



Atmospheric Pollution & Climate Change (APCC)

Environmental Information System Centre (ENVIS)

(Sponsored by Ministry of Environment, Forest & Climate Change, Govt. of India)

ABSTRACTS

India & Global



2017-18

**INDIAN INSTITUTE OF TROPICAL METEOROLOGY
PUNE - 411 008**

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Environmental Information System's (ENVIS) centre on Atmospheric Pollution & Climate Change (APCC) at Indian Institute of Tropical Meteorology (IITM, Pune) is compiling the abstracts of new research done in the field air pollution and climate change categories, for the year 2017. This book has those abstracts which would help scientists, environmentalists and conservationists regarding monitoring, & controlling for atmospheric pollution and climate change.

CONTENTS

Sr. No.	Title	Page No.
	Atmospheric Chemistry & Physics Journal	1-165
1	A missing source of aerosols in Antarctica – beyond long-range transport, phytoplankton, and photochemistry	1
2	Air quality improvement in a megacity: implications from 2015 Beijing Parade Blue pollution control actions	2
3	Atmospheric CO ₂ observations and models suggest strong carbon uptake by forests in New Zealand	2
4	Modeling biogenic and anthropogenic secondary organic aerosol in China	3
5	Development of a high-resolution emission inventory and its evaluation and application through air quality modeling for Jiangsu Province, China	4
6	Global inverse modeling of CH ₄ sources and sinks: an overview of methods	5
7	Variations in O ₃ , CO, and CH ₄ over the Bay of Bengal during the summer monsoon season: shipborne measurements and model simulations	5
8	Characteristics of brown carbon in the urban Po Valley atmosphere	6
9	Size-selected black carbon mass distributions and mixing state in polluted and clean environments of northern India	7
10	Impacts of air pollution and climate on materials in Athens, Greece	7
11	Background aerosol over the Himalayas and Tibetan Plateau: observed characteristics of aerosol mass loading	8
12	Nocturnal new particle formation events in urban environments	9
13	Diurnal variability of the atmospheric boundary layer height over a tropical station in the Indian monsoon region	9
14	Stratospheric tropical warming event and its impact on the polar and tropical troposphere	10
15	The dynamical impact of Rossby wave breaking upon UK PM ₁₀ concentration	10
16	Determination of the atmospheric lifetime and global warming potential of sulfur hexafluoride using a three-dimensional model	11
17	MIX: a mosaic Asian anthropogenic emission inventory under the international collaboration framework of the MICS-Asia and HTAP	12
18	Biomass burning aerosols and the low-visibility events in Southeast Asia	12
19	Quantifying local-scale dust emission from the Arabian Red Sea coastal plain	13
20	Factors controlling black carbon distribution in the Arctic	14
21	Chemistry–climate interactions of aerosol nitrate from lightning	15
22	Data assimilation for volcanic ash plumes using a satellite observational operator: a case study on the 2010 Eyjafjallajökull volcanic eruption	15
23	Direct observations of organic aerosols in common wintertime hazes in North China: insights into direct emissions from Chinese residential stoves	16
24	Influence of enhanced Asian NO _x emissions on ozone in the upper troposphere and lower stratosphere in chemistry–climate model simulations	17
25	Aerosol optical depth thresholds as a tool to assess diffuse radiation fertilization of the land carbon uptake in China	17
26	Qualitative and quantitative analysis of atmospheric organosulfates in Centreville, Alabama	18

27	Simulating ozone dry deposition at a boreal forest with a multi-layer canopy deposition model	19
28	Potential sources and processes affecting speciated atmospheric mercury at Kejimikujik National Park, Canada: comparison of receptor models and data treatment methods	20
29	The impact of resolution on meteorological, chemical and aerosol properties in regional simulations with WRF-Chem	20
30	Halogen chemistry reduces tropospheric O ₃ radiative forcing	21
31	Uptake of nitric acid, ammonia, and organics in orographic clouds: mass spectrometric analyses of droplet residual and interstitial aerosol particles	22
32	Understanding severe winter haze events in the North China Plain in 2014: roles of climate anomalies	22
33	Dust deposition and ambient PM ₁₀ concentration in northwest China: spatial and temporal variability	23
34	Anthropogenic influences on the physical state of submicron particulate matter over a tropical forest	24
35	Cleaning up the air: effectiveness of air quality policy for SO ₂ and NO _x emissions in China	24
36	Particulate-phase mercury emissions from biomass burning and impact on resulting deposition: a modelling assessment	25
37	The CAMS interim Reanalysis of Carbon Monoxide, Ozone and Aerosol for 2003–2015	26
38	Contributions of trans-boundary transport to summertime air quality in Beijing, China	27
39	60 years of UK visibility measurements: impact of meteorology and atmospheric pollutants on visibility	27
40	Nitrate radicals and biogenic volatile organic compounds: oxidation, mechanisms, and organic aerosol	28
41	Impact of a moderate volcanic eruption on chemistry in the lower stratosphere: balloon-borne observations and model calculations	29
42	The recent increase of atmospheric methane from 10 years of ground-based NDACC FTIR observations since 2005	30
43	Emission, transport, and radiative effects of mineral dust from the Taklimakan and Gobi deserts: comparison of measurements and model results	31
44	Influence of rain on the abundance of bioaerosols in fine and coarse particles	31
45	Regional influence of wildfires on aerosol chemistry in the western US and insights into atmospheric aging of biomass burning organic aerosol	32
46	Widespread and persistent ozone pollution in eastern China during the non-winter season of 2015: observations and source attributions	33
47	Global emissions of fluorinated greenhouse gases 2005–2050 with abatement potentials and costs	34
48	A comprehensive biomass burning emission inventory with high spatial and temporal resolution in China	34
49	Improving volcanic ash predictions with the HYSPLIT dispersion model by assimilating MODIS satellite retrievals	35
50	Two global data sets of daily fire emission injection heights since 2003	36
51	US surface ozone trends and extremes from 1980 to 2014: quantifying the	37

	roles of rising Asian emissions, domestic controls, wildfires, and climate	
52	Attributions of meteorological and emission factors to the 2015 winter severe haze pollution episodes in China's Jing-Jin-Ji area	38
53	Effects of ozone-vegetation coupling on surface ozone air quality via biogeochemical and meteorological feedbacks	38
54	Enhanced trans-Himalaya pollution transport to the Tibetan Plateau by cut-off low systems	39
55	Classification of summertime synoptic patterns in Beijing and their associations with boundary layer structure affecting aerosol pollution	40
56	The contribution of wood burning and other pollution sources to wintertime organic aerosol levels in two Greek cities	41
57	Resolving anthropogenic aerosol pollution types – deconvolution and exploratory classification of pollution events	41
58	Sources and formation mechanisms of carbonaceous aerosol at a regional background site in the Netherlands: insights from a year-long radiocarbon study	42
59	Causes of interannual variability over the southern hemispheric tropospheric ozone maximum	43
60	Terpenoid and carbonyl emissions from Norway spruce in Finland during the growing season	44
61	Interpreting the $^{13}\text{C}/^{12}\text{C}$ ratio of carbon dioxide in an urban airshed in the Yangtze River Delta, China	44
62	Bromine atom production and chain propagation during springtime Arctic ozone depletion events in Barrow, Alaska	45
63	Evaluation of the impact of wood combustion on benzo[a]pyrene (BaP) concentrations; ambient measurements and dispersion modeling in Helsinki, Finland	46
64	Black carbon variability since preindustrial times in the eastern part of Europe reconstructed from Mt. Elbrus, Caucasus, ice cores	46
65	Methane fluxes in the high northern latitudes for 2005–2013 estimated using a Bayesian atmospheric inversion	47
66	Composition, size and cloud condensation nuclei activity of biomass burning aerosol from northern Australian savannah fires	48
67	Seasonal variations of triple oxygen isotopic compositions of atmospheric sulfate, nitrate, and ozone at Dumont d'Urville, coastal Antarctica	49
68	Interannual variation, decadal trend, and future change in ozone outflow from East Asia	49
69	Sensitivity of transatlantic dust transport to chemical aging and related atmospheric processes	50
70	Nitrate transboundary heavy pollution over East Asia in winter	51
71	Upper tropospheric cloud systems derived from IR sounders: properties of cirrus anvils in the tropics	52
72	Global distribution of CO_2 in the upper troposphere and stratosphere	53
73	Lower tropospheric distributions of O_3 and aerosol over Raoyang, a rural site in the North China Plain	53
74	An investigation on the origin of regional springtime ozone episodes in the western Mediterranean	54
75	Biomass burning and biogenic aerosols in northern Australia during the	55

	SAFIRE campaign	
76	Constraining sector-specific CO ₂ and CH ₄ emissions in the US	55
77	Satellite retrievals of dust aerosol over the Red Sea and the Persian Gulf (2005–2015)	56
78	Volcanic ash modeling with the online NMMB-MONARCH-ASH v1.0 model: model description, case simulation, and evaluation	57
79	Remote sensing and modelling analysis of the extreme dust storm hitting the Middle East and eastern Mediterranean in September 2015	57
80	Evidence for renoxification in the tropical marine boundary layer	58
81	Wave modulation of the extratropical tropopause inversion layer	58
82	Day and night-time formation of organic nitrates at a forested mountain site in south-west Germany	59
83	Field observations of volatile organic compound (VOC) exchange in red oaks	60
84	Microphysical sensitivity of coupled springtime Arctic stratocumulus to modelled primary ice over the ice pack, marginal ice, and ocean	60
85	Evaluation of the absorption Ångström exponents for traffic and wood burning in the Aethalometer-based source apportionment using radiocarbon measurements of ambient aerosol	61
86	Atmospheric trace metals measured at a regional background site (Welgegund) in South Africa	62
87	Carbonaceous aerosol source apportionment using the Aethalometer model – evaluation by radiocarbon and levoglucosan analysis at a rural background site in southern Sweden	63
88	Chemical transport model simulations of organic aerosol in southern California: model evaluation and gasoline and diesel source contributions	64
89	Source attribution of black carbon and its direct radiative forcing in China	65
90	Influence of 2000–2050 climate change on particulate matter in the United States: results from a new statistical model	65
91	Estimates of the organic aerosol volatility in a boreal forest using two independent methods	66
92	Impacts of coal burning on ambient PM _{2.5} pollution in China	67
93	Probing into the aging dynamics of biomass burning aerosol by using satellite measurements of aerosol optical depth and carbon monoxide	67
94	Constraining N ₂ O emissions since 1940 using firn air isotope measurements in both hemispheres	68
95	A 15-year record of CO emissions constrained by MOPITT CO observations	69
96	Changing trends and emissions of hydrochlorofluorocarbons (HCFCs) and their hydrofluorocarbon (HFCs) replacements	70
97	Impact of typhoons on the composition of the upper troposphere within the Asian summer monsoon anticyclone: the SWOP campaign in Lhasa 2013	71
98	OMI air-quality monitoring over the Middle East	71
99	Long-term air concentrations, wet deposition, and scavenging ratios of inorganic ions, HNO ₃ , and SO ₂ and assessment of aerosol and precipitation acidity at Canadian rural locations	72
100	Emission factors and light absorption properties of brown carbon from household coal combustion in China	73
101	Consistent regional fluxes of CH ₄ and CO ₂ inferred from GOSAT proxy XCH ₄ : XCO ₂ retrievals, 2010–2014	73

102	Impacts of East Asian summer and winter monsoons on interannual variations of mass concentrations and direct radiative forcing of black carbon over eastern China	74
103	Improving PM _{2.5} forecast over China by the joint adjustment of initial conditions and source emissions with an ensemble Kalman filter	75
104	Tropospheric ozone maxima observed over the Arabian Sea during the pre-monsoon	76
105	Contributions of surface solar radiation and precipitation to the spatiotemporal patterns of surface and air warming in China from 1960 to 2003	76
106	Emissions of volatile organic compounds (VOCs) from concentrated animal feeding operations (CAFOs): chemical compositions and separation of sources	77
107	Multi-pollutant emissions from the burning of major agricultural residues in China and the related health-economic effects	78
108	Impact of temperature dependence on the possible contribution of organics to new particle formation in the atmosphere	79
109	Validation of OMI, GOME-2A and GOME-2B tropospheric NO ₂ , SO ₂ and HCHO products using MAX-DOAS observations from 2011 to 2014 in Wuxi, China: investigation of the effects of priori profiles and aerosols on the satellite products	79
110	Relative importance of black carbon, brown carbon, and absorption enhancement from clear coatings in biomass burning emissions	80
111	Real-time aerosol optical properties, morphology and mixing states under clear, haze and fog episodes in the summer of urban Beijing	81
112	Biophysical effects on the interannual variation in carbon dioxide exchange of an alpine meadow on the Tibetan Plateau	82
113	The Climatology of Australian Aerosol	82
114	Investigation of the aerosol–cloud–rainfall association over the Indian summer monsoon region	83
115	Direct radiative effect of carbonaceous aerosols from crop residue burning during the summer harvest season in East China	84
116	Genesis of diamond dust, ice fog and thick cloud episodes observed and modelled above Dome C, Antarctica	85
117	Urbanization-induced urban heat island and aerosol effects on climate extremes in the Yangtze River Delta region of China	86
118	Secondary organic aerosol formation in biomass-burning plumes: theoretical analysis of lab studies and ambient plumes	86
119	How can mountaintop CO ₂ observations be used to constrain regional carbon fluxes?	87
120	Source-sector contributions to European ozone and fine PM in 2010 using AQMEII modeling data	88
121	The constraint of CO ₂ measurements made onboard passenger aircraft on surface–atmosphere fluxes: the impact of transport model errors in vertical mixing	89
122	The radiative role of ozone and water vapour in the annual temperature cycle in the tropical tropopause layer	89
123	Impact of intercontinental pollution transport on North American ozone air	90

	pollution: an HTAP phase 2 multi-model study	
124	Satellite-derived methane hotspot emission estimates using a fast data-driven method	91
125	Observations of atmospheric chemical deposition to high Arctic snow	92
126	Clouds over the summertime Sahara: an evaluation of Met Office retrievals from Meteosat Second Generation using airborne remote sensing	93
127	Comparisons of ground-based tropospheric NO ₂ MAX-DOAS measurements to satellite observations with the aid of an air quality model over the Thessaloniki area, Greece	93
128	Alteration of the size distributions and mixing states of black carbon through transport in the boundary layer in east Asia	94
129	Sensitivity of black carbon concentrations and climate impact to aging and scavenging in OsloCTM2-M7	95
130	Downward particle fluxes of biogenic matter and Saharan dust across the equatorial North Atlantic	96
131	Ozone and haze pollution weakens net primary productivity in China	96
132	Trace gas composition in the Asian summer monsoon anticyclone: a case study based on aircraft observations and model simulations	97
133	New particle formation in the Svalbard region 2006–2015	98
134	Spatial, temporal and source contribution assessments of black carbon over the northern interior of South Africa	99
135	Temporal and spatial variability of ammonia in urban and agricultural regions of northern Colorado, United States	100
136	Technical note: Boundary layer height determination from lidar for improving air pollution episode modeling: development of new algorithm and evaluation	101
137	Impact of Saharan dust on North Atlantic marine stratocumulus clouds: importance of the semidirect effect	101
138	Heterogeneous uptake of ammonia and dimethylamine into sulfuric and oxalic acid particles	102
139	Comparison of emissions inventories of anthropogenic air pollutants and greenhouse gases in China	103
140	Particulate emissions from large North American wildfires estimated using a new top-down method	104
141	Surface ozone in the Southern Hemisphere: 20 years of data from a site with a unique setting in El Tololo, Chile	104
142	Efficient bulk mass accommodation and dissociation of N ₂ O ₅ in neutral aqueous aerosol	105
143	Near-road sampling of PM _{2.5} , BC, and fine-particle chemical components in Kathmandu Valley, Nepal	106
144	Summer ozone in the northern Front Range metropolitan area: weekend–weekday effects, temperature dependences, and the impact of drought	106
145	Glacier evolution in high-mountain Asia under stratospheric sulfate aerosol injection geoengineering	107
146	Regional background O ₃ and NO _x in the Houston–Galveston–Brazoria (TX) region: a decadal-scale perspective	107
147	Influence of urban pollution on the production of organic particulate matter from isoprene epoxydiols in central Amazonia	108

148	The relationship between lower-stratospheric ozone at southern high latitudes and sea surface temperature in the East Asian marginal seas in austral spring	109
149	Secondary ozone peaks in the troposphere over the Himalayas	110
150	Measurement of PM and its chemical composition in real-world emissions from non-road and on-road diesel vehicles	110
151	Influence of biomass burning from South Asia at a high-altitude mountain receptor site in China	111
152	Radiative and climate effects of stratospheric sulfur geoengineering using seasonally varying injection areas	112
153	Quantifying pollution transport from the Asian monsoon anticyclone into the lower stratosphere	112
154	Characterization of fresh and aged organic aerosol emissions from meat charbroiling	113
155	Formation of secondary organic aerosols from gas-phase emissions of heated cooking oils	114
156	Diagnostic methods for atmospheric inversions of long-lived greenhouse gases	114
157	Aerosol effects on the development of cumulus clouds over the Tibetan Plateau	115
158	Methane emissions from dairies in the Los Angeles Basin	116
159	Effect of mid-term drought on Quercus pubescens BVOCs' emission seasonality and their dependency on light and/or temperature	116
160	Size-resolved chemical composition, effective density, and optical properties of biomass burning particles	117
161	Methane emissions from dairies in the Los Angeles Basin	118
162	Particle size dependence of biogenic secondary organic aerosol molecular composition	118
163	Meteorological context of the onset and end of the rainy season in Central Amazonia during the GoAmazon2014/5	119
164	Speciation of anthropogenic emissions of non-methane volatile organic compounds: a global gridded data set for 1970–2012	120
165	Enhanced toxicity of aerosol in fog conditions in the Po Valley, Italy	120
166	Improved provincial emission inventory and speciation profiles of anthropogenic non-methane volatile organic compounds: a case study for Jiangsu, China	121
167	Secondary inorganic aerosols in Europe: sources and the significant influence of biogenic VOC emissions, especially on ammonium nitrate	122
168	Measurement of scattering and absorption properties of dust aerosol in a Gobi farmland region of northwestern China – a potential anthropogenic influence	123
169	Air stagnation in China (1985–2014): climatological mean features and trends	124
170	Estimating the size of a methane emission point source at different scales: from local to landscape	124
171	Optical, physical and chemical properties of aerosols transported to a coastal site in the western Mediterranean: a focus on primary marine aerosols	125
172	Aerosol–landscape–cloud interaction: signatures of topography effect on	126

	cloud droplet formation	
173	Size distribution and source of black carbon aerosol in urban Beijing during winter haze episodes	126
174	Contributions of mobile, stationary and biogenic sources to air pollution in the Amazon rainforest: a numerical study with the WRF-Chem model	127
175	Contribution of different processes to changes in tropical lower-stratospheric water vapor in chemistry–climate models	128
176	Intercomparison of meteorological analyses and trajectories in the Antarctic lower stratosphere with Concordiasi superpressure balloon observations	128
177	Effect of volcanic aerosol on stratospheric NO ₂ and N ₂ O ₅ from 2002–2014 as measured by Odin-OSIRIS and Envisat-MIPAS	129
178	Source apportionment of NMVOCs in the Kathmandu Valley during the SusKat-ABC international field campaign using positive matrix factorization	130
179	Investigation of the mixing layer height derived from ceilometer measurements in the Kathmandu Valley and implications for local air quality	130
180	The boundary condition for vertical velocity and its interdependence with surface gas exchange	131
181	Observation- and model-based estimates of particulate dry nitrogen deposition to the oceans	132
182	Estimating daily surface NO ₂ concentrations from satellite data – a case study over Hong Kong using land use regression models	133
183	Long-term change in the source contribution to surface ozone over Japan	134
184	Changes to the chemical state of the Northern Hemisphere atmosphere during the second half of the twentieth century	134
185	Evaluation of ACCMIP ozone simulations and ozonesonde sampling biases using a satellite-based multi-constituent chemical reanalysis	135
186	Carbon dioxide and methane measurements from the Los Angeles Megacity Carbon Project – Part 1: calibration, urban enhancements, and uncertainty estimates	136
187	Understanding the primary emissions and secondary formation of gaseous organic acids in the oil sands region of Alberta, Canada	137
188	Global O ₃ –CO correlations in a chemistry and transport model during July–August: evaluation with TES satellite observations and sensitivity to input meteorological data and emissions	138
189	Uncertainties of fluxes and ¹³ C / ¹² C ratios of atmospheric reactive-gas emissions	139
190	Lidar ratios of stratospheric volcanic ash and sulfate aerosols retrieved from CALIOP measurements	139
191	Arctic regional methane fluxes by ecotope as derived using eddy covariance from a low-flying aircraft	140
192	Global anthropogenic emissions of particulate matter including black carbon	140
193	Comparison of primary and secondary particle formation from natural gas engine exhaust and of their volatility characteristics	141
194	Response of the global surface ozone distribution to Northern Hemisphere sea surface temperature changes: implications for long-range transport	142
195	Atmospheric chemistry, sources and sinks of carbon suboxide, C ₃ O ₂	143
196	Observations of particles at their formation sizes in Beijing, China	144
197	Organic molecular tracers in the atmospheric aerosols from Lumbini, Nepal,	144

	in the northern Indo-Gangetic Plain: influence of biomass burning	
198	Modeling the role of highly oxidized multifunctional organic molecules for the growth of new particles over the boreal forest region	145
199	Global source attribution of sulfate concentration and direct and indirect radiative forcing	146
200	Evaporation of sulfate aerosols at low relative humidity	146
201	Speciation of organic aerosols in the Saharan Air Layer and in the free troposphere westerlies	147
202	Investigating diesel engines as an atmospheric source of isocyanic acid in urban areas	148
203	Power plant fuel switching and air quality in a tropical, forested environment	148
204	Subtropical subsidence and surface deposition of oxidized mercury produced in the free troposphere	149
205	A wedge strategy for mitigation of urban warming in future climate scenarios	150
206	Global atmospheric chemistry – which air matters	150
207	Differentiating local and regional sources of Chinese urban air pollution based on the effect of the Spring Festival	151
208	OMI-measured increasing SO ₂ emissions due to energy industry expansion and relocation in northwestern China	152
209	Observed trends in ground-level O ₃ in Monterrey, Mexico, during 1993–2014: comparison with Mexico City and Guadalajara	152
210	Chemical composition of ambient PM _{2.5} over China and relationship to precursor emissions during 2005–2012	153
211	Uncertainty assessment and applicability of an inversion method for volcanic ash forecasting	154
212	Wildfire air pollution hazard during the 21st century	155
213	NO _x emission trends over Chinese cities estimated from OMI observations during 2005 to 2015	155
214	An assessment of ozone mini-hole representation in reanalyses over the Northern Hemisphere	156
215	On the multiday haze in the Asian continental outflow: the important role of synoptic conditions combined with regional and local sources	156
216	Net ecosystem exchange and energy fluxes measured with the eddy covariance technique in a western Siberian bog	157
217	Trends and annual cycles in soundings of Arctic tropospheric ozone	158
218	Temporal characteristics of atmospheric ammonia and nitrogen dioxide over China based on emission data, satellite observations and atmospheric transport modeling since 1980	158
219	Plume-exit modeling to determine cloud condensation nuclei activity of aerosols from residential biofuel combustion	159
220	Sources of particulate matter components in the Athabasca oil sands region: investigation through a comparison of trace element measurement methodologies	160
221	A review of current knowledge concerning PM _{2.5} chemical composition, aerosol optical properties and their relationships across China	161
222	Mixing state of oxalic acid containing particles in the rural area of Pearl River Delta, China: implications for the formation mechanism of oxalic acid	162
223	Volatile organic compounds (VOCs) in photochemically aged air from the	162

	eastern and western Mediterranean	
224	Spatial extent of new particle formation events over the Mediterranean Basin from multiple ground-based and airborne measurements	163
225	Possible climatic implications of high-altitude black carbon emissions	164
226	Sources of springtime surface black carbon in the Arctic: an adjoint analysis for April 2008	165
	Aerosol And Air Quality Research	166-236
227	Analysing Temporal Trends in the Ratios of PM _{2.5} /PM ₁₀ in the UK	166
228	Single-Particle Characterizations of Ambient Aerosols during a Wintertime Pollution Episode in Nanning: Local Emissions vs. Regional Transport	166
229	Effects of Polluted Air-Masses Advection on Atmospheric Particles in a Semi-Rural Site in South Italy by SEM-EDX Analysis	167
230	The Concentrations, Formations, Relationships and Modeling of Sulfate, Nitrate and Ammonium (SNA) Aerosols over China	167
231	A Typical Formation Mechanism of Heavy Haze-Fog Induced by Coal Combustion in an Inland City in North-Western China	168
232	Characterization of Traffic-Related Particulate Matter Emissions in a Road Tunnel in Birmingham, UK: Trace Metals and Organic Molecular Markers	168
233	Number Concentrations and Modal Structure of Indoor/Outdoor Fine Particles in Four European Cities	169
234	Changes in Gas-Phase Air Pollutants across New York State, USA	170
235	Investigating Criteria and Organic Air pollutant Emissions from Motorcycles by Using Various Ethanol-Gasoline Blends	170
236	PAHs in Size Fractionate Mainstream Cigarette Smoke, Predictive Deposition and Associated Inhalation Risk	171
237	A Survey of VOC Emissions from Rendering Plants	172
238	An Investigation into the Effects of Off-Shore Shipping Emissions on Coastal Black Carbon Concentrations	172
239	Association of Particulate Matter Impact on Prevalence of Chronic Obstructive Pulmonary Disease in Ahvaz, Southwest Iran during 2009–2013	173
240	Daily Personal Exposure of Women Cooks to Respirable Particulate Matters during Cooking with Solid Bio-Fuels in a Rural Community of West Bengal, India	174
241	Evaluation of PM _{2.5} Surface Concentrations Simulated by Version 1 of NASA's MERRA Aerosol Reanalysis over Israel and Taiwan	174
242	Performance Evaluation of the WRF-Chem Model with Different Physical Parameterization Schemes during an Extremely High PM _{2.5} Pollution Episode in Beijing	175
243	Using Box Modeling to Determine Photodegradation Coefficients Describing the Removal of Gaseous Formaldehyde from Indoor Air	176
244	Development and Application of a New PM _{2.5} Source Apportionment Approach	176
245	Overview of the Special Issue "PM _{2.5} in Asia" for 2015 Asian Aerosol Conference	177
246	Chemical Characteristics of PM _{2.5} during a 2016 Winter Haze Episode in Shijiazhuang, China	177
247	Atmospheric Dispersion of PM _{2.5} Precursor Gases from Two Major Thermal	178

	Power Plants in Andhra Pradesh, India	
248	Extreme Events of Reactive Ambient Air Pollutants and their Distribution Pattern at Urban Hotspots	179
249	Chemical Characteristics of Particulate Matter Emission from a Heavy-Duty Diesel Engine Using ETC Cycle Dynamometer Test	179
250	Trends of PM _{2.5} and Chemical Composition in Beijing, 2000–2015	180
251	Development of an Automated System (PPWD/PILS) for Studying PM _{2.5} Water-Soluble Ions and Precursor Gases: Field Measurements in Two Cities, Taiwan	181
252	Continuous Observation of the Mass and Chemical Composition of PM _{2.5} using an Automatic Analyzer in Kumamoto, Japan	181
253	Analysing PM _{2.5} and its Association with PM ₁₀ and Meteorology in the Arid Climate of Makkah, Saudi Arabia	182
254	Trajectory-Based Models and Remote Sensing for Biomass Burning Assessment in Bangladesh	183
255	Identification of Sources of Fine Particulate Matter in Kandy, Sri Lanka	183
256	Low Molecular Weight Monocarboxylic Acids in PM _{2.5} and PM ₁₀ : Quantification, Seasonal Variation and Source Apportionment	184
257	Characteristics of PM _{2.5} and Assessing Effects of Emission–Reduction Measures in the Heavy Polluted City of Shijiazhuang, before, during, and after the Ceremonial Parade 2015	185
258	A Study of Characteristics and Origins of Haze Pollution in Zhengzhou, China, Based on Observations and Hybrid Receptor Models	185
259	Chemical Fingerprint and Source Identification of Atmospheric Fine Particles Sampled at Three Environments at the Tip of Southern Taiwan	186
260	Impact of Air Humidity Fluctuation on the Rise of PM Mass Concentration Based on the High-Resolution Monitoring Data	187
261	Size Specific Distribution Analysis of Perfluoroalkyl Substances in Atmospheric Particulate Matter – Development of a Sampling Method and their Concentration in Meeting Room/Ambient Atmosphere	187
262	Characteristics, Sources, and Health Risks of Atmospheric PM _{2.5} -Bound Polycyclic Aromatic Hydrocarbons in Hsinchu, Taiwan	188
263	Comparison of Oxidative Abilities of PM _{2.5} Collected at Traffic and Residential Sites in Japan. Contribution of Transition Metals and Primary and Secondary Aerosols	189
264	Significance of PM _{2.5} Air Quality at the Indian Capital	189
265	Infiltration of Ambient PM _{2.5} through Building Envelope in Apartment Housing Units in Korea	190
266	Characteristics of Exposure to Particles due to Incense Burning inside Temples in Kanpur, India	190
267	Source Apportionment of PM _{2.5} Particles: Influence of Outdoor Particles on Indoor Environment of Schools Using Chemical Mass Balance	191
268	PM _{2.5} Emission Reduction by Technical Improvement in a Typical Coal-Fired Power Plant in China	192
269	Transformations of Aerosol Particles from an Outdoor to Indoor Environment	192
270	Relationships between Outdoor and Personal Exposure of Carbonaceous Species and Polycyclic Aromatic Hydrocarbons (PAHs) in Fine Particulate	193

	Matter (PM2.5) at Hong Kong	
271	Evaluating the Effects of Springtime Dust Storms over Beijing and the Associated Characteristics of Sub-Micron Aerosol	193
272	The Impacts of Emission Control and Regional Transport on PM2.5 Ions and Carbon Components in Nanjing during the 2014 Nanjing Youth Olympic Games	194
273	Spatial and Temporal Variability of the PM2.5/PM10 Ratio in Wuhan, Central China	195
274	Propensity to Migrate and Willingness to Pay Related to Air Pollution among Different Populations in Wuhan, China	195
275	Cross-Sectional View of Atmospheric Aerosols over an Urban Location in Central India	196
276	Airborne Bacterial Communities of PM2.5 in Beijing-Tianjin-Hebei Megalopolis, China as Revealed By Illumina MiSeq Sequencing: A Case Study	197
277	Time-Dependent Size-Resolved Bacterial and Fungal Aerosols in Beijing Subway	197
278	Influence of Surgical Smoke on Indoor Air Quality in Hospital Operating Rooms	198
279	Assessment of City Level Human Health Impact and Corresponding Monetary Cost Burden Due to Air Pollution in India taking Agra as a Model City	199
280	Measured Indoor Aerosol Concentration Arising from Commonly-Used Food and Medicinal Powders: A Pilot Study	199
281	Quantification of Non-Linear Dynamics and Chaos of Ambient Particulate Matter Concentrations in Muzaffarabad City	200
282	Particulate Air Pollution at Schools: Indoor-Outdoor Relationship and Determinants of Indoor Concentrations	201
283	Characterization of Inorganic Elements within PM2.5 and PM10 Fractions of Fly Ashes from Coal-Fired Power Plants	201
284	Impact of Magnetic Tube on Pollutant Emissions from the Diesel Engine	202
285	Decomposition Analysis of CO2 Emissions from Coal - Sourced Electricity Production in South Africa	202
286	Effects of Injection Pressure on Geological CO2 Storage in the Northwest Taiwan Basin	203
287	Modeling the Airflow and Particle Dispersion in Street Canyons under Unsteady Thermal Environment with Sinusoidal Variation	204
288	Arsenic Exposure during Preventive Maintenance of an Ion Implanter in a Semiconductor Manufacturing Factory	204
289	A Case Study of Long-Range Transport of Smoke Aerosols from Eastern Siberia to Northeast China in July 2014	205
290	Spatial Variation of Ground Level Ozone Concentrations and its Health Impacts in an Urban Area in India	205
291	Characterization of Ambient Volatile Organic Compounds (VOCs) in the Area Adjacent to a Petroleum Refinery in Jinan, China	206
292	Inorganic Chemical Composition of Fine Particulates in Medium-Sized Urban Areas: A Case Study of Brazilian Cities	207
293	Chemical Characteristics of Aerosols in Coastal and Urban Ambient Atmospheres	207
294	Characterization of Chemical Composition in PM2.5 in Beijing before, during,	208

	and after a Large-Scale International Event	
295	Pollution Characteristics of PM _{2.5} Aerosol during Haze Periods in Changchun, China	209
296	Two-Step Flotation Treatment for Removal of Toxic Matter from Hospital Solid Waste Incinerator Fly Ash	209
297	Radiative Forcing Estimation of Aerosols at an Urban Site near the Thar Desert Using Ground-Based Remote Sensing Measurements	210
298	The Association between Intermodal (PM _{1-2.5}) and PM ₁ , PM _{2.5} , Coarse Fraction and Meteorological Parameters in Various Environments in Central Europe	210
299	Criteria Pollutants and Volatile Organic Compounds Emitted from Motorcycle Exhaust under Various Regulation Phases	211
300	Assessment of Air Pollution around the Panzhihua V-Ti Magnetite Mine Region, Southwest China	212
301	Study of Local and Regional Influence on PM _{2.5} Concentration during Odd-Even Rule in Delhi Using Causal Analysis	212
302	Investigation of Particulate Matter Regional Transport in Beijing Based on Numerical Simulation	213
303	Chemical Characteristics of PM _{2.5} during 2015 Spring Festival in Beijing, China	213
304	Mass Concentration and Size-Distribution of Atmospheric Particulate Matter in an Urban Environment	214
305	Emissions of Polycyclic Aromatic Hydrocarbons and Particle-Bound Metals from a Diesel Engine Generator Fueled with Waste Cooking Oil-Based Biodiesel Blends	215
306	Multiplier Effects of Energy Consumption and CO ₂ Emissions by Input-Output Analysis in South Africa	216
307	Economic Growth, Carbon Abatement Technology and Decoupling Strategy – The Case of Taiwan	216
308	Influences of Copper(II) Chloride Impregnation on Activated Carbon for Low-Concentration Elemental Mercury Adsorption from Simulated Coal Combustion Flue Gas	217
309	Analysis of Long-Range Transport Effects on PM _{2.5} during a Short Severe Haze in Beijing, China	217
310	The Effects of Height on the Accumulation of n-Alkanes and Polycyclic Aromatic Hydrocarbons (PAHs) in Air-Conditioning Filter Dust from High-Rise Apartments	218
311	Identification and Characterization of Size-Segregated Bioaerosols at Different Sites in Delhi	219
312	Part I: PM _{2.5} and Polychlorinated Dibenzo-p-dioxins and Dibenzofurans (PCDD/Fs) in the Ambient Air of Southern China	219
313	Optical Properties, Chemical Composition and the Toxicological Potential of Urban Particulate Matter	220
314	Application of WRF Model for Air Quality Modelling and AERMOD – A Survey	221
315	Evaluation of Model Performance for Forecasting Fine Particle Concentrations in Korea	221
316	Emission Characteristics of Gas-Fired Boilers in Beijing City, China: Category-Specific Emission Factor, Emission Inventory, and Spatial Characteristics	222

317	PM2.5-Bound Polycyclic Aromatic Hydrocarbons (PAHs), Oxygenated-PAHs and Phthalate Esters (PAEs) inside and outside Middle School Classrooms in Xi'an, China: Concentration, Characteristics and Health Risk Assessment	223
318	Atmospheric Deposition of Polychlorinated Dibenzo-p-dioxins and Dibenzofurans in Two Cities of Southern China	223
319	Pollution Characteristics Revealed by Size Distribution Properties of Aerosol Particles at Urban and Suburban Sites, Northwest China	224
320	A Study on Distance Transport of PM2.5 to Xianlin in Nanjing, China and its Source Areas	225
321	Ozone and Secondary Organic Aerosol Formation of Toluene/NOxIrradiations under Complex Pollution Scenarios	225
322	Impact of Atmospheric Flow Conditions on Fine Aerosols in Sydney, Australia	226
323	Organic Nanoaerosol in Coal Mines: Formation Mechanism and Explosibility	227
324	Chemical Composition of Diesel/Biodiesel Particulate Exhaust by FTIR Spectroscopy and Mass Spectrometry: Impact of Fuel and Driving Cycle	227
325	The Construction and Application of a Multipoint Sampling System for Vehicle Exhaust Plumes	228
326	Ozone Catalytic Oxidation of Gaseous Toluene over MnO ₂ -Based Ozone Decomposition Catalysts Immobilized on a Nonwoven Fabric	229
327	Particle Size Distribution of Soot from a Laminar/Diffusion Flame	229
328	Evaluation of $\delta^{13}\text{C}$ in Carbonaceous Aerosol Source Apportionment at a Rural Measurement Site	230
329	Exposure Assessment of Hexavalent Chromium for a Powder Coating Spray Painter Associated with the Development of Lung Cancer	230
330	Fungal Bioaerosol Exposure and Its Effects on the Health of Mushroom and Vegetable Farm Workers in Taiwan	231
331	Ambient Fungal Spore Concentration in a Subtropical Metropolis: Temporal Distributions and Meteorological Determinants	232
332	Persistent Organic Pollutant Reductions from a Diesel Engine Generator Fueled with Waste Cooking Oil-based Biodiesel Blended with Butanol and Acetone	232
333	Part II: PM2.5 and Polychlorinated Dibenzo-p-dioxins and Dibenzofurans (PCDD/Fs) in the Ambient Air of Northern China	233
334	Mobile Monitoring of Personal NO _x Exposures during Scripted Daily Activities in Chicago, IL	234
335	Pollution Characteristics and Source Apportionment of PM2.5-Bound n-Alkanes in the Yangtze River Delta, China	235
336	Particulate Size Distribution and Sources Evaluation of n-Alkanes during Long-Term Haze Episode around Chaohu Lake, Eastern	235
337	Coarse-Particle Passive-Sampler Measurements and Single-Particle Analysis by Transmitted Light Microscopy at Highly Frequented Motorways	236
	Atmospheric Research	237-259
338	Comparison of atmospheric PM2.5-bounded mercury species and their correlation with bromine and iodine at coastal urban and island sites in the eastern China	237
339	Carbonaceous particles and aerosol mass closure in PM2.5 collected in a port city	237

340	Seasonal variation of near surface black carbon and satellite derived vertical distribution of aerosols over a semi-arid station in India	238
341	Water soluble inorganic species of PM10 and PM2.5 at an urban site of Delhi, India: Seasonal variability and sources	238
342	Smog chamber study on aging of combustion soot in isoprene/SO2/NOx system: Changes of mass, size, effective density, morphology and mixing state	239
343	Aerosol black carbon quantification in the central Indo-Gangetic Plain: Seasonal heterogeneity and source apportionment	240
344	Urban air quality in a mid-size city — PM2.5 composition, sources and identification of impact areas: From local to long range contributions	241
345	Source contributions to PM2.5 in Guangdong province, China by numerical modeling: Results and implications	241
346	Model analysis of soil dust impacts on the boundary layer meteorology and air quality over East Asia in April 2015	242
347	Characteristics of lightning activity in tropical cyclones developed over North Indian Ocean basin during 2010–2015	243
348	A study on aerosol-cloud condensation nuclei (CCN) activation over eastern Himalaya in India	243
349	Source apportionment of ambient PM10 and PM2.5 in Haikou, China	244
350	Source apportionment of PM2.5 size distribution and composition data from multiple stationary sites using a mobile platform	245
351	Characterization of size-resolved urban haze particles collected in summer and winter at Taiyuan City, China using quantitative electron probe X-ray microanalysis	245
352	Carbonaceous species in PM2.5 and PM10 in urban area of Zhengzhou in China: Seasonal variations and source apportionment	246
353	Modeling approach for emissions reduction of primary PM2.5 and secondary PM2.5 precursors to achieve the air quality target	247
354	Effect of nitrogen oxides (NO and NO2) and toluene on SO2 photooxidation, nucleation and growth: A smog chamber study	247
355	Estimating Particulate Matter using satellite based aerosol optical depth and meteorological variables in Malaysia	248
356	Sub-micron particle number size distribution characteristics at two urban locations in Leicester	249
357	Estimate of main local sources to ambient ultrafine particle number concentrations in an urban area	250
358	Estimates of spatially and temporally resolved constrained black carbon emission over the Indian region using a strategic integrated modelling approach	250
359	Association of atmospheric pollution and instability indices: A detailed investigation over an Indian urban metropolis	251
360	Characterization, sources and health risk analysis of PM2.5 bound metals during foggy and non-foggy days in sub-urban atmosphere of Agra	252
361	Determination of trace metals in TSP and PM2.5 materials collected in the Metropolitan Area of Monterrey, Mexico: A characterization study by XPS, ICP-AES and SEM-EDS	252
362	Association of atmospheric pollution and instability indices: A detailed investigation over an Indian urban metropolis	253

363	Characterization, sources and health risk analysis of PM _{2.5} bound metals during foggy and non-foggy days in sub-urban atmosphere of Agra	254
364	High ozone episodes at a semi-urban site in India: Photochemical generation and transport	254
365	Influence of air mass origins on optical properties and PM concentrations measured at a high mountain station located in the southwestern Mediterranean	255
366	Source region and sector contributions of atmospheric soot particle in a coalfield region of Dhanbad, eastern part of India	256
367	The association of weather patterns with haze episodes: Recognition by PM _{2.5} oriented circulation classification applied in Xiamen, Southeastern China	257
368	Atmospheric deposition of trace elements at urban and forest sites in central Poland – Insight into seasonal variability and sources	257
369	Chemical characteristics of PM _{2.5} during summer at a background site of the Yangtze River Delta in China	258
370	Temporal and spatial analyses of particulate matter (PM ₁₀ and PM _{2.5}) and its relationship with meteorological parameters over an urban city in northeast China	259
	Atmospheric Environment	260-322
371	Enhanced concentrations of reactive nitrogen species in wildfire smoke	260
372	Solar and thermal radiative effects during the 2011 extreme desert dust episode over Portugal	260
373	Characteristics of aerosol size distribution and vertical backscattering coefficient profile during 2014 APEC in Beijing	261
374	Effect of fiber material on ozone removal and carbonyl production from carpets	262
375	Radiation fog chemical composition and its temporal trend over an eight year period	263
376	Characteristics, sources and evolution of fine aerosol (PM ₁) at urban, coastal and forest background sites in Lithuania	263
377	The impact of Mount Etna sulfur emissions on the atmospheric composition and aerosol properties in the central Mediterranean: A statistical analysis over the period 2000–2013 based on observations and Lagrangian modeling	264
378	Multi-tracer approach to characterize domestic wood burning in Athens (Greece) during wintertime	265
379	Oxidative potential of on-road fine particulate matter (PM _{2.5}) measured on major freeways of Los Angeles, CA, and a 10-year comparison with earlier roadside studies	266
380	Comparison of source apportionment of PM _{2.5} using receptor models in the main hub port city of East Asia: Busan	267
381	Evaluation of fire weather forecasts using PM _{2.5} sensitivity analysis	267
382	Size-segregated particulate matter inside residences of elderly in the Metropolitan Area of São Paulo, Brazil	268
383	Clustered long-range transport routes and potential sources of PM _{2.5} and their chemical characteristics around the Taiwan Strait	269
384	On-road measurements of vehicle NO ₂ /NO _x emission ratios in Denver, Colorado, USA	270

385	Emission characteristics of polycyclic aromatic hydrocarbons and nitro-polycyclic aromatic hydrocarbons from diesel trucks based on on-road measurements	270
386	Analysis of major air pollutants and submicron particles in New York City and Long Island	271
387	Source apportionment of PM _{2.5} chemically speciated mass and particle number concentrations in New York City	272
388	Oxidative potential of subway PM _{2.5}	272
389	Air quality early-warning system for cities in China	273
390	Regionalized PM _{2.5} Community Multiscale Air Quality model performance evaluation across a continuous spatiotemporal domain	274
391	Estimating accidental pollutant releases in the built environment from turbulent concentration signals	274
392	Role of surface wind and vegetation cover in multi-decadal variations of dust emission in the Sahara and Sahel	275
393	Effects of synoptic weather on ground-level PM _{2.5} concentrations in the United States	275
394	Transport of East Asian dust storms to the marginal seas of China and the southern North Pacific in spring 2010	276
395	Determination of n-alkanes, PAHs and nitro-PAHs in PM _{2.5} and PM ₁ sampled in the surroundings of a municipal waste incinerator	277
396	Effect of monitoring network design on land use regression models for estimating residential NO ₂ concentration	278
397	Particulate matter produced during commercial sugarcane harvesting and processing: A respiratory health hazard?	278
398	Multifractal analysis of interactive patterns between meteorological factors and pollutants in urban and rural areas	279
399	Use of tether sonde and aircraft profiles to study the impact of mesoscale and microscale meteorology on air quality	280
400	A comparison study of carbonaceous aerosols in a typical North China Plain urban atmosphere: Seasonal variability, sources and implications to haze formation	280
401	Assessing the effect of long-range pollutant transportation on air quality in Seoul using the conditional potential source contribution function method	281
402	PM _{2.5} and aerosol black carbon in Suva, Fiji	282
403	Effects of future temperature change on PM _{2.5} infiltration in the Greater Boston area	283
404	Short term effects of criteria air pollutants on daily mortality in Delhi, India	283
405	Impact of anomalous forest fire on aerosol radiative forcing and snow cover over Himalayan region	284
406	Effect of tillage and water management on GHG emissions from Mediterranean rice growing ecosystems	285
407	An integrative approach for determination of air pollution and its health effects in a coal fired power plant area by passive sampling	286
408	Chemical characteristics of atmospheric PM _{2.5} loads during air pollution episodes in Giza, Egypt	286
409	Modelling and analysis of ozone concentration by artificial intelligent techniques for estimating air quality	287

410	Emissions of indoor air pollutants from six user scenarios in a model room	288
411	Exposure assessment models for elemental components of particulate matter in an urban environment: A comparison of regression and random forest approaches	289
412	Large reductions in urban black carbon concentrations in the United States between 1965 and 2000	289
413	Trend analysis of CO ₂ and CH ₄ recorded at a semi-natural site in the northern plateau of the Iberian Peninsula	290
414	The effects of marine vessel fuel sulfur regulations on ambient PM _{2.5} at coastal and near coastal monitoring sites in the U.S.	291
415	Characteristics of volatile organic compounds (VOCs) from the evaporative emissions of modern passenger cars	292
416	Climatology of wintertime long-distance transport of surface-layer air masses arriving urban Beijing in 2001–2012	292
417	Emission rates of particle number, mass and black carbon by the Los Angeles International Airport (LAX) and its impact on air quality in Los Angeles	293
418	Lung deposited surface area in Leicester urban background site/UK: Sources and contribution of new particle formation	294
419	Simulating ozone concentrations using precursor emission inventories in Delhi – National Capital Region of India	294
420	Characteristics of an open-cut coal mine fire pollution event	295
421	Atmospheric emissions of typical toxic heavy metals from open burning of municipal solid waste in China	296
422	Combined use of land use regression and BenMAP for estimating public health benefits of reducing PM _{2.5} in Tianjin, China	296
423	Responses of human health and vegetation exposure metrics to changes in ozone concentration distributions in the European Union, United States, and China	297
424	Inhalation exposure and health risk levels to BTEX and carbonyl compounds of traffic policeman working in the inner city of Bangkok, Thailand	298
425	Responses of human health and vegetation exposure metrics to changes in ozone concentration distributions in the European Union, United States, and China	299
426	Comprehensive chemical characterization of industrial PM _{2.5} from steel industry activities	300
427	Ambient air quality measurements from a continuously moving mobile platform: Estimation of area-wide, fuel-based, mobile source emission factors using absolute principal component scores	301
428	Impact of future climate policy scenarios on air quality and aerosol-cloud interactions using an advanced version of CESM/CAM5: Part I. model evaluation for the current decadal simulations	301
429	Relative impact of on-road vehicular and point-source industrial emissions of air pollutants in a medium-sized Andean city	302
430	A modeling study on the effect of urban land surface forcing to regional meteorology and air quality over South China	303
431	A three-year investigation of daily PM _{2.5} main chemical components in four sites: the routine measurement program of the Supersito Project (Po Valley, Italy)	304

432	Heterogeneous reaction of SO ₂ with soot: The roles of relative humidity and surface composition of soot in surface sulfate formation	304
433	Impact of future climate policy scenarios on air quality and aerosol-cloud interactions using an advanced version of CESM/CAM5: Part II. Future trend analysis and impacts of projected anthropogenic emissions	305
434	Large-eddy simulation of dense gas dispersion over a simplified urban area	306
435	Environmental studies in two communes of Santiago de Chile by the analysis of magnetic properties of particulate matter deposited on leaves of roadside trees	307
436	Content, mineral allocation and leaching behavior of heavy metals in urban PM _{2.5}	307
437	Numerical air quality forecasting over eastern China: An operational application of WRF-Chem	308
438	Air pollutants and toxic emissions of various mileage motorcycles for ECE driving cycles	309
439	Uncertainty and dynamics of natural wetland CH ₄ release in China: Research status and priorities	309
440	Variation of the vertical distribution of Nabro volcano aerosol layers in the stratosphere observed by LIDAR	310
441	Assessment of emissions of greenhouse gases and air pollutants in Indonesia and impacts of national policy for elimination of kerosene use in cooking	311
442	Particulate matter air pollution in Europe in a +2 °C warming world	312
443	Variations in particulate matter over Indo-Gangetic Plains and Indo-Himalayan Range during four field campaigns in winter monsoon and summer monsoon: Role of pollution pathways	313
444	Greenhouse gases concentrations and fluxes from subtropical small reservoirs in relation with watershed urbanization	313
445	Emission factors and characteristics of ammonia, hydrogen sulfide, carbon dioxide, and particulate matter at two high-rise layer hen houses	314
446	Reduction of air pollution levels downwind of a road with an upwind noise barrier	315
447	Characterising an intense PM pollution episode in March 2015 in France from multi-site approach and near real time data: Climatology, variabilities, geographical origins and model evaluation	316
448	Composition and mixing states of brown haze particle over the Himalayas along two transboundary south-north transects	316
449	Organic aerosols over Indo-Gangetic Plain: Sources, distributions and climatic implications	317
450	Effectiveness of temporary control measures for lowering PM _{2.5} pollution in Beijing and the implications	318
451	Exposure to fine particulate, black carbon, and particle number concentration in transportation microenvironments	319
452	Daily ambient air pollution metrics for five cities: Evaluation of data-fusion-based estimates and uncertainties	319
453	Role of organic aerosols in CCN activation and closure over a rural background site in Western Ghats, India	320
454	The contribution of residential coal combustion to PM _{2.5} pollution over China's Beijing-Tianjin-Hebei region in winter	321

455	Human health risk due to variations in PM10-PM2.5 and associated PAHs levels	322
456	Atmospheric oxidation of selected chlorinated alkenes by O3, OH, NO3 and Cl	322
	Environmental Science & Pollution Research	324-366
457	Variations of polycyclic aromatic hydrocarbons in ambient air during haze and non-haze episodes in warm seasons in Hangzhou, China	324
458	15N-labeled ammonium nitrogen uptake and physiological responses of poplar exposed to PM2.5 particles	324
459	Chemical characterization of PM1.0 aerosol in Delhi and source apportionment using positive matrix factorization	325
460	Surface properties of PM2.5 calcite fine particulate matter in the presence of same size bacterial cells and exocellular polymeric substances (EPS) of <i>Bacillus mucitagnosus</i>	326
461	Seasonal variations in size distribution, water-soluble ions, and carbon content of size-segregated aerosols over New Delhi	326
462	Modeling the impacts of ambient temperatures on cardiovascular mortality in Yinchuan: evidence from a northwestern city of China	327
463	Impacts of integrated nutrient management on methane emission, global warming potential and carbon storage capacity in rice grown in a northeast India soil	328
464	Acute effects of air pollution on spontaneous abortion, premature delivery, and stillbirth in Ahvaz, Iran: a time-series study	328
465	Elemental composition of PM2.5 in the urban environment of San Juan, Argentina	329
466	Air quality study in the coastal city of Crotone (Southern Italy) hosting a small-size harbor	330
467	Spatial variation of air quality index and urban driving factors linkages: evidence from Chinese cities	330
468	Global warming potential and greenhouse gas emission under different soil nutrient management practices in soybean–wheat system of central India	331
469	Characterization of indoor diesel exhaust emissions from the parking garage of a school	332
470	Influence of transport from urban sources and domestic biomass combustion on the air quality of a mountain area	332
471	Airing ‘clean air’ in Clean India Mission	333
472	Personal exposure to PM2.5 associated with heavy metals in four travel modes of Tianjin during the summer season	334
473	Modeling of air pollutant concentrations in an industrial region of Turkey	334
474	Biomonitoring of atmospheric pollution: possibilities and future challenges	335
475	Assessment of dioxin-like activity in PM10 air samples from an industrial location in Algeria, using the DRE-CALUX bioassay	336
476	Exposure assessment of a cyclist to particles and chemical elements	337
477	Monitoring PAHs in the petrochemical area of Tarragona County, Spain: comparing passive air samplers with lichen transplants	337
478	Magnetic properties and element concentrations in lichens exposed to airborne pollutants released during cement production	338
479	The dynamic relationship between structural change and CO2 emissions in	338

	Malaysia: a cointegrating approach	
480	Optimal air quality policies and health: a multi-objective nonlinear approach	339
481	Traffic aerosol lobar doses deposited in the human respiratory system	340
482	Assessment of indoor climate of Mogiła Abbey in Kraków (Poland) and the application of the analogues method to predict microclimate indoor conditions	340
483	Dynamic of submicrometer particles in urban environment	341
484	Short-term effects of air pollution on daily hospital admissions for cardiovascular diseases in western China	341
485	Quantitative cancer risk assessment and local mortality burden for ambient air pollution in an eastern Mediterranean City	342
486	Carbon dioxide emissions, output, and energy consumption categories in Algeria	343
487	Chemical characteristics and source apportionment of PM _{2.5} using PCA/APCS, UNMIX, and PMF at an urban site of Delhi, India	343
488	Integrated management of ash from industrial and domestic combustion: a new sustainable approach for reducing greenhouse gas emissions from energy conversion	344
489	Impact of shipping emissions on ozone levels over Europe: assessing the relative importance of the Standard Nomenclature for Air Pollution (SNAP) categories	344
490	Effects of fine particulate matter and its constituents on emergency room visits for asthma in southern Taiwan during 2008–2010: a population-based study	345
491	Difference of performance in response to disease admissions between daily time air quality indices and those derived from average and entropy functions	346
492	The short-term associations of weather and air pollution with emergency ambulance calls for paroxysmal atrial fibrillation	346
493	Air pollution and respiratory health among diabetic and non-diabetic subjects in Pune, India—results from the Wellcome Trust Genetic Study	347
494	Portuguese agriculture and the evolution of greenhouse gas emissions—can vegetables control livestock emissions?	348
495	Impact of socioeconomic and meteorological factors on reservoirs' air quality: a case in the Three Gorges Reservoir of Chongqing (TGRC), China over a 10-year period	348
496	Air protection programmes in Poland in the context of the low emission	349
497	Human health impact assessment of exposure to particulate matter: an AirQ software modeling	350
498	Temporal comparison of global inventories of CO ₂ emissions from biomass burning during 2002–2011 derived from remotely sensed data	350
499	Historical and future emission of hazardous air pollutants (HAPs) from gas-fired combustion in Beijing, China	351
500	Does trade openness affect CO ₂ emissions: evidence from ten newly industrialized countries?	351
501	Geographic variations in female breast cancer incidence in relation to ambient air emissions of polycyclic aromatic hydrocarbons	352
502	Different exposure levels of fine particulate matter and preterm birth: a	353

	meta-analysis based on cohort studies	
503	A comparative study of hospital admissions for respiratory diseases during normal and dusty days in Iran	353
504	Influence of polymethyl acrylate additive on the formation of particulate matter and NOX emission of a biodiesel–diesel-fueled engine	354
505	Air pollution and hospital visits for acute upper and lower respiratory infections among children in Ningbo, China: A time-series analysis	354
506	Evaluation of air pollution tolerance index and anticipated performance index of plants and their application in development of green space along the urban areas	355
507	Potential lung carcinogenicity induced by chronic exposure to PM2.5 in the rat	356
508	Physicochemical properties, in vitro cytotoxic and genotoxic effects of PM1.0 and PM2.5 from Shanghai, China	356
509	Chemical characterization of size-segregated PM from different public transport modes and implications of source specific contribution to public exposure	357
510	Chemical characterization of size-segregated PM from different public transport modes and implications of source specific contribution to public exposure	358
511	Effects of air/fuel ratio on gas emissions in a small spark-ignited non-road engine operating with different gasoline/ethanol blends	358
512	Microplastic pollution in deposited urban dust, Tehran metropolis, Iran	359
513	Elemental and isotopic determination of lead (Pb) in particulate matter in the Brazilian city of Goiânia (GO) using ICP-MS technique	359
514	Pilot study investigating ambient air toxics emissions near a Canadian kraft pulp and paper facility in Pictou County, Nova Scotia	360
515	Loss of crop yields in India due to surface ozone: an estimation based on a network of observations	361
516	Seasonal variation of chemical composition and source apportionment of PM2.5 in Pune, India	362
517	Geographic variation in Chinese children' forced vital capacity and its association with long-term exposure to local PM10: a national cross-sectional study	362
518	Ozone generated by air purifier in low concentrations: friend or foe?	363
519	The effect of sources and air mass transport on the variability of trace element deposition in central Poland: a cluster-based approach	363
520	Emission analysis on the effect of nanoparticles on neat biodiesel in unmodified diesel engine	364
521	Real-world vehicle emissions as measured by in situ analysis of exhaust plumes	365
522	Performance, emission, and combustion characteristics of twin-cylinder common rail diesel engine fuelled with butanol-diesel blends	365
523	Simulations of the impacts of building height layout on air quality in natural-ventilated rooms around street canyons	366
	Science Of Total Environment	367-415
524	Variability, drivers, and effects of atmospheric nitrogen inputs across an	367

	urban area: Emerging patterns among human activities, the atmosphere, and soils	
525	Co-digestion of rice straw and cow dung to supply cooking fuel and fertilizers in rural India: Impact on human health, resource flows and climate change	367
526	Characteristics of black carbon in snow from Laohugou No. 12 glacier on the northern Tibetan Plateau	368
527	Temporal trends of surface urban heat islands and associated determinants in major Chinese cities	369
528	Projections of NH ₃ emissions from manure generated by livestock production in China to 2030 under six mitigation scenarios	369
529	Temporal and spatial distribution of PM _{2.5} chemical composition in a coastal city of Southeast China	370
530	Source apportionment of PM ₁₀ and PM _{2.5} in major urban Greek agglomerations using a hybrid source-receptor modeling process	371
531	Chemical characteristics and source apportionment of PM _{2.5} in Lanzhou, China	372
532	Spatio-temporal analysis of particulate matter intake fractions for vehicular emissions: Hourly variation by micro-environments in the Greater Toronto and Hamilton Area, Canada	372
533	Meteorological factors had more impact on airborne bacterial communities than air pollutants	373
534	A one-year, on-line, multi-site observational study on water-soluble inorganic ions in PM _{2.5} over the Pearl River Delta region, China	374
535	Refined 2013-based vehicle emission inventory and its spatial and temporal characteristics in Zhengzhou, China	374
536	Impact of dust and smoke mixing on column-integrated aerosol properties from observations during a severe wildfire episode over Valencia (Spain)	375
537	Acid rain footprint three decades after peak deposition: Long-term recovery from pollutant sulphate in the Uhlirská catchment (Czech Republic)	376
538	Impacts of Himalayas on black carbon over the Tibetan Plateau during summer monsoon	376
539	Airborne dust absorption by semi-arid forests reduces PM pollution in nearby urban environments	377
540	Review and analysis of strengths and weaknesses of agro-ecosystem models for simulating C and N fluxes	378
541	Substantial air quality and climate co-benefits achievable now with sectoral mitigation strategies in China	379
542	Ozone exposure affects tree defoliation in a continental climate	379
543	Source-receptor relationships for PM _{2.5} during typical pollution episodes in the Pearl River Delta city cluster, China	380
544	Biomass burning sources and their contributions to the local air quality in Hong Kong	381
545	Atmospheric removal of PM _{2.5} by man-made Three Northern Regions Shelter Forest in Northern China estimated using satellite retrieved PM _{2.5} concentration	381
546	Physicochemical characterization of ambient PM _{2.5} in Tehran air and its potential cytotoxicity in human lung epithelial cells (A549)	382
547	Temporal patterns of CO ₂ and CH ₄ in a rural area in northern Spain	383

	described by a harmonic equation over 2010–2016	
548	Influence of seasonality, air mass origin and particulate matter chemical composition on airborne bacterial community structure in the Po Valley, Italy	383
549	Long-term exposure to high air pollution induces cumulative DNA damages in traffic policemen	384
550	Ambient PM _{2.5} in the residential area near industrial complexes: Spatiotemporal variation, source apportionment, and health impact	385
551	Indoor air pollutants, ventilation rate determinants and potential control strategies in Chinese dwellings: A literature review	385
552	Extreme air pollution with contaminants originating from the mining–metallurgical processes	386
553	Linking potential heat source and sink to urban heat island: Heterogeneous effects of landscape pattern on land surface temperature	387
554	Vehicle emission trends in China's Guangdong Province from 1994 to 2014	387
555	Heavy pollution episodes, transport pathways and potential sources of PM _{2.5} during the winter of 2013 in Chengdu (China)	388
556	Particulate pollution in urban Chongqing of southwest China: Historical trends of variation, chemical characteristics and source apportionment	389
557	Emission factors for PM _{2.5} , CO, CO ₂ , NO _x , SO ₂ and particle size distributions from the combustion of wood species using a new controlled combustion chamber 3CE	389
558	Particulate matter pollution over China and the effects of control policies	390
559	Regional transport of anthropogenic pollution and dust aerosols in spring to Tianjin — A coastal megacity in China	391
560	Utilising green and bluespace to mitigate urban heat island intensity	391
561	An emission processing system for air quality modelling in the Mexico City metropolitan area: Evaluation and comparison of the MOBILE6.2-Mexico and MOVES-Mexico traffic emissions	392
562	An anomalous African dust event and its impact on aerosol radiative forcing on the Southwest Atlantic coast of Europe in February 2016	393
563	Long-range and regional transported size-resolved atmospheric aerosols during summertime in urban Shanghai	394
564	Estimating the contribution of regional transport to PM _{2.5} air pollution in a rural area on the North China Plain	394
565	The analysis and application of a new hybrid pollutants forecasting model using modified Kolmogorov–Zurbenko filter	395
566	Trends in population exposure to particulate matter in urban areas of Greece during the last decade	396
567	Shipping emissions and their impacts on air quality in China	396
568	Characteristics of PAHs in street dust of Beijing and the annual wash-off load using an improved load calculation method	397
569	Long-term trend of chemical composition of atmospheric precipitation at a regional background station in Northern China	398
570	Statistical analysis of Seoul air quality to assess the efficacy of emission abatement strategies since 1987	398
571	Modelling impacts of atmospheric deposition and temperature on long-term DOC trends	399
572	Benefits on public health from transport-related greenhouse gas mitigation	400

	policies in Southeastern European cities	
573	Atmospheric deposition of particles at a sensitive alpine lake: Size-segregated daily and annual fluxes from passive sampling techniques	400
574	Formation, features and controlling strategies of severe haze-fog pollutions in China	401
575	Haze, public health and mitigation measures in China: A review of the current evidence for further policy response	402
576	How does elevated ozone reduce methane emissions from peatlands?	402
577	Factors influencing the atmospheric concentrations of PCBs at an abandoned e-waste recycling site in South China	403
578	Seasonal and temperature modifications of the association between fine particulate air pollution and cardiovascular hospitalization in New York state	404
579	Seasonal analysis of the short-term effects of air pollution on daily mortality in Northeast Asia	404
580	Exposures to and origins of carbonaceous PM _{2.5} in a cookstove intervention in Northern Ghana	405
581	Impacts of household coal and biomass combustion on indoor and ambient air quality in China: Current status and implication	406
582	Effects of landscape composition and pattern on land surface temperature: An urban heat island study in the megacities of Southeast Asia	407
583	Air quality across a European hotspot: Spatial gradients, seasonality, diurnal cycles and trends in the Veneto region, NE Italy	407
584	A synchronous observation of enhanced aerosol and NO ₂ over Beijing, China, in winter 2015	408
585	Ozone pollution in China: A review of concentrations, meteorological influences, chemical precursors, and effects	409
586	The covariance of air quality conditions in six cities in Southern Germany - The role of meteorology	409
587	Forest soils in France are sequestering substantial amounts of carbon	410
588	Optical and radiative properties of aerosols over Desalpar, a remote site in western India: Source identification, modification processes and aerosol type discrimination	411
589	Tropospheric volatile organic compounds in China	411
590	Air quality perception of pedestrians in an urban outdoor Mediterranean environment: A field survey approach	412
591	Vertical distribution of ambient aerosol extinctive properties during haze and haze-free periods based on the Micro-Pulse Lidar observation in Shanghai	412
592	Estimating ground-level PM _{2.5} concentrations in Beijing, China using aerosol optical depth and parameters of the temperature inversion layer	413
593	A new-generation 3D ozone FACE (Free Air Controlled Exposure)	414
594	Assessment of PM _{2.5} sources and their corresponding level of uncertainty in a coastal urban area using EPA PMF 5.0 enhanced diagnostics	414
595	Quantifying the relationship between visibility degradation and PM _{2.5} constituents at a suburban site in Hong Kong: Differentiating contributions from hydrophilic and hydrophobic organic compounds	415

Atmospheric Chemistry & Physics Journal

A missing source of aerosols in Antarctica – beyond long-range transport, phytoplankton, and photochemistry

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Source: Atmos. Chem. Phys., 17, 1-20, 2017

<http://www.atmos-chem-phys.net/17/1/2017/doi:10.5194/acp-17-1-2017>

Understanding the sources and evolution of aerosols is crucial for constraining the impacts that aerosols have on a global scale. An unanswered question in atmospheric science is the source and evolution of the Antarctic aerosol population. Previous work over the continent has primarily utilized low temporal resolution aerosol filters to answer questions about the chemical composition of Antarctic aerosols. Bulk aerosol sampling has been useful in identifying seasonal cycles in the aerosol populations, especially in populations that have been attributed to Southern Ocean phytoplankton emissions. However, real-time, high-resolution chemical composition data are necessary to identify the mechanisms and exact timing of changes in the Antarctic aerosol. The recent ZODIAC (2-Season Ozone Depletion and Interaction with Aerosols Campaign) field campaign saw the first ever deployment of a real-time, high-resolution aerosol mass spectrometer (SP-AMS – soot particle aerosol mass spectrometer – or AMS) to the continent. Data obtained from the AMS, and a suite of other aerosol, gas-phase, and meteorological instruments, are presented here. In particular, this paper focuses on the aerosol population over coastal Antarctica and the evolution of that population in austral spring. Results indicate that there exists a sulfate mode in Antarctica that is externally mixed with a mass mode vacuum aerodynamic diameter of 250 nm. Springtime increases in sulfate aerosol are observed and attributed to biogenic sources, in agreement with previous research identifying phytoplankton activity as the source of the aerosol. Furthermore, the total Antarctic aerosol population is shown to undergo three distinct phases during the winter to summer transition. The first phase is dominated by highly aged sulfate particles comprising the majority of the aerosol mass at low wind speed. The second phase, previously unidentified, is the generation of a sub-250 nm aerosol population of unknown composition. The second phase appears as a transitional phase during the extended polar sunrise. The third phase is marked by an increased importance of biogenically derived sulfate to the total aerosol population (photolysis of dimethyl sulfate and methanesulfonic acid (DMS and MSA)). The increased importance of MSA is identified both through the direct, real-time measurement of aerosol MSA and through the use of positive matrix factorization on the sulfur-containing ions in the high-resolution mass-spectral data. Given the importance of sub-250 nm particles, the aforementioned second phase suggests that early austral spring is the season where new particle formation mechanisms are likely to have the largest contribution to the aerosol population in Antarctica.

Air quality improvement in a megacity: implications from 2015 Beijing Parade Blue pollution control actions

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Source: *Atmos. Chem. Phys.*, 17, 31-46, 2017

<http://www.atmos-chem-phys.net/17/31/2017/doi:10.5194/acp-17-31-2017>

The implementation of strict emission control measures in Beijing and surrounding regions during the 2015 China Victory Day Parade provided a valuable opportunity to investigate related air quality improvements in a megacity. We measured NH₃, NO₂ and PM_{2.5} at multiple sites in and outside Beijing and summarized concentrations of PM_{2.5}, PM₁₀, NO₂, SO₂ and CO in 291 cities across China from a national urban air quality monitoring network between August and September 2015. Consistently significant reductions of 12–35 % for NH₃ and 33–59 % for NO₂ in different areas of Beijing during the emission control period (referred to as the Parade Blue period) were observed compared with measurements in the pre- and post-Parade Blue periods without emission controls. Average NH₃ and NO₂ concentrations at sites near traffic were strongly correlated and showed positive and significant responses to traffic reduction measures, suggesting that traffic is an important source of both NH₃ and NO_x in urban Beijing. Daily concentrations of PM_{2.5} and secondary inorganic aerosol (sulfate, ammonium and nitrate) at the urban and rural sites both decreased during the Parade Blue period. During (after) the emission control period, concentrations of PM_{2.5}, PM₁₀, NO₂, SO₂ and CO from the national city-monitoring network showed the largest decrease (increase) of 34–72 % (50–214 %) in Beijing, a smaller decrease (a moderate increase) of 1–32 % (16–44 %) in emission control regions outside Beijing and an increase (decrease) of 6–16 % (–2–7 %) in non-emission-control regions of China. Integrated analysis of modelling and monitoring results demonstrated that emission control measures made a major contribution to air quality improvement in Beijing compared with a minor contribution from favourable meteorological conditions during the Parade Blue period. These results show that controls of secondary aerosol precursors (NH₃, SO₂ and NO_x) locally and regionally are key to curbing air pollution in Beijing and probably in other mega cities worldwide.

Atmospheric CO₂ observations and models suggest strong carbon uptake by forests in New Zealand

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Source: *Atmos. Chem. Phys.*, 17, 47-76, 2017

<http://www.atmos-chem-phys.net/17/47/2017/doi:10.5194/acp-17-47-2017>

A regional atmospheric inversion method has been developed to determine the spatial and temporal distribution of CO₂ sinks and sources across New Zealand for 2011–2013. This approach infers net air–sea and air–land CO₂ fluxes from measurement records, using back-trajectory simulations from the Numerical Atmospheric dispersion Modelling Environment (NAME) Lagrangian dispersion model, driven by meteorology from the New Zealand Limited Area Model (NZLAM) weather prediction model. The inversion uses in situ measurements from two fixed sites, Baring Head on the southern tip of New Zealand's North Island (41.408° S, 174.871° E) and Lauder from the central South Island (45.038° S, 169.684° E), and ship board data from monthly cruises between Japan, New Zealand, and Australia. A range of scenarios is used to assess the sensitivity of the inversion method to underlying assumptions and to ensure robustness of the results. The results indicate a strong seasonal cycle in terrestrial land fluxes from the South Island of New Zealand, especially in western regions covered by indigenous forest, suggesting higher photosynthetic and respiratory activity than is evident in the current a priori land process model. On the annual scale, the terrestrial biosphere in New Zealand is estimated to be a net CO₂ sink, removing 98 (±37) Tg CO₂ yr^{−1} from the atmosphere on average during 2011–2013. This sink is much larger than the reported 27 Tg CO₂ yr^{−1} from the national inventory for the same time period. The difference can be partially reconciled when factors related to forest and agricultural management and exports, fossil fuel emission estimates, hydrologic fluxes, and soil carbon change are considered, but some differences are likely to remain. Baseline uncertainty, model transport uncertainty, and limited sensitivity to the northern half of the North Island are the main contributors to flux uncertainty.

Modeling biogenic and anthropogenic secondary organic aerosol in China

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Source: Atmos. Chem. Phys., 17, 77–92, 2017

<http://www.atmos-chem-phys.net/17/77/2017/doi:10.5194/acp-17-77-2017>

A revised Community Multi-scale Air Quality (CMAQ) model with updated secondary organic aerosol (SOA) yields and a more detailed description of SOA formation from isoprene oxidation was applied to study the spatial and temporal distribution of SOA in China in the entire year of 2013. Predicted organic carbon (OC), elemental carbon and volatile organic compounds agreed favorably with observations at several urban areas, although the high OC concentrations in wintertime in Beijing were under-predicted. Predicted summer SOA was generally higher (10–15 µg m^{−3}) due to large contributions of isoprene (country average, 61 %), although the relative importance varies in different regions. Winter SOA was slightly lower and was mostly due to emissions of alkane and aromatic compounds (51 %). Contributions of monoterpene SOA was relatively constant (8–10 %). Overall, biogenic SOA accounted for approximately 75 % of total SOA in summer, 50–60 % in autumn and spring, and 24 % in winter. The Sichuan Basin had the highest predicted SOA concentrations in the country in all seasons, with hourly concentrations up to 50 µg m^{−3}. Approximately half of the SOA in all seasons was due to the traditional equilibrium partitioning of semivolatile components followed by oligomerization, while the remaining SOA was mainly due to reactive surface uptake of isoprene epoxide (5–14 %), glyoxal (14–25 %) and methylglyoxal (23–

28 %). Sensitivity analyses showed that formation of SOA from biogenic emissions was significantly enhanced due to anthropogenic emissions. Removing all anthropogenic emissions while keeping the biogenic emissions unchanged led to total SOA concentrations of less than $1 \mu\text{g m}^{-3}$, which suggests that manmade emissions facilitated biogenic SOA formation and controlling anthropogenic emissions would result in reduction of both anthropogenic and biogenic SOA.

Development of a high-resolution emission inventory and its evaluation and application through air quality modeling for Jiangsu Province, China

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Source: Atmos. Chem. Phys., 17, 211-233, 2017

<http://www.atmos-chem-phys.net/17/211/2017/doi:10.5194/acp-17-211-2017>

Improved emission inventories combining detailed source information are crucial for better understanding of the atmospheric chemistry and effectively making emission control policies using air quality simulation, particularly at regional or local scales. With the downscaled inventories directly applied, chemical transport models might not be able to reproduce the authentic evolution of atmospheric pollution processes at small spatial scales. Using the bottom-up approach, a high-resolution emission inventory was developed for Jiangsu China, including SO_2 , NO_x , CO, NH_3 , volatile organic compounds (VOCs), total suspended particulates (TSP), PM_{10} , $\text{PM}_{2.5}$, black carbon (BC), organic carbon (OC), and CO_2 . The key parameters relevant to emission estimation for over 6000 industrial sources were investigated, compiled, and revised at plant level based on various data sources and on-site surveys. As a result, the emission fractions of point sources were significantly elevated for most species. The improvement of this provincial inventory was evaluated through comparisons with other inventories at larger spatial scales, using satellite observation and air quality modeling. Compared to the downscaled Multi-resolution Emission Inventory for China (MEIC), the spatial distribution of NO_x emissions in our provincial inventory was more consistent with summer tropospheric NO_2 VCDs observed from OMI, particularly for the grids with moderate emission levels, implying the improved emission estimation for small and medium industrial plants by this work. Three inventories (national, regional, and provincial by this work) were applied in the Models-3 Community Multi-scale Air Quality (CMAQ) system for southern Jiangsu October 2012, to evaluate the model performances with different emission inputs. The best agreement between available ground observation and simulation was found when the provincial inventory was applied, indicated by the smallest normalized mean bias (NMB) and normalized mean errors (NME) for all the concerned species SO_2 , NO_2 , O_3 , and $\text{PM}_{2.5}$. The result thus implied the advantage of improved emission inventory at local scale for high-resolution air quality modeling. Under the unfavorable meteorology in which horizontal and vertical movement of atmosphere was limited, the simulated SO_2 concentrations at downtown Nanjing (the capital city of Jiangsu) using the regional or national inventories were much higher than those observed, implying that the urban emissions were overestimated when economy or population densities were applied to downscale or allocate the emissions. With more accurate spatial distribution of emissions at city level, the simulated concentrations using the provincial inventory were much closer to observation. Sensitivity analysis of $\text{PM}_{2.5}$ and O_3 formation was conducted using the improved provincial inventory through the brute force method. Iron and steel plants and cement plants were identified as important contributors to the $\text{PM}_{2.5}$ concentrations in Nanjing. The O_3 formation was VOC-limited in

southern Jiangsu, and the concentrations were negatively correlated with NO_x emissions in urban areas owing to the accumulated NO_x from transportation. More evaluations are further suggested for the impacts of speciation and temporal and vertical distribution of emissions on air quality modeling at regional or local scales in China.

Global inverse modeling of CH₄ sources and sinks: an overview of methods

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Source: Atmos. Chem. Phys., 17, 235-256, 2017

<http://www.atmos-chem-phys.net/17/235/2017/doi:10.5194/acp-17-235-2017>

The aim of this paper is to present an overview of inverse modeling methods that have been developed over the years for estimating the global sources and sinks of CH₄. It provides insight into how techniques and estimates have evolved over time and what the remaining shortcomings are. As such, it serves a didactical purpose of introducing apprentices to the field, but it also takes stock of developments so far and reflects on promising new directions. The main focus is on methodological aspects that are particularly relevant for CH₄, such as its atmospheric oxidation, the use of methane isotopologues, and specific challenges in atmospheric transport modeling of CH₄. The use of satellite retrievals receives special attention as it is an active field of methodological development, with special requirements on the sampling of the model and the treatment of data uncertainty. Regional scale flux estimation and attribution is still a grand challenge, which calls for new methods capable of combining information from multiple data streams of different measured parameters. A process model representation of sources and sinks in atmospheric transport inversion schemes allows the integrated use of such data. These new developments are needed not only to improve our understanding of the main processes driving the observed global trend but also to support international efforts to reduce greenhouse gas emissions.

Variations in O₃, CO, and CH₄ over the Bay of Bengal during the summer monsoon season: shipborne measurements and model simulations

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Source: Atmos. Chem. Phys., 17, 257-275, 2017

<http://www.atmos-chem-phys.net/17/257/2017/doi:10.5194/acp-17-257-2017>

We present shipborne measurements of surface ozone (O₃), carbon monoxide (CO), and methane (CH₄) over the Bay of Bengal (BoB), the first time such measurements have been performed during the summer monsoon season, as a part of the Continental Tropical Convergence Zone (CTCZ) experiment during 2009. O₃, CO, and CH₄ mixing ratios exhibited significant spatial and temporal

variability in the ranges of 8–54 nmol mol⁻¹, 50–200 nmol mol⁻¹, and 1.57–2.15 μmol mol⁻¹, with means of 29.7 ± 6.8 nmol mol⁻¹, 96 ± 25 nmol mol⁻¹, and 1.83 ± 0.14 μmol mol⁻¹, respectively. The average mixing ratios of trace gases over BoB in air masses from central/northern India (O₃: 30 ± 7 nmol mol⁻¹; CO: 95 ± 25 nmol mol⁻¹; CH₄: 1.86 ± 0.12 μmol mol⁻¹) were not statistically different from those in air masses from southern India (O₃: 27 ± 5 nmol mol⁻¹; CO: 101 ± 27 nmol mol⁻¹; CH₄: 1.72 ± 0.14 μmol mol⁻¹). Spatial variability is observed to be most significant for CH₄ with higher mixing ratios in the air masses from central/northern India, where higher CH₄ levels are seen in the SCIAMACHY (SCanning Imaging Absorption spectroMeter for Atmospheric CartographY) data. O₃ mixing ratios over the BoB showed large reductions (by ~ 20 nmol mol⁻¹) during four rainfall events. Temporal changes in the meteorological parameters, in conjunction with O₃ vertical profile, indicate that these low-O₃ events are associated with downdrafts of free-tropospheric O₃-poor air masses. While the observed variations of O₃ and CO are successfully reproduced using the Weather Research and Forecasting model with Chemistry (WRF-Chem), this model overestimates mean concentrations by about 6 and 16 % for O₃ and CO, respectively, generally overestimating O₃ mixing ratios during the rainfall events. An analysis of modelled O₃ along air mass trajectories show mean en route O₃ production rate of about 4.6 nmol mol⁻¹ day⁻¹ in the outflow towards the BoB. Analysis of the various tendencies from model simulations during an event on 10 August 2009, reproduced by the model, shows horizontal advection rapidly transporting O₃-rich air masses from near the coast across the BoB. This study fills a gap in the availability of trace gas measurements over the BoB and, when combined with data from previous campaigns, reveals large seasonal amplitude (~ 39 and ~ 207 nmol mol⁻¹ for O₃ and CO, respectively) over the northern BoB.

Characteristics of brown carbon in the urban Po Valley atmosphere

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Source: *Atmos. Chem. Phys.*, 17, 313–326, 2017

<http://www.atmos-chem-phys.net/17/313/2017/doi:10.5194/acp-17-313-2017>

We investigate optical–microphysical–chemical properties of brown carbon (BrC) in the urban ambient atmosphere of the Po Valley. In situ ground measurements of aerosol spectral optical properties, PM₁ chemical composition (HR-ToF-AMS), and particle size distributions were carried out in Bologna. BrC was identified through its wavelength dependence of light absorption at visible wavelengths, as indicated by the absorption Ångström exponent (AAE). We found that BrC occurs in particles with a narrow monomodal size distribution peaking in the droplet mode, enriched in ammonium nitrate and poor in black carbon (BC), with a strong dependence on OA-to-BC ratios, and SSA₅₃₀ of 0.98 ± 0.01. We demonstrate that specific complex refractive index values (k₅₃₀ = 0.017 ± 0.001) are necessary in addition to a proper particle size range to match the large AAEs measured for this BrC (AAE_{467–660} = 3.2 ± 0.9 with values up to 5.3). In terms of consistency of these findings with literature, this study

i. provides experimental evidence of the size distribution of BrC associated with the formation of secondary aerosol;

ii. shows that in the lower troposphere AAE increases with increasing OA-to-BC ratios rather than with increasing OA – contributing to sky radiometer retrieval techniques (e.g., AERONET);

iii. extends the dependence of AAE on BC-to-OA ratios previously observed in chamber experiments to ambient aerosol dominated by wood-burning emissions.

These findings are expected to bear important implications for atmospheric modeling studies and remote sensing observations as regards the parametrization and identification of BrC in the atmosphere.

Size-selected black carbon mass distributions and mixing state in polluted and clean environments of northern India

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Source: *Atmos. Chem. Phys.*, 17, 371-383, 2017

<http://www.atmos-chem-phys.net/17/371/2017/doi:10.5194/acp-17-371-2017>

We have measured black carbon properties by using a size-selected single-particle soot photometer (SP2). The measurements were conducted in northern India at two sites: Gual Pahari is located at the Indo-Gangetic Plain (IGP) and Mukteshwar at the Himalayan foothills. Northern India is known as one of the absorbing aerosol hot spots, but detailed information about absorbing aerosol mixing state is still largely missing. Previous equivalent black carbon (eBC) mass concentration measurements are available for this region, and these are consistent with our observations showing that refractory black carbon (rBC) concentrations are about 10 times higher in Gual Pahari than those at Mukteshwar. Also, the number fraction of rBC-containing particles is higher in Gual Pahari, but individual rBC-containing particles and their size distributions are fairly similar. These findings indicate that particles at both sites have similar local and regional emission sources, but aerosols are also transported from the main source regions (IGP) to the less polluted regions (Himalayan foothills). Detailed examination of the rBC-containing particle properties revealed that they are most likely irregular particles such as fractal aggregates, but the exact structure remains unknown.

Impacts of air pollution and climate on materials in Athens, Greece

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Source: *Atmos. Chem. Phys.*, 17, 439-448, 2017

<http://www.atmos-chem-phys.net/17/439/2017/doi:10.5194/acp-17-439-2017>

For more than 10 years now the National and Kapodistrian University of Athens, Greece, has contributed to the UNECE (United Nations Economic Commission for Europe) ICP Materials (International Co-operative Programme on Effects on Materials including Historic and Cultural Monuments) programme for monitoring the corrosion/soiling levels of different kinds of materials

due to environmental air-quality parameters. In this paper we present the results obtained from the analysis of observational data that were collected in Athens during the period 2003–2012. According to these results, the corrosion/soiling of the particular exposed materials tends to decrease over the years, except for the case of copper. Based on this long experimental database that is applicable to the multi-pollutant situation in the Athens basin, we present dose–response functions (DRFs) considering that dose stands for the air pollutant concentration, response for the material mass loss (normally per annum) and function, the relationship derived by the best statistical fit to the data.

Background aerosol over the Himalayas and Tibetan Plateau: observed characteristics of aerosol mass loading

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Source: Atmos. Chem. Phys., 17, 449–463, 2017

<http://www.atmos-chem-phys.net/17/449/2017/doi:10.5194/acp-17-449-2017>

To investigate the atmospheric aerosols of the Himalayas and Tibetan Plateau (HTP), an observation network was established within the region's various ecosystems, including at the Ngari, Qomolangma (QOMS), Nam Co, and Southeastern Tibetan (SET) stations. In this paper we illustrate aerosol mass loadings by integrating in situ measurements with satellite and ground-based remote sensing datasets for the 2011–2013 period, on both local and large scales. Mass concentrations of these surface atmospheric aerosols were relatively low and varied with land cover, showing a general tendency of Ngari and QOMS (barren sites) > Nam Co (grassland site) > SET (forest site). Daily averages of online PM_{2.5} (particulates with aerodynamic diameters below 2.5 μm) at these sites were sequentially 18.2 ± 8.9 , 14.5 ± 7.4 , 11.9 ± 4.9 and $11.7 \pm 4.7 \mu\text{g m}^{-3}$. Correspondingly, the ratios of PM_{2.5} to total suspended particles (TSP) were 27.4 ± 6.65 , 22.3 ± 10.9 , 37.3 ± 11.1 and $54.4 \pm 6.72\%$. Bimodal mass distributions of size-segregated particles were found at all sites, with a relatively small peak in accumulation mode and a more notable peak in coarse mode. Diurnal variations in fine-aerosol masses generally displayed a bi-peak pattern at the QOMS, Nam Co and SET stations and a single-peak pattern at the Ngari station, controlled by the effects of local geomorphology, mountain-valley breeze circulation and aerosol emissions. Dust aerosol content in PM_{2.1} samples gave fractions of 26 % at the Ngari station and 29 % at the QOMS station, or ~ 2 –3 times that of reported results at human-influenced sites. Furthermore, observed evidence confirmed the existence of the aerodynamic conditions necessary for the uplift of fine particles from a barren land surface. Combining surface aerosol data and atmospheric-column aerosol optical properties, the TSP mass and aerosol optical depth (AOD) of the Multi-angle Imaging Spectroradiometer (MISR) generally decreased as land cover changed from barren to forest, in inverse relation to the PM_{2.5} ratios. The seasonality of aerosol mass parameters was land-cover dependent. Over forest and grassland areas, TSP mass, PM_{2.5} mass, MISR-AOD and fine-mode AOD were higher in spring and summer, followed by relatively lower values in autumn and winter. At the barren site (the QOMS station), there were inconsistent seasonal patterns between surface TSP mass (PM_{2.5} mass) and atmospheric column AOD (fine-mode AOD). Our findings implicate that HTP aerosol masses (especially their regional characteristics and fine-particle emissions) need to be treated sensitively in relation to assessments of their climatic effect and potential role as cloud condensation nuclei and ice nuclei.

Nocturnal new particle formation events in urban environments

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Source: *Atmos. Chem. Phys.*, 17, 521-530, 2017

<http://www.atmos-chem-phys.net/17/521/2017/doi:10.5194/acp-17-521-2017>

Few studies have investigated nocturnal new particle formation (NPF) events, and none of them were conducted in urban environments. Nocturnal NPF can potentially be a significant source of particles in urban areas, and studying them would improve our understanding of nucleation mechanisms. To address this, our study was conducted in an urban environment to investigate the physical characteristics of NPF events, with a particular focus on nocturnal events and the differences between them and the daytime NPF events. Particle number size distribution (PNSD) was measured for 2 weeks at each of 25 sites across an urban environment. A new method was proposed to automatically categorise NPF events based on growth rate (GR) in order to remove the bias related to the manual procedure. Out of 219 observed events, 118 and 101 were categorised into class I and II respectively and 73 happened during the nighttime which included more than 30 % of the events. GR and condensation sink (CS) were calculated and a slight negative relationship between GR and CS was observed. Nocturnal events displayed higher GRs compared to daylight ones which were on average about 10 %. Back trajectory analysis was also conducted to estimate the locations of the sources of daylight and nocturnal precursors. While the precursors related to daylight events originated from different locations with no particular pattern, back-trajectory analysis showed many air masses associated with nocturnal NPF events were transported from over the ocean. Overall, nocturnal NPF events were found to be a significant source of particles in the studied environment with different physical characteristics and/or sources compared to daylight events.

Diurnal variability of the atmospheric boundary layer height over a tropical station in the Indian monsoon region

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Source: *Atmos. Chem. Phys.*, 17, 531-549, 2017

<http://www.atmos-chem-phys.net/17/531/2017/doi:10.5194/acp-17-531-2017>

The diurnal variation of atmospheric boundary layer (ABL) height is studied using high-resolution radiosonde observations available at 3 h intervals for 3 days continuously from 34 intensive campaigns conducted during the period December 2010–March 2014 over a tropical station Gadanki (13.5° N, 79.2° E; 375 m), in the Indian monsoon region. The heights of the ABL during the different stages of its diurnal evolution, namely, the convective boundary layer (CBL), the stable boundary layer (SBL), and the residual layer (RL) are obtained to study the diurnal variabilities. A clear diurnal variation is observed in 9 campaigns out of the 34 campaigns. In 7 campaigns the SBL did not form in the entire day and in the remaining 18 campaigns the SBL formed intermittently. The SBL forms for 33–55 % of the time during nighttime and 9 and 25 % during the evening and morning hours, respectively. The mean SBL height is within 0.3 km above the surface which increases slightly just after midnight (02:00 IST) and remains almost constant until the morning. The mean CBL height is within 3.0 km above the surface, which generally increases from morning to

evening. The mean RL height is within 2 km above the surface which generally decreases slowly as the night progresses. The diurnal variation of the ABL height over the Indian region is stronger during the pre-monsoon and weaker during winter season. The CBL is higher during the summer monsoon and lower during the winter season while the RL is higher during the winter season and lower during the summer season. During all the seasons, the ABL height peaks during the afternoon (~ 14:00 IST) and remains elevated until evening (~ 17:00 IST). The ABL suddenly collapses at 20:00 IST and increases slightly in the night. Interestingly, it is found that the low level clouds have an effect on the ABL height variability, but the deep convective clouds do not. The lifting condensation level (LCL) is generally found to occur below the ABL for the majority of the database and they are randomly related.

Stratospheric tropical warming event and its impact on the polar and tropical troposphere

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Source: *Atmos. Chem. Phys.*, 17, 615-625, 2017

<http://www.atmos-chem-phys.net/17/615/2017/doi:10.5194/acp-17-615-2017>

Stratosphere–troposphere coupling is investigated in relation to middle atmospheric subtropical jet (MASTJ) variations in boreal winter. An exceptional strengthening of the MASTJ occurred in association with a sudden equatorward shift of the stratospheric polar night jet (PNJ) in early December 2011. This abrupt transformation of the MASTJ and PNJ had no apparent relation to the upward propagation of planetary waves from the troposphere. The impact of this stratospheric event penetrated into the troposphere in two regions: in the northern polar region and the tropics. Due to the strong MASTJ, planetary waves at higher latitudes were deflected and trapped in the northern polar region. Trapping of the planetary waves resulted in amplification of zonal wave number 1 component, which appeared in the troposphere as the development of a trough over the Atlantic sector and a ridge over the Eurasian sector. A strong MASTJ also suppressed the equatorward propagation of planetary waves, which resulted in weaker tropical stratospheric upwelling and produced anomalous warming in the tropical stratosphere. In the tropical tropopause layer (TTL), however, sublimation of ice clouds kept the temperature change minor. In the troposphere, an abrupt termination of a Madden–Julian Oscillation (MJO) event occurred following the static stability increase in the TTL. This termination suggests that the stratospheric event affected the convective episode in the troposphere.

The dynamical impact of Rossby wave breaking upon UK PM10 concentration

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Source: *Atmos. Chem. Phys.*, 17, 867-881, 2017

<http://www.atmos-chem-phys.net/17/867/2017/doi:10.5194/acp-17-867-2017>

Coarse particulate matter (PM₁₀) has long been understood to be hazardous to human health, with mortality rates increasing as a result of raised ground level concentrations. We explore the influence of synoptic-scale meteorology on daily mean observed PM₁₀ concentration ([PM₁₀]) using Rossby wave breaking (RWB). Meteorological reanalysis data for the winter months (DJF) between January 1999 and December 2008 and observed PM₁₀ data for three urban background UK (Midland) sites were analysed. Three RWB diagnostics were used to identify RWB that had significant influence on UK Midland PM₁₀. RWB events were classified according to whether the RWB was cyclonic or anticyclonic in its direction of breaking and whether the RWB event was influenced more by poleward or equatorial air masses.

We find that there is a strong link between RWB events and UK [PM₁₀]. Significant increases ($p < 0.01$) in UK [PM₁₀] were seen 1 day following RWB occurring in spatially constrained northeast Atlantic–European regions. Analysis into episodic PM₁₀ exceedance events shows increased probability of [PM₁₀] exceedance associated with all RWB subsets. The greatest probability of exceeding the UK [PM₁₀] threshold was associated with cyclonic RWB preceded by anticyclonic RWB forming an Ω block synoptic pattern. This mechanism suggests an easterly advection of European PM₁₀ followed by prolonged stagnant conditions within the UK and led to an almost threefold increase in the probability of the UK Midlands exceeding a hazardous [PM₁₀] threshold (0.383), when compared to days where no RWB was detected (0.129).

Determination of the atmospheric lifetime and global warming potential of sulfur hexafluoride using a three-dimensional model

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Source: Atmos. Chem. Phys., 17, 883–898, 2017

<http://www.atmos-chem-phys.net/17/883/2017/doi:10.5194/acp-17-883-2017>

We have used the Whole Atmosphere Community Climate Model (WACCM), with an updated treatment of loss processes, to determine the atmospheric lifetime of sulfur hexafluoride (SF₆). The model includes the following SF₆ removal processes: photolysis, electron attachment and reaction with mesospheric metal atoms. The Sodankylä Ion Chemistry (SIC) model is incorporated into the standard version of WACCM to produce a new version with a detailed D region ion chemistry with cluster ions and negative ions. This is used to determine a latitude- and altitude-dependent scaling factor for the electron density in the standard WACCM in order to carry out multi-year SF₆ simulations. The model gives a mean SF₆ lifetime over an 11-year solar cycle (τ) of 1278 years (with a range from 1120 to 1475 years), which is much shorter than the currently widely used value of 3200 years, due to the larger contribution (97.4 %) of the modelled electron density to the total atmospheric loss. The loss of SF₆ by reaction with mesospheric metal atoms (Na and K) is far too slow to affect the lifetime. We investigate how this shorter atmospheric lifetime impacts the use of SF₆ to derive stratospheric age of air. The age of air derived from this shorter lifetime SF₆ tracer is longer by 9 % in polar latitudes at 20 km compared to a passive SF₆ tracer. We also present laboratory measurements of the infrared spectrum of SF₆ and find good agreement with previous

studies. We calculate the resulting radiative forcings and efficiencies to be, on average, very similar to those reported previously. Our values for the 20-, 100- and 500-year global warming potentials are 18 000, 23 800 and 31 300, respectively.

MIX: a mosaic Asian anthropogenic emission inventory under the international collaboration framework of the MICS-Asia and HTAP

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Source: Atmos. Chem. Phys., 17, 935-963, 2017

<http://www.atmos-chem-phys.net/17/935/2017/doi:10.5194/acp-17-935-2017>

The MIX inventory is developed for the years 2008 and 2010 to support the Model Inter-Comparison Study for Asia (MICS-Asia) and the Task Force on Hemispheric Transport of Air Pollution (TF HTAP) by a mosaic of up-to-date regional emission inventories. Emissions are estimated for all major anthropogenic sources in 29 countries and regions in Asia. We conducted detailed comparisons of different regional emission inventories and incorporated the best available ones for each region into the mosaic inventory at a uniform spatial and temporal resolution. Emissions are aggregated to five anthropogenic sectors: power, industry, residential, transportation, and agriculture. We estimate the total Asian emissions of 10 species in 2010 as follows: 51.3 Tg SO₂, 52.1 Tg NO_x, 336.6 Tg CO, 67.0 Tg NMVOC (non-methane volatile organic compounds), 28.8 Tg NH₃, 31.7 Tg PM₁₀, 22.7 Tg PM_{2.5}, 3.5 Tg BC, 8.3 Tg OC, and 17.3 Pg CO₂. Emissions from China and India dominate the emissions of Asia for most of the species. We also estimated Asian emissions in 2006 using the same methodology of MIX. The relative change rates of Asian emissions for the period of 2006–2010 are estimated as follows: –8.1 % for SO₂, +19.2 % for NO_x, +3.9 % for CO, +15.5 % for NMVOC, +1.7 % for NH₃, –3.4 % for PM₁₀, –1.6 % for PM_{2.5}, +5.5 % for BC, +1.8 % for OC, and +19.9 % for CO₂. Model-ready speciated NMVOC emissions for SAPRC-99 and CB05 mechanisms were developed following a profile-assignment approach. Monthly gridded emissions at a spatial resolution of 0.25° × 0.25° are developed and can be accessed from <http://www.meicmodel.org/dataset-mix>.

Biomass burning aerosols and the low-visibility events in Southeast Asia

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Source: Atmos. Chem. Phys., 17, 965-980, 2017

<http://www.atmos-chem-phys.net/17/965/2017/doi:10.5194/acp-17-965-2017>

Fires including peatland burning in Southeast Asia have become a major concern to the general public as well as governments in the region. This is because aerosols emitted from such fires can cause persistent haze events under certain weather conditions in downwind locations, degrading visibility and causing human health issues. In order to improve our understanding of the spatiotemporal coverage and influence of biomass burning aerosols in Southeast Asia, we have used surface visibility and particulate matter concentration observations, supplemented by decade-long (2003 to 2014) simulations using the Weather Research and Forecasting (WRF) model with a fire aerosol module, driven by high-resolution biomass burning emission inventories. We find that in the past decade, fire aerosols are responsible for nearly all events with very low visibility (< 7 km). Fire aerosols alone are also responsible for a substantial fraction of low-visibility events (visibility < 10 km) in the major metropolitan areas of Southeast Asia: up to 39 % in Bangkok, 36 % in Kuala Lumpur, and 34 % in Singapore. Biomass burning in mainland Southeast Asia accounts for the largest contribution to total fire-produced PM_{2.5} in Bangkok (99 %), while biomass burning in Sumatra is a major contributor to fire-produced PM_{2.5} in Kuala Lumpur (50 %) and Singapore (41 %). To examine the general situation across the region, we have further defined and derived a new integrated metric for 50 cities of the Association of Southeast Asian Nations (ASEAN): the haze exposure day (HED), which measures the annual exposure days of these cities to low visibility (< 10 km) caused by particulate matter pollution. It is shown that HEDs have increased steadily in the past decade across cities with both high and low populations. Fire events alone are found to be responsible for up to about half of the total HEDs. Our results suggest that in order to improve the overall air quality in Southeast Asia, mitigation policies targeting both biomass burning and fossil fuel burning sources need to be implemented.

Quantifying local-scale dust emission from the Arabian Red Sea coastal plain

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Source: *Atmos. Chem. Phys.*, 17, 993-1015, 2017

<http://www.atmos-chem-phys.net/17/993/2017/doi:10.5194/acp-17-993-2017>

Dust plumes emitted from the narrow Arabian Red Sea coastal plain are often observed on satellite images and felt in local population centers. Despite its relatively small area, the coastal plain could be a significant dust source; however, its effect is not well quantified as it is not well approximated in global or even regional models. In addition, because of close proximity to the Red Sea, a significant amount of dust from the coastal areas could be deposited into the Red Sea and serve as a vital component of the nutrient balance of marine ecosystems.

In the current study, we apply the offline Community Land Model version 4 (CLM4) to better quantify dust emission from the coastal plain during the period of 2009–2011. We verify the spatial and temporal variability in model results using independent weather station reports. We also compare the results with the MERRA Aerosol Reanalysis (MERRAero). We show that the best results are obtained with 1 km model spatial resolution and dust source function based on Meteosat Second Generation Spinning Enhanced Visible and InfraRed Imager (SEVIRI) measurements. We present the dust emission spatial pattern, as well as estimates of seasonal and diurnal variability in dust event frequency and intensity, and discuss the emission regime in the

major dust generation hot spot areas. We demonstrate the contrasting seasonal dust cycles in the northern and southern parts of the coastal plain and discuss the physical mechanisms responsible for dust generation. This study provides the first estimates of the fine-scale spatial and temporal distribution of dust emissions from the Arabian Red Sea coastal plain constrained by MERRAero and short-term WRF-Chem simulations. The estimate of total dust emission from the coastal plain, tuned to fit emissions in MERRAero, is $7.5 \pm 0.5 \text{ Mt a}^{-1}$. Small interannual variability indicates that the study area is a stable dust source. The mineralogical composition analysis shows that the coastal plain generates around $76 \pm 5 \text{ kt}$ of iron oxides and $6 \pm 0.4 \text{ kt}$ of phosphorus annually. Over 65 % of dust is emitted from the northern part of the coastal plain.

Factors controlling black carbon distribution in the Arctic

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Source: *Atmos. Chem. Phys.*, 17, 1037-1059, 2017

<http://www.atmos-chem-phys.net/17/1037/2017/doi:10.5194/acp-17-1037-2017>

We investigate the sensitivity of black carbon (BC) in the Arctic, including BC concentration in snow (BC_{snow}, ng g⁻¹) and surface air (BC_{air}, ng m⁻³), as well as emissions, dry deposition, and wet scavenging using the global three-dimensional (3-D) chemical transport model (CTM) GEOS-Chem. We find that the model underestimates BC_{snow} in the Arctic by 40 % on average (median = 11.8 ng g⁻¹). Natural gas flaring substantially increases total BC emissions in the Arctic (by ~ 70 %). The flaring emissions lead to up to 49 % increases (0.1–8.5 ng g⁻¹) in Arctic BC_{snow}, dramatically improving model comparison with observations (50 % reduction in discrepancy) near flaring source regions (the western side of the extreme north of Russia). Ample observations suggest that BC dry deposition velocities over snow and ice in current CTMs (0.03 cm s⁻¹ in the GEOS-Chem) are too small. We apply the resistance-in-series method to compute a dry deposition velocity (vd) that varies with local meteorological and surface conditions. The resulting velocity is significantly larger and varies by a factor of 8 in the Arctic (0.03–0.24 cm s⁻¹), which increases the fraction of dry to total BC deposition (16 to 25 %) yet leaves the total BC deposition and BC_{snow} in the Arctic unchanged. This is largely explained by the offsetting higher dry and lower wet deposition fluxes. Additionally, we account for the effect of the Wegener–Bergeron–Findeisen (WBF) process in mixed-phase clouds, which releases BC particles from condensed phases (water drops and ice crystals) back to the interstitial air and thereby substantially reduces the scavenging efficiency of clouds for BC (by 43–76 % in the Arctic). The resulting BC_{snow} is up to 80 % higher, BC loading is considerably larger (from 0.25 to 0.43 mg m⁻²), and BC lifetime is markedly prolonged (from 9 to 16 days) in the Arctic. Overall, flaring emissions increase BC_{air} in the Arctic (by ~ 20 ng m⁻³), the updated vd more than halves BC_{air} (by ~ 20 ng m⁻³), and the WBF effect increases BC_{air} by 25–70 % during winter and early spring. The resulting model simulation of BC_{snow} is substantially improved (within 10 % of the observations) and the discrepancies of BC_{air} are much smaller during the snow season at Barrow, Alert, and Summit (from –67––47 % to –46–3 %). Our results point toward an urgent need for better characterization of flaring emissions of BC (e.g., the emission factors, temporal, and spatial distribution), extensive measurements of both the dry deposition of BC over snow and ice, and the scavenging efficiency of BC in mixed-phase

clouds. In addition, we find that the poorly constrained precipitation in the Arctic may introduce large uncertainties in estimating BC_{snow}. Doubling precipitation introduces a positive bias approximately as large as the overall effects of flaring emissions and the WBF effect; halving precipitation produces a similarly large negative bias.

Chemistry–climate interactions of aerosol nitrate from lightning

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Source: *Atmos. Chem. Phys.*, 17, 1125-1142, 2017

<http://www.atmos-chem-phys.net/17/1125/2017/doi:10.5194/acp-17-1125-2017>

Lightning represents one of the dominant emission sources for NO_x in the troposphere. The direct release of oxidised nitrogen in the upper troposphere does not only affect ozone formation, but also chemical and microphysical properties of aerosol particles in this region. This study investigates the direct impact of LNO_x emissions on upper-tropospheric nitrate using a global chemistry climate model. The simulation results show a substantial influence of the lightning emissions on the mixing ratios of nitrate aerosol in the upper troposphere of more than 50 %. In addition to the impact on nitrate, lightning substantially affects the oxidising capacity of the atmosphere with substantial implications for gas-phase sulfate formation and new particle formation in the upper troposphere. In conjunction with the condensation of nitrates, substantial differences in the aerosol size distribution occur in the upper troposphere as a consequence of lightning. This has implications for the extinction properties of the aerosol particles and for the cloud optical properties. While the extinction is generally slightly enhanced due to the LNO_x emissions, the response of the clouds is ambiguous due to compensating effects in both liquid and ice clouds. Resulting shortwave flux perturbations are of $\sim -100 \text{ mW m}^{-2}$ as determined from several sensitivity scenarios, but an uncertainty range of almost 50 % has to be defined due to the large internal variability of the system and the uncertainties in the multitude of involved processes. Despite the clear statistical significance of the influence of lightning on the nitrate concentrations, the robustness of the findings gradually decreases towards the determination of the radiative flux perturbations.

Data assimilation for volcanic ash plumes using a satellite observational operator: a case study on the 2010 Eyjafjallajökull volcanic eruption

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Source: *Atmos. Chem. Phys.*, 17, 1187-1205, 2017

<http://www.atmos-chem-phys.net/17/1187/2017/doi:10.5194/acp-17-1187-2017>

Using data assimilation (DA) to improve model forecast accuracy is a powerful approach that requires available observations. Infrared satellite measurements of volcanic ash mass loadings are often used as input observations for the assimilation scheme. However, because these primary satellite-retrieved data are often two-dimensional (2-D) and the ash plume is usually vertically

located in a narrow band, directly assimilating the 2-D ash mass loadings in a three-dimensional (3-D) volcanic ash model (with an integral observational operator) can usually introduce large artificial/spurious vertical correlations.

In this study, we look at an approach to avoid the artificial vertical correlations by not involving the integral operator. By integrating available data of ash mass loadings and cloud top heights, as well as data-based assumptions on thickness, we propose a satellite observational operator (SOO) that translates satellite-retrieved 2-D volcanic ash mass loadings to 3-D concentrations. The 3-D SOO makes the analysis step of assimilation comparable in the 3-D model space.

Ensemble-based DA is used to assimilate the extracted measurements of ash concentrations. The results show that satellite DA with SOO can improve the estimate of volcanic ash state and the forecast. Comparison with both satellite-retrieved data and aircraft in situ measurements shows that the effective duration of the improved volcanic ash forecasts for the distal part of the Eyjafjallajökull volcano is about 6 h.

Direct observations of organic aerosols in common wintertime hazes in North China: insights into direct emissions from Chinese residential stoves

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Source: Atmos. Chem. Phys., 17, 1259-1270, 2017

<http://www.atmos-chem-phys.net/17/1259/2017/doi:10.5194/acp-17-1259-2017>

Many studies have focused on the physicochemical properties of aerosol particles in unusually severe haze episodes in North China instead of the more frequent and less severe hazes. Consistent with this lack of attention, the morphology and mixing state of organic matter (OM) particles in the frequent light and moderate (L & M) hazes in winter in the North China Plain (NCP) have not been examined, even though OM dominates these fine particles. In the present work, morphology, mixing state, and size of organic aerosols in the L & M hazes were systematically characterized using transmission electron microscopy coupled with energy-dispersive X-ray spectroscopy, atomic force microscopy, and nanoscale secondary ion mass spectrometer, with the comparisons among an urban site (Jinan, S1), a mountain site (Mt. Tai, S2), and a background island site (Changdao, S3) in the same hazes. Based on their morphologies, the OM particles were divided into six different types: spherical (type 1), near-spherical (type 2), irregular (type 3), domelike (type 4), dispersed-OM (type 5), and OM-coating (type 6). In the three sampling sites, types 1–3 of OM particles were most abundant in the L & M hazes and most of them were internally mixed with non-OM particles. The abundant near-spherical OM particles with higher sphericity and lower aspect ratio indicate that these primary OM particles formed in the cooling process after polluted plumes were emitted from coal combustion and biomass burning. Based on the Si-O-C ratio in OM particles, we estimated that 71 % of type 1–3 OM particles were associated with coal combustion. Our result suggests that coal combustion in residential stoves was a widespread source from urban to rural areas in NCP. Average OM thickness which correlates with the age of the air masses in type 6 particles only slightly increased from S1 to S2 to S3, suggesting that the L & M hazes were usually dry (relative humidity < 60 %) with weak photochemistry and heterogeneous reactions between particles and

gases. We conclude that the direct emissions from these coal stoves without any pollution controls in rural areas and in urban outskirts contribute large amounts of primary OM particles to the regional L & M hazes in North China.

Influence of enhanced Asian NO_x emissions on ozone in the upper troposphere and lower stratosphere in chemistry–climate model simulations

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Source: Atmos. Chem. Phys., 17, 1297-1311, 2017

<http://www.atmos-chem-phys.net/17/1297/2017/doi:10.5194/acp-17-1297-2017>

The Asian summer monsoon (ASM) anticyclone is the most pronounced circulation pattern in the upper troposphere and lower stratosphere (UTLS) during northern hemispheric summer. ASM convection plays an important role in efficient vertical transport from the surface to the upper-level anticyclone. In this paper we investigate the potential impact of enhanced anthropogenic nitrogen oxide (NO_x) emissions on the distribution of ozone in the UTLS using the fully coupled aerosol–chemistry–climate model, ECHAM5-HAMMOZ. Ozone in the UTLS is influenced both by the convective uplift of ozone precursors and by the uplift of enhanced-NO_x-induced tropospheric ozone anomalies. We performed anthropogenic NO_x emission sensitivity experiments over India and China. In these simulations, covering the years 2000–2010, anthropogenic NO_x emissions have been increased by 38 % over India and by 73 % over China with respect to the emission base year 2000. These emission increases are comparable to the observed linear trends of 3.8 % per year over India and 7.3 % per year over China during the period 2000 to 2010. Enhanced NO_x emissions over India by 38 % and China by 73 % increase the ozone radiative forcing in the ASM anticyclone (15–40° N, 60–120° E) by 16.3 and 78.5 mW m^{−2} respectively. These elevated NO_x emissions produce significant warming over the Tibetan Plateau and increase precipitation over India due to a strengthening of the monsoon Hadley circulation. However, increase in NO_x emissions over India by 73 % (similar to the observed increase over China) results in large ozone production over the Indo-Gangetic Plain and Tibetan Plateau. The higher ozone concentrations, in turn, induce a reversed monsoon Hadley circulation and negative precipitation anomalies over India. The associated subsidence suppresses vertical transport of NO_x and ozone into the ASM anticyclone.

Aerosol optical depth thresholds as a tool to assess diffuse radiation fertilization of the land carbon uptake in China

Xu Yue and Nadine Unger

Source: Atmos. Chem. Phys., 17, 1329-1342, 2017

<http://www.atmos-chem-phys.net/17/1329/2017/doi:10.5194/acp-17-1329-2017>

China suffers from frequent haze pollution episodes that alter the surface solar radiation and influence regional carbon uptake by the land biosphere. Here, we apply combined vegetation and radiation modeling and multiple observational datasets to assess the radiative effects of aerosol pollution in China on the regional land carbon uptake for the 2009–2011 period. First, we assess the inherent sensitivity of China's land biosphere to aerosol pollution by defining and calculating two thresholds of aerosol optical depth (AOD) at 550 nm, (i) AODt1, resulting in the maximum net primary productivity (NPP), and (ii) AODt2, such that if local AOD < AODt2, the aerosol diffuse fertilization effect (DFE) always promotes local NPP compared with aerosol-free conditions. Then, we apply the thresholds, satellite data, and interactive vegetation modeling to estimate current impacts of aerosol pollution on land ecosystems. In the northeast, observed AOD is 55 % lower than AODt1, indicating a strong aerosol DFE on local NPP. In the southeastern coastal regions, observed AOD is close to AODt1, suggesting that regional NPP is promoted by the current level of aerosol loading, but that further increases in AOD in this region will weaken the fertilization effects. The North China Plain experiences limited enhancement of NPP by aerosols because observed AOD is 77 % higher than AODt1 but 14 % lower than AODt2. Aerosols always inhibit regional NPP in the southwest because of the persistent high cloud coverage that already substantially reduces the total light availability there. Under clear-sky conditions, simulated NPP shows widespread increases of 20–60 % (35.0 ± 0.9 % on average) by aerosols. Under all-sky conditions, aerosol pollution has spatially contrasting opposite sign effects on NPP from –3 % to +6 % (1.6 ± 0.5 % on average), depending on the local AOD relative to the regional thresholds. Stringent aerosol pollution reductions motivated by public health concerns, especially in the North China Plain and the southwest, will help protect land ecosystem functioning in China and mitigate long-term global warming.

Qualitative and quantitative analysis of atmospheric organosulfates in Centreville, Alabama

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Source: *Atmos. Chem. Phys.*, 17, 1343–1359, 2017

<http://www.atmos-chem-phys.net/17/1343/2017/doi:10.5194/acp-17-1343-2017>

Organosulfates are components of secondary organic aerosols (SOA) that form from oxidation of volatile organic compounds (VOCs) in the presence of sulfate. In this study, the composition and abundance of organosulfates were determined in fine particulate matter (PM_{2.5}) collected from Centreville, AL, during the Southern Oxidant and Aerosol Study (SOAS) in summer 2013. Six organosulfates were quantified using hydrophilic interaction liquid chromatography (HILIC) with triple quadrupole mass spectrometry (TQD) against authentic standards. Among these, the three most abundant species were glycolic acid sulfate (0.5–52.5 ng m^{–3}), lactic acid sulfate (0.5–36.7 ng m^{–3}), and hydroxyacetone sulfate (0.5–14.3 ng m^{–3}). These three species were strongly inter-correlated, suggesting similar precursors and/or formation pathways. Further correlations with sulfate, isoprene, and isoprene oxidation products indicate important roles for these precursors in organosulfate formation in Centreville. Positive filter sampling artifacts associated with these organosulfates due to gas adsorption or reaction of gas phase precursors of organosulfates with sulfuric acid were assessed for a subset of samples and were less than 7.8 % of their PM_{2.5} concentrations. Together, the quantified organosulfates accounted for < 0.3 % of

organic carbon mass in PM_{2.5}. To gain insights into other organosulfates in PM_{2.5} collected from Centreville, semi-quantitative analysis was employed by way of monitoring characteristic product ions of organosulfates (HSO₄⁻ at *m/z* 97 and SO₄⁻ at *m/z* 96) and evaluating relative signal strength by HILIC-TQD. Molecular formulas of organosulfates were determined by high-resolution time-of-flight (TOF) mass spectrometry. The major organosulfate signal across all samples corresponded to 2-methyltetrol sulfates, which accounted for 42–62 % of the total bisulfate ion signal. Conversely, glycolic acid sulfate, the most abundant organosulfate quantified in this study, was 0.13–0.57 % of the total bisulfate ion signal. Precursors of *m/z* 96 mainly consisted of nitro-oxy organosulfates. Organosulfates identified were mainly associated with biogenic VOC precursors, particularly isoprene and to a lesser extent monoterpenes and 2-methyl-3-buten-2-ol (MBO). While a small number of molecules dominated the total organosulfate signal, a large number of minor species were also present. This study provides insights into the major organosulfate species in the southeastern US, as measured by tandem mass spectrometry that should be targets for future standard development and quantitative analysis.

Simulating ozone dry deposition at a boreal forest with a multi-layer canopy deposition model

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Source: *Atmos. Chem. Phys.*, 17, 1361-1379, 2017

<http://www.atmos-chem-phys.net/17/1361/2017/doi:10.5194/acp-17-1361-2017>

A multi-layer ozone (O₃) dry deposition model has been implemented into SOSAA (a model to Simulate the concentrations of Organic vapours, Sulphuric Acid and Aerosols) to improve the representation of O₃ concentration and flux within and above the forest canopy in the planetary boundary layer. We aim to predict the O₃ uptake by a boreal forest canopy under varying environmental conditions and analyse the influence of different factors on total O₃ uptake by the canopy as well as the vertical distribution of deposition sinks inside the canopy. The newly implemented dry deposition model was validated by an extensive comparison of simulated and observed O₃ turbulent fluxes and concentration profiles within and above the boreal forest canopy at SMEAR II (Station to Measure Ecosystem-Atmosphere Relations II) in Hyytiälä, Finland, in August 2010.

In this model, the fraction of wet surface on vegetation leaves was parametrised according to the ambient relative humidity (RH). Model results showed that when RH was larger than 70 % the O₃ uptake onto wet skin contributed ~ 51 % to the total deposition during nighttime and ~ 19 % during daytime. The overall contribution of soil uptake was estimated about 36 %. The contribution of sub-canopy deposition below 4.2 m was modelled to be ~ 38 % of the total O₃ deposition during daytime, which was similar to the contribution reported in previous studies. The chemical contribution to O₃ removal was evaluated directly in the model simulations. According to the simulated averaged diurnal cycle the net chemical production of O₃ compensated up to ~ 4 % of dry deposition loss from about 06:00 to 15:00 LT. During nighttime, the net chemical loss of O₃ further enhanced removal by dry deposition by a maximum ~ 9 %. Thus the results indicated an overall relatively small contribution of airborne chemical processes to O₃ removal at this site.

Potential sources and processes affecting speciated atmospheric mercury at Kejimikujik National Park, Canada: comparison of receptor models and data treatment methods

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Source: Atmos. Chem. Phys., 17, 1381-1400, 2017

<http://www.atmos-chem-phys.net/17/1381/2017/doi:10.5194/acp-17-1381-2017>

Source apportionment analysis was conducted with positive matrix factorization (PMF) and principal component analysis (PCA) methods using concentrations of speciated mercury (Hg), i.e., gaseous elemental mercury (GEM), gaseous oxidized mercury (GOM), and particulate-bound mercury (PBM), and other air pollutants collected at Kejimikujik National Park, Nova Scotia, Canada, in 2009 and 2010. The results were largely consistent between the 2 years for both methods. The same four source factors were identified in each year using PMF method. In both years, factor photochemistry and re-emission had the largest contributions to atmospheric Hg, while the contributions of combustion emission and industrial sulfur varied slightly between the 2 years. Four components were extracted with air pollutants only in each year using PCA method. Consistencies between the results of PMF and PCA include (1) most or all PMF factors overlapped with PCA components, (2) both methods suggest strong impact of photochemistry but little association between ambient Hg and sea salt, and (3) shifting of PMF source profiles and source contributions from one year to another was echoed in PCA. Inclusion of meteorological parameters led to identification of an additional component, Hg wet deposition in PCA, while it did not affect the identification of other components.

The PMF model performance was comparable in 2009 and 2010. Among the three Hg forms, the agreements between model-reproduced and observed annual mean concentrations were excellent for GEM, very good for PBM, and acceptable for GOM. However, on a daily basis, the agreement was very good for GEM but poor for GOM and PBM. Sensitivity tests suggest that increasing sample size by imputation is not effective in improving model performance, while reducing the fraction of concentrations below method detection limit, by either scaling GOM and PBM to higher concentrations or combining them to reactive mercury, is effective. Most of the data treatment options considered had little impact on the source identification or contribution.

The impact of resolution on meteorological, chemical and aerosol properties in regional simulations with WRF-Chem

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Source: Atmos. Chem. Phys., 17, 1511-1528, 2017

<http://www.atmos-chem-phys.net/17/1511/2017/doi:10.5194/acp-17-1511-2017>

Limited area (regional) models applied at high resolution over specific regions of interest are generally expected to more accurately capture the spatiotemporal variability of key meteorological and climate parameters. However, improved performance is not inevitable, and there remains a need to optimize use of numerical resources and to quantify the impact on simulation fidelity that

derives from increased resolution. The application of regional models for climate forcing assessment is currently limited by the lack of studies quantifying the sensitivity to horizontal spatial resolution and the physical–dynamical–chemical schemes driving the simulations. Here we investigate model skill in simulating meteorological, chemical and aerosol properties as a function of spatial resolution, by applying the Weather Research and Forecasting model with coupled Chemistry (WRF-Chem) over eastern North America at different resolutions. Using Brier skill scores and other statistical metrics it is shown that enhanced resolution (from 60 to 12 km) improves model performance for all of the meteorological parameters and gas-phase concentrations considered, in addition to both mean and extreme aerosol optical depth (AOD) in three wavelengths in the visible relative to satellite observations, principally via increase of potential skill. Some of the enhanced model performance for AOD appears to be attributable to improved simulation of meteorological conditions and the concentration of key aerosol precursor gases (e.g., SO₂ and NH₃). Among other reasons, a dry bias in the specific humidity in the boundary layer and a substantial underestimation of total monthly precipitation in the 60 km simulations are identified as causes for the better performance of WRF-Chem simulations at 12 km.

Halogen chemistry reduces tropospheric O₃ radiative forcing

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Source: *Atmos. Chem. Phys.*, 17, 1557-1569, 2017

<http://www.atmos-chem-phys.net/17/1557/2017/doi:10.5194/acp-17-1557-2017>

Tropospheric ozone (O₃) is a global warming gas, but the lack of a firm observational record since the preindustrial period means that estimates of its radiative forcing (RFTO₃) rely on model calculations. Recent observational evidence shows that halogens are pervasive in the troposphere and need to be represented in chemistry-transport models for an accurate simulation of present-day O₃. Using the GEOS-Chem model we show that tropospheric halogen chemistry is likely more active in the present day than in the preindustrial. This is due to increased oceanic iodine emissions driven by increased surface O₃, higher anthropogenic emissions of bromo-carbons, and an increased flux of bromine from the stratosphere. We calculate preindustrial to present-day increases in the tropospheric O₃ burden of 113 Tg without halogens but only 90 Tg with, leading to a reduction in RFTO₃ from 0.43 to 0.35 Wm⁻². We attribute ~ 50 % of this reduction to increased bromine flux from the stratosphere, ~ 35 % to the ocean–atmosphere iodine feedback, and ~ 15 % to increased tropospheric sources of anthropogenic halogens. This reduction of tropospheric O₃ radiative forcing due to halogens (0.087 Wm⁻²) is greater than that from the radiative forcing of stratospheric O₃ (~ 0.05 Wm⁻²). Estimates of RFTO₃ that fail to consider halogen chemistry are likely overestimates (~ 25 %).

Uptake of nitric acid, ammonia, and organics in orographic clouds: mass spectrometric analyses of droplet residual and interstitial aerosol particles

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Source: *Atmos. Chem. Phys.*, 17, 1571-1593, 2017

<http://www.atmos-chem-phys.net/17/1571/2017/doi:10.5194/acp-17-1571-2017>

Concurrent in situ analyses of interstitial aerosol and cloud droplet residues have been conducted at the Schmücke mountain site during the Hill Cap Cloud Thuringia campaign in central Germany in September and October 2010. Cloud droplets were sampled from warm clouds (temperatures between -3 and $+16$ °C) by a counterflow virtual impactor and the submicron-sized residues were analyzed by a compact time-of-flight aerosol mass spectrometer (C-ToF-AMS), while the interstitial aerosol composition was measured by an high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS). During cloud-free periods, the submicron out-of-cloud aerosol was analyzed using both instruments, allowing for intercomparison between the two instruments. Further instrumentation included black carbon measurements and optical particle counters for the aerosol particles as well as optical sizing instrumentation for the cloud droplets. The results show that, under cloud conditions, on average 85 % of the submicron aerosol mass partitioned into the cloud liquid phase. Scavenging efficiencies of nitrate, ammonium, sulfate, and organics ranged between 60 and 100 %, with nitrate having, in general, the highest values. For black carbon, the scavenging efficiency was markedly lower (about 24 %). The nitrate and ammonium mass fractions were found to be markedly enhanced in cloud residues, indicating uptake of gaseous nitric acid and ammonia into the aqueous phase. This effect was found to be temperature dependent: at lower temperatures, the nitrate and ammonium mass fractions in the residues were higher. Also, the oxidation state of the organic matter in cloud residues was found to be temperature dependent: the O : C ratio was lower at higher temperatures. A possible explanation for this observation is a more effective uptake and/or higher concentrations of low-oxidized water-soluble volatile organic compounds, possibly of biogenic origin, at higher temperatures. Organic nitrates were observed in cloud residuals as well as in the out-of-cloud aerosol, but no indication of a preferred partitioning of organic nitrates into the aqueous phase or into the gas phase was detected. Assuming the uptake of nitric acid and ammonia in cloud droplets will be reversible, it will lead to a redistribution of nitrate and ammonium among the aerosol particles, leading to more uniform, internally mixed particles after several cloud passages.

Understanding severe winter haze events in the North China Plain in 2014: roles of climate anomalies

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Source: *Atmos. Chem. Phys.*, 17, 1641-1651, 2017

<http://www.atmos-chem-phys.net/17/1641/2017/doi:10.5194/acp-17-1641-2017>

Atmospheric pollution has become a serious environmental and social problem in China. Over the past 30 years, the number of winter (December–February) haze days over the North China Plain (WHDNCP) was greatest in 2014. In addition to anthropogenic influence, climate anomalies also played a role. Thus, it is necessary to analyze the anomalous atmosphere circulations associated with haze pollution of this year in detail. Near the surface, the weaker East Asian winter monsoon pattern, causing southerly winds over the North China Plain, could aggravate the situation of haze. In the lower and middle troposphere, taking the anticyclone circulation over North China as an intermediate system, the positive phases of the eastern Atlantic/western Russia (EA/WR), the western Pacific (WP), and the Eurasia (EU) patterns led to a worse air pollution dispersion condition that contributed to a larger number of WHDNCP. In 2014, these three patterns could be recognized from the wind anomalies in the lower troposphere. The preceding autumn (September–November) Arctic sea ice (ASI) anomalies over the eastern Hemisphere and the warmer winter surface over Eurasia might have induced or intensified the positive EA/WR pattern in 2014. These two external forcings, together with the pre-autumn sea surface temperature anomalies in the Pacific, might have also stimulated or enhanced the positive EU-like patterns. The anomalous surface temperature in autumn 2014 was efficient in intensifying anomalous circulations such as the positive phase of the WP pattern. The opposite case of minimum WHDNCP in 2010 further supports the mechanism of how EA/WR and WP patterns and associated external factors altered the local climate conditions to impact the WHDNCP.

Dust deposition and ambient PM₁₀ concentration in northwest China: spatial and temporal variability

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Source: *Atmos. Chem. Phys.*, 17, 1699–1711, 2017

<http://www.atmos-chem-phys.net/17/1699/2017/doi:10.5194/acp-17-1699-2017>

Eolian dust transport and deposition are important geophysical processes which influence global bio-geochemical cycles. Currently, reliable deposition data are scarce in central and east Asia. Located at the boundary of central and east Asia, Xinjiang Province of northwestern China has long played a strategic role in cultural and economic trade between Asia and Europe. In this paper, we investigated the spatial distribution and temporal variation in dust deposition and ambient PM₁₀ (particulate matter in aerodynamic diameter $\leq 10 \mu\text{m}$) concentration from 2000 to 2013 in Xinjiang Province. This variation was assessed using environmental monitoring records from 14 stations in the province. Over the 14 years, annual average dust deposition across stations in the province ranged from 255.7 to 421.4 t km⁻². Annual dust deposition was greater in southern Xinjiang (663.6 t km⁻²) than northern (147.8 t km⁻²) and eastern Xinjiang (194.9 t km⁻²). Annual average PM₁₀ concentration across stations in the province varied from 100 to 196 $\mu\text{g m}^{-3}$ and was 70, 115 and 239 $\mu\text{g m}^{-3}$ in northern, eastern and southern Xinjiang, respectively. The highest annual dust deposition (1394.1 t km⁻²) and ambient PM₁₀ concentration (352 $\mu\text{g m}^{-3}$) were observed in Hotan, which is located in southern Xinjiang and at the southern boundary of the Taklamakan Desert. Dust deposition was more intense during the spring and summer than other seasons. PM₁₀ was the main air pollutant that significantly influenced regional air quality. Annual average dust deposition increased logarithmically with ambient PM₁₀ concentration ($R^2 \geq 0.81$). While the annual average dust storm frequency remained unchanged from 2000 to 2013, there was

a positive relationship between dust storm days and dust deposition and PM₁₀ concentration across stations. This study suggests that sand storms are a major factor affecting the temporal variability and spatial distribution of dust deposition in northwest China.

Anthropogenic influences on the physical state of submicron particulate matter over a tropical forest

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The occurrence of nonliquid and liquid physical states of submicron atmospheric particulate matter (PM) downwind of an urban region in central Amazonia was investigated. Measurements were conducted during two intensive operating periods (IOP1 and IOP2) that took place during the wet and dry seasons of the GoAmazon2014/5 campaign. Air masses representing variable influences of background conditions, urban pollution, and regional- and continental-scale biomass burning passed over the research site. As the air masses varied, particle rebound fraction, an indicator of physical state, was measured in real time at ground level using an impactor apparatus. Micrographs collected by transmission electron microscopy confirmed that liquid particles adhered, while nonliquid particles rebounded. Relative humidity (RH) was scanned to collect rebound curves. When the apparatus RH matched ambient RH, 95 % of the particles adhered as a campaign average. Secondary organic material, produced for the most part by the oxidation of volatile organic compounds emitted from the forest, produces liquid PM over this tropical forest. During periods of anthropogenic influence, by comparison, the rebound fraction dropped to as low as 60 % at 95 % RH. Analyses of the mass spectra of the atmospheric PM by positive-matrix factorization (PMF) and of concentrations of carbon monoxide, total particle number, and oxides of nitrogen were used to identify time periods affected by anthropogenic influences, including both urban pollution and biomass burning. The occurrence of nonliquid PM at high RH correlated with these indicators of anthropogenic influence. A linear model having as output the rebound fraction and as input the PMF factor loadings explained up to 70 % of the variance in the observed rebound fractions. Anthropogenic influences can contribute to the presence of nonliquid PM in the atmospheric particle population through the combined effects of molecular species that increase viscosity when internally mixed with background PM and increased concentrations of nonliquid anthropogenic particles in external mixtures of anthropogenic and biogenic PM.

Cleaning up the air: effectiveness of air quality policy for SO₂ and NO_x emissions in China

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Source: *Atmos. Chem. Phys.*, 17, 1775-1789, 2017

<http://www.atmos-chem-phys.net/17/1775/2017/doi:10.5194/acp-17-1775-2017>

Air quality observations by satellite instruments are global and have a regular temporal resolution, which makes them very useful in studying long-term trends in atmospheric species. To monitor air quality trends in China for the period 2005–2015, we derive SO₂ columns and NO_x emissions on a provincial level with improved accuracy. To put these trends into perspective they are compared with public data on energy consumption and the environmental policies of China. We distinguish the effect of air quality regulations from economic growth by comparing them relatively to fossil fuel consumption. Pollutant levels, per unit of fossil fuel, are used to assess the effectiveness of air quality regulations. We note that the desulfurization regulations enforced in 2005–2006 only had a significant effect in the years 2008–2009, when a much stricter control of the actual use of the installations began. For national NO_x emissions a distinct decreasing trend is only visible from 2012 onwards, but the emission peak year differs from province to province. Unlike SO₂, emissions of NO_x are highly related to traffic. Furthermore, regulations for NO_x emissions are partly decided on a provincial level. The last 3 years show a reduction both in SO₂ and NO_x emissions per fossil fuel unit, since the authorities have implemented several new environmental regulations. Despite an increasing fossil fuel consumption and a growing transport sector, the effects of air quality policy in China are clearly visible. Without the air quality regulations the concentration of SO₂ would be about 2.5 times higher and the NO₂ concentrations would be at least 25 % higher than they are today in China.

Particulate-phase mercury emissions from biomass burning and impact on resulting deposition: a modelling assessment

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Source: *Atmos. Chem. Phys.*, 17, 1881-1899, 2017

<http://www.atmos-chem-phys.net/17/1881/2017/doi:10.5194/acp-17-1881-2017>

Mercury (Hg) emissions from biomass burning (BB) are an important source of atmospheric Hg and a major factor driving the interannual variation of Hg concentrations in the troposphere. The greatest fraction of Hg from BB is released in the form of elemental Hg (Hg⁰(g)). However, little is known about the fraction of Hg bound to particulate matter (HgP) released from BB, and the factors controlling this fraction are also uncertain. In light of the aims of the Minamata Convention to reduce intentional Hg use and emissions from anthropogenic activities, the relative importance of Hg emissions from BB will have an increasing impact on Hg deposition fluxes. Hg speciation is one of the most important factors determining the redistribution of Hg in the atmosphere and the geographical distribution of Hg deposition. Using the latest version of the Global Fire Emissions Database (GFEDv4.1s) and the global Hg chemistry transport model, ECHMERIT, the impact of Hg speciation in BB emissions, and the factors which influence speciation, on Hg deposition have been investigated for the year 2013. The role of other uncertainties related to physical and chemical atmospheric processes involving Hg and the influence of model parametrisations were also

investigated, since their interactions with Hg speciation are complex. The comparison with atmospheric HgP concentrations observed at two remote sites, Amsterdam Island (AMD) and Manaus (MAN), in the Amazon showed a significant improvement when considering a fraction of HgP from BB. The set of sensitivity runs also showed how the quantity and geographical distribution of HgP emitted from BB has a limited impact on a global scale, although the inclusion of increasing fractions HgP does limit Hg₀(g) availability to the global atmospheric pool. This reduces the fraction of Hg from BB which deposits to the world's oceans from 71 to 62 %. The impact locally is, however, significant on northern boreal and tropical forests, where fires are frequent, uncontrolled and lead to notable Hg inputs to local ecosystems. In the light of ongoing climatic changes this effect could be potentially be exacerbated in the future.

The CAMS interim Reanalysis of Carbon Monoxide, Ozone and Aerosol for 2003–2015

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Source: *Atmos. Chem. Phys.*, 17, 1945–1983, 2017

<http://www.atmos-chem-phys.net/17/1945/2017/doi:10.5194/acp-17-1945-2017>

A new global reanalysis data set of atmospheric composition (AC) for the period 2003–2015 has been produced by the Copernicus Atmosphere Monitoring Service (CAMS). Satellite observations of total column (TC) carbon monoxide (CO) and aerosol optical depth (AOD), as well as several TC and profile observations of ozone, have been assimilated with the Integrated Forecasting System for Composition (C-IFS) of the European Centre for Medium-Range Weather Forecasting. Compared to the previous Monitoring Atmospheric Composition and Climate (MACC) reanalysis (MACCRA), the new CAMS interim reanalysis (CAMSiRA) is of a coarser horizontal resolution of about 110 km, compared to 80 km, but covers a longer period with the intent to be continued to present day. This paper compares CAMSiRA with MACCRA and a control run experiment (CR) without assimilation of AC retrievals. CAMSiRA has smaller biases than the CR with respect to independent observations of CO, AOD and stratospheric ozone. However, ozone at the surface could not be improved by the assimilation because of the strong impact of surface processes such as dry deposition and titration with nitrogen monoxide (NO), which were both unchanged by the assimilation. The assimilation of AOD led to a global reduction of sea salt and desert dust as well as an exaggerated increase in sulfate. Compared to MACCRA, CAMSiRA had smaller biases for AOD, surface CO and TC ozone as well as for upper stratospheric and tropospheric ozone. Finally, the temporal consistency of CAMSiRA was better than the one of MACCRA. This was achieved by using a revised emission data set as well as by applying careful selection and bias correction to the assimilated retrievals. CAMSiRA is therefore better suited than MACCRA for the study of interannual variability, as demonstrated for trends in surface CO.

Contributions of trans-boundary transport to summertime air quality in Beijing, China

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Source: *Atmos. Chem. Phys.*, 17, 2035-2051, 2017

<http://www.atmos-chem-phys.net/17/2035/2017/doi:10.5194/acp-17-2035-2017>

In the present study, the WRF-CHEM model is used to evaluate the contributions of trans-boundary transport to the air quality in Beijing during a persistent air pollution episode from 5 to 14 July 2015 in Beijing–Tianjin–Hebei (BTH), China. Generally, the predicted temporal variations and spatial distributions of PM_{2.5} (fine particulate matter), O₃ (ozone), and NO₂ are in good agreement with observations in BTH. The WRF-CHEM model also reproduces reasonably well the temporal variations of aerosol species compared to measurements in Beijing. The factor separation approach is employed to evaluate the contributions of trans-boundary transport of non-Beijing emissions to the PM_{2.5} and O₃ levels in Beijing. On average, in the afternoon during the simulation episode, the local emissions contribute 22.4 % to the O₃ level in Beijing, less than 36.6 % from non-Beijing emissions. The O₃ concentrations in Beijing are decreased by 5.1 % in the afternoon due to interactions between local and non-Beijing emissions. The non-Beijing emissions play a dominant role in the PM_{2.5} level in Beijing, with a contribution of 61.5 %, much higher than 13.7 %, from Beijing local emissions. The emission interactions between local and non-Beijing emissions enhance the PM_{2.5} concentrations in Beijing, with a contribution of 5.9 %. Therefore, the air quality in Beijing is generally determined by the trans-boundary transport of non-Beijing emissions during summertime, showing that the cooperation with neighboring provinces to mitigate pollutant emissions is key for Beijing to improve air quality.

60 years of UK visibility measurements: impact of meteorology and atmospheric pollutants on visibility

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Source: *Atmos. Chem. Phys.*, 17, 2085-2101, 2017

<http://www.atmos-chem-phys.net/17/2085/2017/doi:10.5194/acp-17-2085-2017>

Reduced visibility is an indicator of poor air quality. Moreover, degradation in visibility can be hazardous to human safety; for example, low visibility can lead to road, rail, sea and air accidents. In this paper, we explore the combined influence of atmospheric aerosol particle and gas characteristics, and meteorology, on long-term visibility. We use visibility data from eight meteorological stations, situated in the UK, which have been running since the 1950s. The site locations include urban, rural and marine environments.

Most stations show a long-term trend of increasing visibility, which is indicative of reductions in air pollution, especially in urban areas. Additionally, the visibility at all sites shows a very clear dependence on relative humidity, indicating the importance of aerosol hygroscopicity on the ability of aerosol particles to scatter radiation. The dependence of visibility on other meteorological

parameters, such as wind speed and wind direction, is also investigated. Most stations show long-term increases in temperature which can be ascribed to climate change, land-use changes (e.g. urban heat island effects) or a combination of both; the observed effect is greatest in urban areas. The impact of this temperature change upon local relative humidity is discussed.

To explain the long-term visibility trends and their dependence on meteorological conditions, the measured data were fitted to a newly developed light-extinction model to generate predictions of historic aerosol and gas scattering and absorbing properties. In general, an excellent fit was achieved between measured and modelled visibility for all eight sites. The model incorporates parameterizations of aerosol hygroscopicity, particle concentration, particle scattering, and particle and gas absorption. This new model should be applicable and is easily transferrable to other data sets worldwide. Hence, historical visibility data can be used to assess trends in aerosol particle properties. This approach may help constrain global model simulations which attempt to generate aerosol fields for time periods when observational data are scarce or non-existent. Both the measured visibility and the modelled aerosol properties reported in this paper highlight the success of the UK's Clean Air Act, which was passed in 1956, in cleaning the atmosphere of visibility-reducing pollutants.

Nitrate radicals and biogenic volatile organic compounds: oxidation, mechanisms, and organic aerosol

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Source: *Atmos. Chem. Phys.*, 17, 2103-2162, 2017

<http://www.atmos-chem-phys.net/17/2103/2017/doi:10.5194/acp-17-2103-2017>

Oxidation of biogenic volatile organic compounds (BVOC) by the nitrate radical (NO₃) represents one of the important interactions between anthropogenic emissions related to combustion and natural emissions from the biosphere. This interaction has been recognized for more than 3 decades, during which time a large body of research has emerged from laboratory, field, and modeling studies. NO₃-BVOC reactions influence air quality, climate and visibility through regional and global budgets for reactive nitrogen (particularly organic nitrates), ozone, and organic aerosol. Despite its long history of research and the significance of this topic in atmospheric chemistry, a number of important uncertainties remain. These include an incomplete understanding of the rates, mechanisms, and organic aerosol yields for NO₃-BVOC reactions, lack of constraints on the role of heterogeneous oxidative processes associated with the NO₃ radical, the difficulty of characterizing the spatial distributions of BVOC and NO₃ within the poorly mixed nocturnal atmosphere, and the challenge of constructing appropriate boundary layer schemes and non-photochemical mechanisms for use in state-of-the-art chemical transport and chemistry-climate models.

This review is the result of a workshop of the same title held at the Georgia Institute of Technology

in June 2015. The first half of the review summarizes the current literature on NO₃-BVOC chemistry, with a particular focus on recent advances in instrumentation and models, and in organic nitrate and secondary organic aerosol (SOA) formation chemistry. Building on this current understanding, the second half of the review outlines impacts of NO₃-BVOC chemistry on air quality and climate, and suggests critical research needs to better constrain this interaction to improve the predictive capabilities of atmospheric models.

Impact of a moderate volcanic eruption on chemistry in the lower stratosphere: balloon-borne observations and model calculations

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Source: Atmos. Chem. Phys., 17, 2229-2253, 2017

<http://www.atmos-chem-phys.net/17/2229/2017/doi:10.5194/acp-17-2229-2017>

The major volcanic eruption of Mount Pinatubo in 1991 has been shown to have significant effects on stratospheric chemistry and ozone depletion even at midlatitudes. Since then, only moderate but recurrent volcanic eruptions have modulated the stratospheric aerosol loading and are assumed to be one cause for the reported increase in the global aerosol content over the past 15 years. This particularly enhanced aerosol context raises questions about the effects on stratospheric chemistry which depend on the latitude, altitude and season of injection. In this study, we focus on the midlatitude Sarychev volcano eruption in June 2009, which injected 0.9 Tg of sulfur dioxide (about 20 times less than Pinatubo) into a lower stratosphere mainly governed by high-stratospheric temperatures. Together with in situ measurements of aerosol amounts, we analyse high-resolution in situ and/or remote-sensing observations of NO₂, HNO₃ and BrO from balloon-borne infrared and UV-visible spectrometers launched in Sweden in August–September 2009. It is shown that differences between observations and three-dimensional (3-D) chemistry-transport model (CTM) outputs are not due to transport calculation issues but rather reflect the chemical impact of the volcanic plume below 19 km altitude. Good measurement–model agreement is obtained when the CTM is driven by volcanic aerosol loadings derived from in situ or space-borne data. As a result of enhanced N₂O₅ hydrolysis in the Sarychev volcanic aerosol conditions, the model calculates reductions of ~ 45 % and increases of ~ 11 % in NO₂ and HNO₃ amounts respectively over the August–September 2009 period. The decrease in NO_x abundances is limited due to the expected saturation effect for high aerosol loadings. The links between the various chemical catalytic cycles involving chlorine, bromine, nitrogen and HO_x compounds in the lower stratosphere are discussed. The increased BrO amounts (~ 22 %) compare rather well with the balloon-borne observations when volcanic aerosol levels are accounted for in the CTM and appear to be mainly controlled by the coupling with nitrogen chemistry rather than by enhanced BrONO₂ hydrolysis. We show that the chlorine partitioning is significantly controlled by enhanced BrONO₂ hydrolysis. However, simulated effects of the Sarychev eruption on chlorine activation are very limited in the high-temperature conditions in the stratosphere in the period considered, inhibiting the effect of ClONO₂ hydrolysis. As a consequence, the simulated chemical ozone loss due to the Sarychev aerosols is low with a reduction of –22 ppbv (–1.5 %) of the ozone budget around 16 km. This is at

least 10 times lower than the maximum ozone depletion from chemical processes (up to -20 %) reported in the Northern Hemisphere lower stratosphere over the first year following the Pinatubo eruption. This study suggests that moderate volcanic eruptions have limited chemical effects when occurring at midlatitudes (restricted residence times) and outside winter periods (high-temperature conditions). However, it would be of interest to investigate longer-lasting tropical volcanic plumes or sulfur injections in the wintertime low-temperature conditions.

The recent increase of atmospheric methane from 10 years of ground-based NDACC FTIR observations since 2005

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Source: *Atmos. Chem. Phys.*, 17, 2255-2277, 2017

<http://www.atmos-chem-phys.net/17/2255/2017/doi:10.5194/acp-17-2255-2017>

Changes of atmospheric methane total columns (CH_4) since 2005 have been evaluated using Fourier transform infrared (FTIR) solar observations carried out at 10 ground-based sites, affiliated to the Network for Detection of Atmospheric Composition Change (NDACC). From this, we find an increase of atmospheric methane total columns of $0.31 \pm 0.03 \text{ \% year}^{-1}$ (2σ level of uncertainty) for the 2005–2014 period. Comparisons with in situ methane measurements at both local and global scales show good agreement. We used the GEOS-Chem chemical transport model tagged simulation, which accounts for the contribution of each emission source and one sink in the total methane, simulated over 2005–2012. After regridding according to NDACC vertical layering using a conservative regridding scheme and smoothing by convolving with respective FTIR seasonal averaging kernels, the GEOS-Chem simulation shows an increase of atmospheric methane total columns of $0.35 \pm 0.03 \text{ \% year}^{-1}$ between 2005 and 2012, which is in agreement with NDACC measurements over the same time period ($0.30 \pm 0.04 \text{ \% year}^{-1}$, averaged over 10 stations). Analysis of the GEOS-Chem-tagged simulation allows us to quantify the contribution of each tracer to the global methane change since 2005. We find that natural sources such as wetlands and biomass burning contribute to the interannual variability of methane. However, anthropogenic emissions, such as coal mining, and gas and oil transport and exploration, which are mainly emitted in the Northern Hemisphere and act as secondary contributors to the global budget of methane, have played a major role in the increase of atmospheric methane observed since 2005. Based on the GEOS-Chem-tagged simulation, we discuss possible cause(s) for the increase of methane since 2005, which is still unexplained.

Emission, transport, and radiative effects of mineral dust from the Taklimakan and Gobi deserts: comparison of measurements and model results

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Source: *Atmos. Chem. Phys.*, 17, 2401-2421, 2017

<http://www.atmos-chem-phys.net/17/2401/2017/doi:10.5194/acp-17-2401-2017>

The Weather Research and Forecasting Model with chemistry (WRF-Chem model) was used to investigate a typical dust storm event that occurred from 18 to 23 March 2010 and swept across almost all of China, Japan, and Korea. The spatial and temporal variations in dust aerosols and the meteorological conditions over East Asia were well reproduced by the WRF-Chem model. The simulation results were used to further investigate the details of processes related to dust emission, long-range transport, and radiative effects of dust aerosols over the Taklimakan Desert (TD) and Gobi Desert (GD). The results indicated that weather conditions, topography, and surface types in dust source regions may influence dust emission, uplift height, and transport at the regional scale. The GD was located in the warm zone in advance of the cold front in this case. Rapidly warming surface temperatures and cold air advection at high levels caused strong instability in the atmosphere, which strengthened the downward momentum transported from the middle and low troposphere and caused strong surface winds. Moreover, the GD is located in a relatively flat, high-altitude region influenced by the confluence of the northern and southern westerly jets. Therefore, the GD dust particles were easily lofted to 4 km and were the primary contributor to the dust concentration over East Asia. In the dust budget analysis, the dust emission flux over the TD was $27.2 \pm 4.1 \mu\text{g m}^{-2} \text{s}^{-1}$, which was similar to that over the GD ($29 \pm 3.6 \mu\text{g m}^{-2} \text{s}^{-1}$). However, the transport contribution of the TD dust (up to 0.8 ton d^{-1}) to the dust sink was much smaller than that of the GD dust (up to 3.7 ton d^{-1}) because of the complex terrain and the prevailing wind in the TD. Notably, a small amount of the TD dust (PM_{2.5} dust concentration of approximately $8.7 \mu\text{g m}^{-3}$) was lofted to above 5 km and transported over greater distances under the influence of the westerly jets. Moreover, the direct radiative forcing induced by dust was estimated to be -3 and -7 W m^{-2} at the top of the atmosphere, -8 and -10 W m^{-2} at the surface, and $+5$ and $+3 \text{ W m}^{-2}$ in the atmosphere over the TD and GD, respectively. This study provides confidence for further understanding the climate effects of the GD dust.

Influence of rain on the abundance of bioaerosols in fine and coarse particles

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Source: *Atmos. Chem. Phys.*, 17, 2459-2475, 2017

<http://www.atmos-chem-phys.net/17/2459/2017/doi:10.5194/acp-17-2459-2017>

Assessing the environmental, health, and climate impacts of bioaerosols requires knowledge of their size and abundance. These two properties were assessed through daily measurements of

chemical tracers for pollens (sucrose, fructose, and glucose), fungal spores (mannitol and glucans), and Gram-negative bacterial endotoxins in two particulate matter (PM) size modes: fine particles ($< 2.5 \mu\text{m}$) and coarse particles ($2.5\text{--}10 \mu\text{m}$) as determined by their aerodynamic diameter. Measurements were made during the spring tree pollen season (mid-April to early May) and late summer ragweed season (late August to early September) in the Midwestern US in 2013. Under dry conditions, pollen, and fungal spore tracers were primarily in coarse PM ($> 75 \%$), as expected for particles greater than $2.5 \mu\text{m}$. Rainfall on 2 May corresponded to maximum atmospheric pollen tracer levels and a redistribution of pollen tracers to the fine PM fraction ($> 80 \%$). Both changes were attributed to the osmotic rupture of pollen grains that led to the suspension of fine-sized pollen fragments. Fungal spore tracers peaked in concentration following spring rain events and decreased in particle size, but to a lesser extent than pollens. A short, heavy thunderstorm in late summer corresponded to an increase in endotoxin and glucose levels, with a simultaneous shift to smaller particle sizes. Simultaneous increase in bioaerosol levels and decrease in their size have significant implications for population exposures to bioaerosols, particularly during rain events. Chemical mass balance (CMB) source apportionment modeling and regionally specific pollen profiles were used to apportion PM mass to pollens and fungal spores. Springtime pollen contributions to the mass of particles $< 10 \mu\text{m}$ (PM₁₀) ranged from 0.04 to $0.8 \mu\text{g m}^{-3}$ ($0.2\text{--}38 \%$, averaging 4%), with maxima occurring on rainy days. Fungal spore contributions to PM₁₀ mass ranged from 0.1 to $1.5 \mu\text{g m}^{-3}$ ($0.8\text{--}17 \%$, averaging 5%), with maxima occurring after rain. Overall, this study defines changes to the fine- and coarse-mode distribution of PM, pollens, fungal spores, and endotoxins in response to rain in the Midwestern United States and advances the ability to apportion PM mass to pollens.

Regional influence of wildfires on aerosol chemistry in the western US and insights into atmospheric aging of biomass burning organic aerosol

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Source: *Atmos. Chem. Phys.*, 17, 2477-2493, 2017

<http://www.atmos-chem-phys.net/17/2477/2017/doi:10.5194/acp-17-2477-2017>

Biomass burning (BB) is one of the most important contributors to atmospheric aerosols on a global scale, and wildfires are a large source of emissions that impact regional air quality and global climate. As part of the Biomass Burning Observation Project (BBOP) field campaign in summer 2013, we deployed a high-resolution time-of-flight aerosol mass spectrometer (HR-AMS) coupled with a thermodenuder at the Mt. Bachelor Observatory (MBO, $\sim 2.8 \text{ km}$ above sea level) to characterize the impact of wildfire emissions on aerosol loading and properties in the Pacific Northwest region of the United States. MBO represents a remote background site in the western US, and it is frequently influenced by transported wildfire plumes during summer. Very clean conditions were observed at this site during periods without BB influence where the 5 min average ($\pm 1\sigma$) concentration of non-refractory submicron aerosols (NR-PM₁) was $3.7 \pm 4.2 \mu\text{g m}^{-3}$. Aerosol concentration increased substantially (reaching up to $210 \mu\text{g m}^{-3}$ of NR-PM₁) for periods impacted by transported BB plumes, and aerosol composition was overwhelmingly organic. Based on positive matrix factorization (PMF) of the HR-AMS data, three types of BB organic aerosol (BBOA) were identified, including a fresh, semivolatile BBOA-1 (O/C = 0.35; 20% of OA mass) that correlated well with ammonium nitrate; an intermediately oxidized BBOA-2 (O/C = 0.60; 17% of OA mass);

and a highly oxidized BBOA-3 (O/C = 1.06; 31 % of OA mass) that showed very low volatility with only ~ 40 % mass loss at 200 °C. The remaining 32 % of the OA mass was attributed to a boundary layer (BL) oxygenated OA (BL-OOA; O/C = 0.69) representing OA influenced by BL dynamics and a low-volatility oxygenated OA (LV-OOA; O/C = 1.09) representing regional aerosols in the free troposphere. The mass spectrum of BBOA-3 resembled that of LV-OOA and had negligible contributions from the HR-AMS BB tracer ions – C₂H₄O₂⁺ (m/z = 60.021) and C₃H₅O₂⁺ (m/z = 73.029); nevertheless, it was unambiguously related to wildfire emissions. This finding highlights the possibility that the influence of BB emission could be underestimated in regional air masses where highly oxidized BBOA (e.g., BBOA-3) might be a significant aerosol component but where primary BBOA tracers, such as levoglucosan, are depleted. We also examined OA chemical evolution for persistent BB plume events originating from a single fire source and found that longer solar radiation led to higher mass fraction of the chemically aged BBOA-2 and BBOA-3 and more oxidized aerosol. However, an analysis of the enhancement ratios of OA relative to CO ($\Delta\text{OA}/\Delta\text{CO}$) showed little difference between BB plumes transported primarily at night versus during the day, despite evidence of substantial chemical transformation in OA induced by photooxidation. These results indicate negligible net OA production in photochemically aged wildfire plumes observed in this study, for which a possible reason is that SOA formation was almost entirely balanced by BBOA volatilization. Nevertheless, the formation and chemical transformation of BBOA during atmospheric transport can significantly influence downwind sites with important implications for health and climate.

Widespread and persistent ozone pollution in eastern China during the non-winter season of 2015: observations and source attributions

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Source: *Atmos. Chem. Phys.*, 17, 2759-2774, 2017

<http://www.atmos-chem-phys.net/17/2759/2017/doi:10.5194/acp-17-2759-2017>

Rapid growth of industrialization, transportation, and urbanization has caused increasing emissions of ozone (O₃) precursors recently, enhancing the O₃ formation in eastern China. We show here that eastern China has experienced widespread and persistent O₃ pollution from April to September 2015 based on the O₃ observations in 223 cities. The observed maximum 1 h O₃ concentrations exceed 200 µg m⁻³ in almost all the cities, 400 µg m⁻³ in more than 25 % of the cities, and even 800 µg m⁻³ in six cities in eastern China. The average daily maximum 1 h O₃ concentrations are more than 160 µg m⁻³ in 45 % of the cities, and the 1 h O₃ concentrations of 200 µg m⁻³ have been exceeded on over 10 % of days from April to September in 129 cities. Analyses of pollutant observations from 2013 to 2015 have shown that the concentrations of CO, SO₂, NO₂, and PM_{2.5} from April to September in eastern China have considerably decreased, but the O₃ concentrations have increased by 9.9 %. A widespread and severe O₃ pollution episode from 22 to 28 May 2015 in eastern China has been simulated using the Weather Research and Forecasting model coupled to chemistry (WRF-CHEM) to evaluate the O₃ contribution of biogenic and various anthropogenic sources. The model generally performs reasonably well in simulating the temporal variations and spatial distributions of near-surface O₃ concentrations. Using the factor separation approach, sensitivity studies have indicated that the industry source plays the

most important role in the O₃ formation and constitutes the culprit of the severe O₃ pollution in eastern China. The transportation source contributes considerably to the O₃ formation, and the O₃ contribution of the residential source is not significant generally. The biogenic source provides a background O₃ source, and also plays an important role in the south of eastern China. Further model studies are needed to comprehensively investigate O₃ formation for supporting the design and implementation of O₃ control strategies, considering rapid changes of emission inventories and photolysis caused by the Atmospheric Pollution Prevention and Control Action Plan released by the Chinese State Council in 2013.

Global emissions of fluorinated greenhouse gases 2005–2050 with abatement potentials and costs

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Source: *Atmos. Chem. Phys.*, 17, 2795–2816, 2017

<http://www.atmos-chem-phys.net/17/2795/2017/doi:10.5194/acp-17-2795-2017>

This study uses the GAINS model framework to estimate current and future emissions of fluorinated greenhouse gases (F-gases), their abatement potentials, and costs for twenty source sectors and 162 countries and regions, which are aggregated to produce global estimates. Global F-gas (HFCs, PFCs, and SF₆) emissions are estimated at 0.7 Pg CO₂ eq. in 2005 with an expected increase to 3.7 Pg CO₂ eq. in 2050 if application of control technology remains at the current level. There are extensive opportunities to reduce emissions using existing technology and alternative substances with low global warming potential. Estimates show that it would be technically feasible to reduce cumulative F-gas emissions from 81 to 11 Pg CO₂ eq. between 2018 and 2050. A reduction in cumulative emissions to 23 Pg CO₂ eq. is estimated to be possible at a marginal abatement cost below 10 EUR t⁻¹ CO₂ eq. We also find that future F-gas abatement is expected to become relatively more costly for developing than developed countries due to differences in the sector contribution to emissions and abatement potentials.

A comprehensive biomass burning emission inventory with high spatial and temporal resolution in China

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Source: *Atmos. Chem. Phys.*, 17, 2839–2864, 2017

<http://www.atmos-chem-phys.net/17/2839/2017/doi:10.5194/acp-17-2839-2017>

Biomass burning injects many different gases and aerosols into the atmosphere that could have a harmful effect on air quality, climate, and human health. In this study, a comprehensive biomass burning emission inventory including domestic and in-field straw burning, firewood burning, livestock excrement burning, and forest and grassland fires is presented, which was developed for mainland China in 2012 based on county-level activity data, satellite data, and updated source-

specific emission factors (EFs). The emission inventory within a $1 \times 1 \text{ km}^2$ grid was generated using geographical information system (GIS) technology according to source-based spatial surrogates. A range of key information related to emission estimation (e.g. province-specific proportion of domestic and in-field straw burning, detailed firewood burning quantities, uneven temporal distribution coefficient) was obtained from field investigation, systematic combing of the latest research, and regression analysis of statistical data. The established emission inventory includes the major precursors of complex pollution, greenhouse gases, and heavy metal released from biomass burning. The results show that the emissions of SO_2 , NO_x , PM_{10} , $\text{PM}_{2.5}$, NMVOC, NH_3 , CO, EC, OC, CO_2 , CH_4 , and Hg in 2012 are 336.8 Gg, 990.7 Gg, 3728.3 Gg, 3526.7 Gg, 3474.2 Gg, 401.2 Gg, 34 380.4 Gg, 369.7 Gg, 1189.5 Gg, 675 299.0 Gg, 2092.4 Gg, and 4.12 Mg, respectively. Domestic straw burning, in-field straw burning, and firewood burning are identified as the dominant biomass burning sources. The largest contributing source is different for various pollutants. Domestic straw burning is the largest source of biomass burning emissions for all the pollutants considered, except for NH_3 , EC (firewood), and NO_x (in-field straw). Corn, rice, and wheat represent the major crop straws. The combined emission of these three straw types accounts for 80 % of the total straw-burned emissions for each specific pollutant mentioned in this study. As for the straw burning emission of various crops, corn straw burning has the largest contribution to all of the pollutants considered, except for CH_4 ; rice straw burning has highest contribution to CH_4 and the second largest contribution to other pollutants, except for SO_2 , OC, and Hg; wheat straw burning is the second largest contributor to SO_2 , OC, and Hg and the third largest contributor to other pollutants. Heilongjiang, Shandong, and Henan provinces located in the north-eastern and central-southern regions of China have higher emissions compared to other provinces in China. Gridded emissions, which were obtained through spatial allocation based on the gridded rural population and fire point data from emission inventories at county resolution, could better represent the actual situation. High biomass burning emissions are concentrated in the areas with more agricultural and rural activity. The months of April, May, June, and October account for 65 % of emissions from in-field crop residue burning, while, regarding EC, the emissions in January, February, October, November, and December are relatively higher than other months due to biomass domestic burning in heating season. There are regional differences in the monthly variations of emissions due to the diversity of main planted crops and climatic conditions. Furthermore, $\text{PM}_{2.5}$ component results showed that OC, Cl⁻, EC, K⁺, NH_4^+ , elemental K, and SO_4^{2-} are the main $\text{PM}_{2.5}$ species, accounting for 80 % of the total emissions. The species with relatively high contribution to NMVOC emission include ethylene, propylene, toluene, m,p-xylene, and ethyl benzene, which are key species for the formation of secondary air pollution. The detailed biomass burning emission inventory developed by this study could provide useful information for air-quality modelling and could support the development of appropriate pollution-control strategies.

Improving volcanic ash predictions with the HYSPLIT dispersion model by assimilating MODIS satellite retrievals

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Source: *Atmos. Chem. Phys.*, 17, 2865-2879, 2017

<http://www.atmos-chem-phys.net/17/2865/2017/doi:10.5194/acp-17-2865-2017>

Currently, the National Oceanic and Atmospheric Administration (NOAA) National Weather Service (NWS) runs the HYSPLIT dispersion model with a unit mass release rate to predict the transport and dispersion of volcanic ash. The model predictions provide information for the Volcanic Ash Advisory Centers (VAAC) to issue advisories to meteorological watch offices, area control centers, flight information centers, and others. This research aims to provide quantitative forecasts of ash distributions generated by objectively and optimally estimating the volcanic ash source strengths, vertical distribution, and temporal variations using an observation-modeling inversion technique. In this top-down approach, a cost functional is defined to quantify the differences between the model predictions and the satellite measurements of column-integrated ash concentrations weighted by the model and observation uncertainties. Minimizing this cost functional by adjusting the sources provides the volcanic ash emission estimates. As an example, MODIS (Moderate Resolution Imaging Spectroradiometer) satellite retrievals of the 2008 Kasatochi volcanic ash clouds are used to test the HYSPLIT volcanic ash inverse system. Because the satellite retrievals include the ash cloud top height but not the bottom height, there are different model diagnostic choices for comparing the model results with the observed mass loadings. Three options are presented and tested. Although the emission estimates vary significantly with different options, the subsequent model predictions with the different release estimates all show decent skill when evaluated against the unassimilated satellite observations at later times. Among the three options, integrating over three model layers yields slightly better results than integrating from the surface up to the observed volcanic ash cloud top or using a single model layer. Inverse tests also show that including the ash-free region to constrain the model is not beneficial for the current case. In addition, extra constraints on the source terms can be given by explicitly enforcing no-ash for the atmosphere columns above or below the observed ash cloud top height. However, in this case such extra constraints are not helpful for the inverse modeling. It is also found that simultaneously assimilating observations at different times produces better hindcasts than only assimilating the most recent observations.

Two global data sets of daily fire emission injection heights since 2003

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Source: *Atmos. Chem. Phys.*, 17, 2921-2942, 2017

<http://www.atmos-chem-phys.net/17/2921/2017/doi:10.5194/acp-17-2921-2017>

The Global Fire Assimilation System (GFAS) assimilates fire radiative power (FRP) observations from satellite-based sensors to produce daily estimates of biomass burning emissions. It has been extended to include information about injection heights derived from fire observations and meteorological information from the operational weather forecasts of ECMWF.

Injection heights are provided by two distinct methods: the Integrated Monitoring and Modelling System for wildland fires (IS4FIRES) parameterisation and the one-dimensional plume rise model (PRM). A global database of daily biomass burning emissions and injection heights at 0.1° resolution has been produced for 2003–2015 and is continuously extended in near-real time with the operational GFAS service of the Copernicus Atmospheric Monitoring Service (CAMS).

In this study, the two injection height data sets were compared with the new MHP2 (MISR Plume Height Project 2) satellite-based plume height retrievals. The IS4FIRES parameterisation showed a better overall agreement than the observations, while the PRM was better at capturing the variability of injection heights. The performance of both parameterisations is also dependent on the type of vegetation.

Furthermore, the use of biomass burning emission heights from GFAS in atmospheric composition forecasts was assessed in two case studies: the South American Biomass Burning Analysis (SAMBBA) campaign which took place in September 2012 in Brazil, and a series of large fire events in the western USA in August 2013. For these case studies, forecasts of biomass burning aerosol species by the Composition Integrated Forecasting System (C-IFS) of CAMS were found to better reproduce the observed vertical distribution when using PRM injection heights from GFAS compared to aerosols emissions being prescribed at the surface.

The globally available GFAS injection heights introduced and evaluated in this study provide a comprehensive data set for future fire and atmospheric composition modelling studies.

US surface ozone trends and extremes from 1980 to 2014: quantifying the roles of rising Asian emissions, domestic controls, wildfires, and climate

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Source: Atmos. Chem. Phys., 17, 2943-2970, 2017

<http://www.atmos-chem-phys.net/17/2943/2017/doi:10.5194/acp-17-2943-2017>

US surface O₃ responds to varying global-to-regional precursor emissions, climate, and extreme weather, with implications for designing effective air quality control policies. We examine these conjoined processes with observations and global chemistry-climate model (GFDL-AM3) hindcasts over 1980–2014. The model captures the salient features of observed trends in daily maximum 8 h average O₃: (1) increases over East Asia (up to 2 ppb yr⁻¹), (2) springtime increases at western US (WUS) rural sites (0.2–0.5 ppb yr⁻¹) with a baseline sampling approach, and (3) summertime decreases, largest at the 95th percentile, and wintertime increases in the 50th to 5th percentiles over the eastern US (EUS). Asian NO_x emissions have tripled since 1990, contributing as much as 65 % to modeled springtime background O₃ increases (0.3–0.5 ppb yr⁻¹) over the WUS, outpacing O₃ decreases attained via 50 % US NO_x emission controls. Methane increases over this period contribute only 15 % of the WUS background O₃ increase. Springtime O₃ observed in Denver has increased at a rate similar to remote rural sites. During summer, increasing Asian emissions approximately offset the benefits of US emission reductions, leading to weak or insignificant observed O₃ trends at WUS rural sites. Mean springtime WUS O₃ is projected to increase by ~ 10 ppb from 2010 to 2030 under the RCP8.5 global change scenario. While historical wildfire emissions can enhance summertime monthly mean O₃ at individual sites by 2–8 ppb, high temperatures and the associated buildup of O₃ produced from regional anthropogenic emissions contribute most to elevating observed summertime O₃ throughout the USA. GFDL-AM3 captures the observed interannual variability of summertime EUS O₃. However, O₃ deposition sink to vegetation must be reduced by 35 % for the model to accurately simulate observed high-O₃ anomalies during the severe drought of 1988. Regional NO_x reductions alleviated the

O₃ buildup during the recent heat waves of 2011 and 2012 relative to earlier heat waves (e.g., 1988, 1999). The O₃ decreases driven by NO_x controls were more pronounced in the southeastern US, where the seasonal onset of biogenic isoprene emissions and NO_x-sensitive O₃ production occurs earlier than in the northeast. Without emission controls, the 95th percentile summertime O₃ in the EUS would have increased by 0.2–0.4 ppb yr⁻¹ over 1988–2014 due to more frequent hot extremes and rising biogenic isoprene emissions.

Attributions of meteorological and emission factors to the 2015 winter severe haze pollution episodes in China's Jing-Jin-Ji area

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Source: Atmos. Chem. Phys., 17, 2971–2980, 2017

<http://www.atmos-chem-phys.net/17/2971/2017/doi:10.5194/acp-17-2971-2017>

In the 2015 winter month of December, northern China witnessed the most severe air pollution phenomena since the 2013 winter haze events occurred. This triggered the first-ever red alert in the air pollution control history of Beijing, with an instantaneous fine particulate matter (PM_{2.5}) concentration over 1 mg m⁻³. Air quality observations reveal large temporal–spatial variations in PM_{2.5} concentrations over the Beijing–Tianjin–Hebei (Jing-Jin-Ji) area between 2014 and 2015. Compared to 2014, the PM_{2.5} concentrations over the area decreased significantly in all months except November and December of 2015, with an increase of 36 % in December. Analysis shows that the PM_{2.5} concentrations are significantly correlated with the local meteorological parameters in the Jing-Jin-Ji area such as the stable conditions, relative humidity (RH), and wind field. A comparison of two month simulations (December 2014 and 2015) with the same emission data was performed to explore and quantify the meteorological impacts on the PM_{2.5} over the Jing-Jin-Ji area. Observation and modeling results show that the worsening meteorological conditions are the main reasons behind this unusual increase of air pollutant concentrations and that the emission control measures taken during this period of time have contributed to mitigate the air pollution (~ 9 %) in the region. This work provides a scientific insight into the emission control measures vs. the meteorology impacts for the period.

Effects of ozone–vegetation coupling on surface ozone air quality via biogeochemical and meteorological feedbacks

Mehliyar Sadiq, Amos P. K. Tai, Danica Lombardozzi, and Maria Val Martin

Source: Atmos. Chem. Phys., 17, 3055–3066, 2017

<http://www.atmos-chem-phys.net/17/3055/2017/doi:10.5194/acp-17-3055-2017>

Tropospheric ozone is one of the most hazardous air pollutants as it harms both human health and plant productivity. Foliage uptake of ozone via dry deposition damages photosynthesis and causes

stomatal closure. These foliage changes could lead to a cascade of biogeochemical and biogeophysical effects that not only modulate the carbon cycle, regional hydrometeorology and climate, but also cause feedbacks onto surface ozone concentration itself. In this study, we implement a semi-empirical parameterization of ozone damage on vegetation in the Community Earth System Model to enable online ozone-vegetation coupling, so that for the first time ecosystem structure and ozone concentration can coevolve in fully coupled land-atmosphere simulations. With ozone-vegetation coupling, present-day surface ozone is simulated to be higher by up to 4–6 ppbv over Europe, North America and China. Reduced dry deposition velocity following ozone damage contributes to ~40–100 % of those increases, constituting a significant positive biogeochemical feedback on ozone air quality. Enhanced biogenic isoprene emission is found to contribute to most of the remaining increases, and is driven mainly by higher vegetation temperature that results from lower transpiration rate. This isoprene-driven pathway represents an indirect, positive meteorological feedback. The reduction in both dry deposition and transpiration is mostly associated with reduced stomatal conductance following ozone damage, whereas the modification of photosynthesis and further changes in ecosystem productivity are found to play a smaller role in contributing to the ozone-vegetation feedbacks. Our results highlight the need to consider two-way ozone-vegetation coupling in Earth system models to derive a more complete understanding and yield more reliable future predictions of ozone air quality.

Enhanced trans-Himalaya pollution transport to the Tibetan Plateau by cut-off low systems

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Source: *Atmos. Chem. Phys.*, 17, 3083–3095, 2017

<http://www.atmos-chem-phys.net/17/3083/2017/doi:10.5194/acp-17-3083-2017>

Long-range transport followed by deposition of black carbon on glaciers of Tibet is one of the key issues of climate research as it induces changes on radiative forcing and subsequently impacting the melting of glaciers. The transport mechanism, however, is not well understood. In this study, we use short-lived reactive aromatics as proxies to diagnose transport of pollutants to Tibet. In situ observations of short-lived reactive aromatics across the Tibetan Plateau are analyzed using a regional chemistry and transport model. The model performance using the current emission inventories over the region is poor due to problems in the inventories and model transport. Top-down emissions constrained by satellite observations of glyoxal are a factor of 2–6 higher than the a priori emissions over the industrialized Indo-Gangetic Plain. Using the top-down emissions, agreement between model simulations and surface observations of aromatics improves. We find enhancements of reactive aromatics over Tibet by a factor of 6 on average due to rapid transport from India and nearby regions during the presence of a high-altitude cut-off low system. Our results suggest that the cut-off low system is a major pathway for long-range transport of pollutants such as black carbon. The modeling analysis reveals that even the state-of-the-science high-resolution reanalysis cannot simulate this cut-off low system accurately, which probably explains in part the underestimation of black carbon deposition over Tibet in previous modeling studies. Another model deficiency of underestimating pollution transport from the south is due to the complexity of terrain, leading to enhanced transport. It is therefore challenging for coarse-resolution global

climate models to properly represent the effects of long-range transport of pollutants on the Tibetan environment and the subsequent consequence for regional climate forcing.

Classification of summertime synoptic patterns in Beijing and their associations with boundary layer structure affecting aerosol pollution

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Source: *Atmos. Chem. Phys.*, 17, 3097-3110, 2017

<http://www.atmos-chem-phys.net/17/3097/2017/doi:10.5194/acp-17-3097-2017>

Meteorological conditions within the planetary boundary layer (PBL) are closely governed by large-scale synoptic patterns and play important roles in air quality by directly and indirectly affecting the emission, transport, formation, and deposition of air pollutants. Partly due to the lack of long-term fine-resolution observations of the PBL, the relationships between synoptic patterns, PBL structure, and aerosol pollution in Beijing have not been well understood. This study applied the obliquely rotated principal component analysis in T-mode to classify the summertime synoptic conditions over Beijing using the National Centers for Environmental Prediction reanalysis from 2011 to 2014, and investigated their relationships with PBL structure and aerosol pollution by combining numerical simulations, measurements of surface meteorological variables, fine-resolution soundings, the concentration of particles with diameters less than or equal to 2.5 μm , total cloud cover (CLD), and reanalysis data. Among the seven identified synoptic patterns, three types accounted for 67 % of the total number of cases studied and were associated with heavy aerosol pollution events. These particular synoptic patterns were characterized by high-pressure systems located to the east or southeast of Beijing at the 925 hPa level, which blocked the air flow seaward, and southerly PBL winds that brought in polluted air from the southern industrial zone. The horizontal transport of pollutants induced by the synoptic forcings may be the most important factor affecting the air quality of Beijing in summer. In the vertical dimension, these three synoptic patterns featured a relatively low boundary layer height (BLH) in the afternoon, accompanied by high CLD and southerly cold advection from the seas within the PBL. The high CLD reduced the solar radiation reaching the surface, and suppressed the thermal turbulence, leading to lower BLH. Besides, the numerical sensitive experiments show that cold advection induced by the large-scale synoptic forcing may have cooled the PBL, leading to an increase in near-surface stability and a decrease in the BLH in the afternoon. Moreover, when warm advection appeared simultaneously above the top level of the PBL, the thermal inversion layer capping the PBL may have been strengthened, resulting in the further suppression of PBL and thus the deterioration of aerosol pollution levels. This study has important implications for understanding the crucial roles that meteorological factors (at both synoptic and local scales) play in modulating and forecasting aerosol pollution in Beijing and its surrounding area.

The contribution of wood burning and other pollution sources to wintertime organic aerosol levels in two Greek cities

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Source: *Atmos. Chem. Phys.*, 17, 3145-3163, 2017

<http://www.atmos-chem-phys.net/17/3145/2017/doi:10.5194/acp-17-3145-2017>

The composition of fine particulate matter (PM) in two major Greek cities (Athens and Patras) was measured during two wintertime campaigns, one conducted in 2013 and the other in 2012. A major goal of this study is to quantify the sources of organic aerosol (OA) and especially residential wood burning, which has dramatically increased due to the Greek financial crisis. A high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) was deployed at both sites. PM with diameter less than 1 μm (PM₁) consisted mainly of organics (60–75 %), black carbon (5–20 %), and inorganic salts (around 20 %) in both Patras and Athens. In Patras, during evening hours, PM₁ concentrations were as high as 100 $\mu\text{g m}^{-3}$, of which 85 % was OA. In Athens, the maximum hourly value observed during nighttime was 140 $\mu\text{g m}^{-3}$, of which 120 $\mu\text{g m}^{-3}$ was OA. Forty to 60 % of the average OA was due to biomass burning for both cities, while the remaining mass originated from traffic (12–17 %), cooking (12–16 %), and long-range transport (18–24 %). The contribution of residential wood burning was even higher (80–90 %) during the nighttime peak concentration periods, and less than 10 % during daytime. Cooking OA contributed up to 75 % during mealtime hours in Patras, while traffic-related OA was responsible for 60–70 % of the OA during the morning rush hour.

Resolving anthropogenic aerosol pollution types – deconvolution and exploratory classification of pollution events

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Source: *Atmos. Chem. Phys.*, 17, 3165-3197, 2017

<http://www.atmos-chem-phys.net/17/3165/2017/doi:10.5194/acp-17-3165-2017>

Mass spectrometric measurements commonly yield data on hundreds of variables over thousands of points in time. Refining and synthesizing this raw data into chemical information necessitates the use of advanced, statistics-based data analytical techniques. In the field of analytical aerosol chemistry, statistical, dimensionality reductive methods have become widespread in the last decade, yet comparable advanced chemometric techniques for data classification and identification remain marginal. Here we present an example of combining data dimensionality reduction (factorization) with exploratory classification (clustering), and show that the results cannot only reproduce and corroborate earlier findings, but also complement and broaden our current perspectives on aerosol chemical classification. We find that applying positive matrix factorization to extract spectral characteristics of the organic component of air pollution plumes, together with an unsupervised clustering algorithm, k-means+, for classification, reproduces classical organic

aerosol speciation schemes. Applying appropriately chosen metrics for spectral dissimilarity along with optimized data weighting, the source-specific pollution characteristics can be statistically resolved even for spectrally very similar aerosol types, such as different combustion-related anthropogenic aerosol species and atmospheric aerosols with similar degree of oxidation. In addition to the typical oxidation level and source-driven aerosol classification, we were also able to classify and characterize outlier groups that would likely be disregarded in a more conventional analysis. Evaluating solution quality for the classification also provides means to assess the performance of mass spectral similarity metrics and optimize weighting for mass spectral variables. This facilitates algorithm-based evaluation of aerosol spectra, which may prove invaluable for future development of automatic methods for spectra identification and classification. Robust, statistics-based results and data visualizations also provide important clues to a human analyst on the existence and chemical interpretation of data structures. Applying these methods to a test set of data, aerosol mass spectrometric data of organic aerosol from a boreal forest site, yielded five to seven different recurring pollution types from various sources, including traffic, cooking, biomass burning and nearby sawmills. Additionally, three distinct, minor pollution types were discovered and identified as amine-dominated aerosols.

Sources and formation mechanisms of carbonaceous aerosol at a regional background site in the Netherlands: insights from a year-long radiocarbon study

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Source: *Atmos. Chem. Phys.*, 17, 3233-3251, 2017

<http://www.atmos-chem-phys.net/17/3233/2017/doi:10.5194/acp-17-3233-2017>

We measured the radioactive carbon isotope ^{14}C (radiocarbon) in various fractions of the carbonaceous aerosol sampled between February 2011 and March 2012 at the Cesar Observatory in the Netherlands. Based on the radiocarbon content in total carbon (TC), organic carbon (OC), water-insoluble organic carbon (WIOC), and elemental carbon (EC), we estimated the contribution of major sources to the carbonaceous aerosol. The main source categories were fossil fuel combustion, biomass burning, and other contemporary carbon, which is mainly biogenic secondary organic aerosol material (SOA).

A clear seasonal variation is seen in EC from biomass burning (EC_{bb}), with lowest values in summer and highest values in winter, but EC_{bb} is a minor fraction of EC in all seasons. WIOC from contemporary sources is highly correlated with EC_{bb}, indicating that biomass burning is a dominant source of contemporary WIOC. This suggests that most biogenic SOA is water soluble and that water-insoluble carbon stems mainly from primary sources. Seasonal variations in other carbon fractions are less clear and hardly distinguishable from variations related to air mass history.

Air masses originating from the ocean sector presumably contain little carbonaceous aerosol from outside the Netherlands, and during these conditions measured carbon concentrations reflect regional sources. In these situations absolute TC concentrations are usually rather low, around $1.5\text{ }\mu\text{g m}^{-3}$, and EC_{bb} is always very low ($\sim 0.05\text{ }\mu\text{g m}^{-3}$), even in winter, indicating that biomass

burning is not a strong source of carbonaceous aerosol in the Netherlands. In continental air masses, which usually arrive from the east or south and have spent several days over land, TC concentrations are on average by a factor of 3.5 higher. EC_{bb} increases more strongly than TC to 0.2 µg m⁻³. Fossil EC and fossil WIOC, which are indicative of primary emissions, show a more moderate increase by a factor of 2.5 on average.

An interesting case is fossil water-soluble organic carbon (WSOC, calculated as OC-WIOC), which can be regarded as a proxy for SOA from fossil precursors. Fossil WSOC has low concentrations when regional sources are sampled and increases by more than a factor of 5 in continental air masses. A longer residence time of air masses over land seems to result in increased SOA concentrations from fossil origin.

Causes of interannual variability over the southern hemispheric tropospheric ozone maximum

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Source: Atmos. Chem. Phys., 17, 3279-3299, 2017
<https://doi.org/10.5194/acp-17-3279-2017>

We examine the relative contribution of processes controlling the interannual variability (IAV) of tropospheric ozone over four sub-regions of the southern hemispheric tropospheric ozone maximum (SHTOM) over a 20-year period. Our study is based on hindcast simulations from the National Aeronautics and Space Administration Global Modeling Initiative chemistry transport model (NASA GMI-CTM) of tropospheric and stratospheric chemistry, driven by assimilated Modern Era Retrospective Analysis for Research and Applications (MERRA) meteorological fields. Our analysis shows that over SHTOM region, the IAV of the stratospheric contribution is the most important factor driving the IAV of upper tropospheric ozone (270 hPa), where ozone has a strong radiative effect. Over the South Atlantic region, the contribution from surface emissions to the IAV of ozone exceeds that from stratospheric input at and below 430 hPa. Over the South Indian Ocean, the IAV of stratospheric ozone makes the largest contribution to the IAV of ozone with little or no influence from surface emissions at 270 and 430 hPa in austral winter. Over the tropical South Atlantic region, the contribution from IAV of stratospheric input dominates in austral winter at 270 hPa and drops to less than half but is still significant at 430 hPa. Emission contributions are not significant at these two levels. The IAV of lightning over this region also contributes to the IAV of ozone in September and December. Over the tropical southeastern Pacific, the contribution of the IAV of stratospheric input is significant at 270 and 430 hPa in austral winter, and emissions have little influence.

Terpenoid and carbonyl emissions from Norway spruce in Finland during the growing season

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Source: Atmos. Chem. Phys., 17, 3357-3370, 2017
<https://doi.org/10.5194/acp-17-3357-2017>

We present spring and summer volatile organic compound (VOC) emission rate measurements from Norway spruce (*Picea abies* L. Karst) growing in a boreal forest in southern Finland. The measurements were conducted using in situ gas chromatograph with 1 to 2 h time resolution to reveal quantitative and qualitative short-term and seasonal variability of the emissions. The measurements cover altogether 14 weeks in years 2011, 2014 and 2015. Monoterpene (MT) and sesquiterpene (SQT) emission rates were measured all the time, but isoprene only in 2014 and 2015 and acetone and C4–C10 aldehydes only in 2015. The emission rates of all the compounds were low in spring, but MT, acetone, and C4–C10 aldehyde emission rates increased as summer proceeded, reaching maximum emission rates in July. Late summer mean values (late July and August) were 29, 17, and 33 ng g(dw)^{−1} h^{−1} for MTs, acetone, and aldehydes respectively. SQT emission rates increased during the summer and highest emissions were measured in late summer (late summer mean value 84 ng g(dw)^{−1} h^{−1}) concomitant with highest linalool emissions most likely due to stress effects. The between-tree variability of emission pattern was studied by measuring seven different trees during the same afternoon using adsorbent tubes. Especially the contributions of limonene, terpinolene, and camphene were found to vary between trees, whereas proportions of α -pinene ($25 \pm 5\%$) and β -pinene ($7 \pm 3\%$) were more stable. Our results show that it is important to measure emissions at canopy level due to irregular emission pattern, but reliable SQT emission data can be measured only from enclosures. SQT emissions contributed more than 90 % of the ozone reactivity most of the time, and about 70 % of the OH reactivity during late summer. The contribution of aldehydes to OH reactivity was comparable to that of MT during late summer, 10–30 % most of the time.

Interpreting the ¹³C / ¹²C ratio of carbon dioxide in an urban airshed in the Yangtze River Delta, China

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Source: Atmos. Chem. Phys., 17, 3385-3399, 2017
<https://doi.org/10.5194/acp-17-3385-2017>

Observations of atmospheric CO₂ mole fraction and the ¹³C/¹²C ratio (expressed as $\delta^{13}\text{C}$) in urban airsheds provide constraints on the roles of anthropogenic and natural sources and sinks in local and regional carbon cycles. In this study, we report observations of these quantities in Nanjing at hourly intervals from March 2013 to August 2015, using a laser-based optical instrument. Nanjing is the second largest city located in the highly industrialized Yangtze River Delta (YRD), eastern China. The mean CO₂ mole fraction and $\delta^{13}\text{C}$ were $(439.7 \pm 7.5) \mu\text{mol mol}^{-1}$ and

(-8.48 ± 0.56) ‰ over this observational period. The peak monthly mean $\delta^{13}\text{C}$ (-7.44 ‰, July 2013) was 0.74 ‰ higher than that observed at Mount Waliguan, a WMO (World Meteorological Organization) baseline site on the Tibetan Plateau and upwind of the YRD region. The highly ^{13}C -enriched signal was partly attributed to the influence of cement production in the region. By applying the Miller-Tans method to nighttime and daytime observations to represent signals from the city of Nanjing and the YRD, respectively, we showed that the $^{13}\text{C}/^{12}\text{C}$ ratio of CO_2 sources in the Nanjing municipality was (0.21 ± 0.53) ‰ lower than that in the YRD. Flux partitioning calculations revealed that natural ecosystems in the YRD were a negligibly small source of atmospheric CO_2 .

Bromine atom production and chain propagation during springtime Arctic ozone depletion events in Barrow, Alaska

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Source: Atmos. Chem. Phys., 17, 3401-3421, 2017
<https://doi.org/10.5194/acp-17-3401-2017>

Ozone depletion events (ODEs) in the Arctic are primarily controlled by a bromine radical-catalyzed destruction mechanism that depends on the efficient production and recycling of Br atoms. Numerous laboratory and modeling studies have suggested the importance of heterogeneous recycling of Br through HOBr reaction with bromide on saline surfaces. On the other hand, the gas-phase regeneration of bromine atoms through BrO–BrO radical reactions has been assumed to be an efficient, if not dominant, pathway for Br reformation and thus ozone destruction. Indeed, it has been estimated that the rate of ozone depletion is approximately equal to twice the rate of the BrO self-reaction. Here, we use a zero-dimensional, photochemical model, largely constrained to observations of stable atmospheric species from the 2009 Ocean–Atmosphere–Sea Ice–Snowpack (OASIS) campaign in Barrow, Alaska, to investigate gas-phase bromine radical propagation and recycling mechanisms of bromine atoms for a 7-day period during late March. This work is a continuation of that presented in Thompson et al. (2015) and utilizes the same model construct. Here, we use the gas-phase radical chain length as a metric for objectively quantifying the efficiency of gas-phase recycling of bromine atoms. The gas-phase bromine chain length is determined to be quite small, at < 1.5 , and highly dependent on ambient O_3 concentrations. Furthermore, we find that Br atom production from photolysis of Br_2 and BrCl , which is predominately emitted from snow and/or aerosol surfaces, can account for between 30 and 90 % of total Br atom production. This analysis suggests that condensed-phase production of bromine is at least as important as, and at times greater than, gas-phase recycling for the occurrence of Arctic ODEs. Therefore, the rate of the BrO self-reaction is not a sufficient estimate for the rate of O_3 depletion.

Evaluation of the impact of wood combustion on benzo[a]pyrene (BaP) concentrations; ambient measurements and dispersion modeling in Helsinki, Finland

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Source: Atmos. Chem. Phys., 17, 3475-3487, 2017
<https://doi.org/10.5194/acp-17-3475-2017>

Even though emission inventories indicate that wood combustion is a major source of polycyclic aromatic hydrocarbons (PAHs), estimating its impacts on PAH concentration in ambient air remains challenging. In this study the effect of local small-scale wood combustion on the benzo[a]pyrene (BaP) concentrations in ambient air in the Helsinki metropolitan area in Finland is evaluated, using ambient air measurements, emission estimates, and dispersion modeling. The measurements were conducted at 12 different locations during the period from 2007 to 2015. The spatial distributions of annual average BaP concentrations originating from wood combustion were predicted for four of those years: 2008, 2011, 2013, and 2014. According to both the measurements and the dispersion modeling, the European Union target value for the annual average BaP concentrations (1 ng m^{-3}) was clearly exceeded in certain suburban detached-house areas. However, in most of the other urban areas, including the center of Helsinki, the concentrations were below the target value. The measured BaP concentrations highly correlated with the measured levoglucosan concentrations in the suburban detached-house areas. In street canyons, the measured concentrations of BaP were at the same level as those in the urban background, clearly lower than those in suburban detached-house areas. The predicted annual average concentrations matched with the measured concentrations fairly well. Both the measurements and the modeling clearly indicated that wood combustion was the main local source of ambient air BaP in the Helsinki metropolitan area.

Black carbon variability since preindustrial times in the eastern part of Europe reconstructed from Mt. Elbrus, Caucasus, ice cores

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Black carbon (BC), emitted by fossil fuel combustion and biomass burning, is the second largest man-made contributor to global warming after carbon dioxide (Bond et al., 2013). However, limited information exists on its past emissions and atmospheric variability. In this study, we present the first high-resolution record of refractory BC (rBC, including mass concentration and size) reconstructed from ice cores drilled at a high-altitude eastern European site in Mt. Elbrus (ELB), Caucasus (5115 m a.s.l.). The ELB ice core record, covering the period 1825–2013, reflects the atmospheric load of rBC particles at the ELB site transported from the European continent with a larger rBC input from sources located in the eastern part of Europe. In the first half of the 20th century, European anthropogenic emissions resulted in a 1.5-fold increase in the ice core rBC mass concentrations with respect to its level in the preindustrial era (before 1850). The summer (winter) rBC mass concentrations increased 5-fold (3.3-fold) in 1960–1980, followed by a decrease until

~ 2000. Over the last decade, the rBC signal for summertime slightly increased. We have compared the signal with the atmospheric BC load simulated using past BC emissions (ACCMIP and MACCity inventories) and taken into account the contribution of different geographical regions to rBC distribution and deposition at the ELB site. Interestingly, the observed rBC variability in the ELB ice core record since the 1960s is not in perfect agreement with the simulated atmospheric BC load. Similar features between the ice core rBC record and the best scenarios for the atmospheric BC load support anthropogenic BC increase in the 20th century being reflected in the ELB ice core record. However, the peak in BC mass concentration observed in ~ 1970 in the ice core is estimated to occur a decade later from past inventories. BC emission inventories for the period 1960s–1970s may be underestimating European anthropogenic emissions. Furthermore, for summertime snow layers of the 2000s, the slightly increasing trend of rBC deposition likely reflects recent changes in anthropogenic and biomass burning BC emissions in the eastern part of Europe. Our study highlights that the past changes in BC emissions of eastern Europe need to be considered in assessing ongoing air quality regulation.

Methane fluxes in the high northern latitudes for 2005–2013 estimated using a Bayesian atmospheric inversion

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Source: Atmos. Chem. Phys., 17, 3553–3572, 2017
<https://doi.org/10.5194/acp-17-3553-2017>

We present methane (CH₄) flux estimates for 2005 to 2013 from a Bayesian inversion focusing on the high northern latitudes (north of 50° N). Our inversion is based on atmospheric transport modelled by the Lagrangian particle dispersion model FLEXPART and CH₄ observations from 17 in situ and five discrete flask-sampling sites distributed over northern North America and Eurasia. CH₄ fluxes are determined at monthly temporal resolution and on a variable grid with maximum resolution of 1° × 1°. Our inversion finds a CH₄ source from the high northern latitudes of 82 to 84 Tg yr⁻¹, constituting ~ 15 % of the global total, compared to 64 to 68 Tg yr⁻¹ (~ 12 %) in the prior estimates. For northern North America, we estimate a mean source of 16.6 to 17.9 Tg yr⁻¹, which is dominated by fluxes in the Hudson Bay Lowlands (HBL) and western Canada, specifically the province of Alberta. Our estimate for the HBL, of 2.7 to 3.4 Tg yr⁻¹, is close to the prior estimate (which includes wetland fluxes from the land surface model, LPX-Bern) and to other independent inversion estimates. However, our estimate for Alberta, of 5.0 to 5.8 Tg yr⁻¹, is significantly higher than the prior (which also includes anthropogenic sources from the EDGAR-4.2FT2010 inventory). Since the fluxes from this region persist throughout the winter, this may signify that the anthropogenic emissions are underestimated. For northern Eurasia, we find a mean source of 52.2 to 55.5 Tg yr⁻¹, with a strong contribution from fluxes in the Western Siberian Lowlands (WSL) for which we estimate a source of 19.3 to 19.9 Tg yr⁻¹. Over the 9-year inversion period, we find significant year-to-year variations in the fluxes, which in North America, and specifically in the HBL, appear to be driven at least in part by soil temperature, while in the WSL, the variability is more dependent on soil moisture. Moreover, we find significant positive trends in the CH₄ fluxes in North America of 0.38 to 0.57 Tg yr⁻², and northern Eurasia of 0.76 to 1.09 Tg yr⁻². In North America,

this could be due to an increase in soil temperature, while in North Eurasia, specifically Russia, the trend is likely due, at least in part, to an increase in anthropogenic sources.

Composition, size and cloud condensation nuclei activity of biomass burning aerosol from northern Australian savannah fires

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Source: Atmos. Chem. Phys., 17, 3605-3617, 2017
<https://doi.org/10.5194/acp-17-3605-2017>

The vast majority of Australia's fires occur in the tropical north of the continent during the dry season. These fires are a significant source of aerosol and cloud condensation nuclei (CCN) in the region, providing a unique opportunity to investigate the biomass burning aerosol (BBA) in the absence of other sources. CCN concentrations at 0.5 % supersaturation and aerosol size and chemical properties were measured at the Australian Tropical Atmospheric Research Station (ATARS) during June 2014. CCN concentrations reached over 10^4 cm^{-3} when frequent and close fires were burning – up to 45 times higher than periods with no fires. Both the size distribution and composition of BBA appeared to significantly influence CCN concentrations. A distinct diurnal trend in the proportion of BBA activating to cloud droplets was observed, with an activation ratio of $40 \pm 20 \%$ during the night and $60 \pm 20 \%$ during the day. BBA was, on average, less hygroscopic during the night ($\kappa = 0.04 \pm 0.03$) than during the day ($\kappa = 0.07 \pm 0.05$), with a maximum typically observed just before midday. Size-resolved composition of BBA showed that organics comprised a constant 90 % of the aerosol volume for aerodynamic diameters between 100 and 200 nm. While this suggests that the photochemical oxidation of organics led to an increase in the hygroscopic growth and an increase in daytime activation ratios, it does not explain the decrease in hygroscopicity after midday. Modelled CCN concentrations assuming typical continental hygroscopicities produced very large overestimations of up to 200 %. Smaller, but still significant, overpredictions up to $\sim 100 \%$ were observed using aerosol mass spectrometer (AMS)- and hygroscopicity tandem differential mobility analyser (H-TDMA)-derived hygroscopicities as well as campaign night and day averages. The largest estimations in every case occurred during the night, when the small variations in very weakly hygroscopic species corresponded to large variations in the activation diameters. Trade winds carry the smoke generated from these fires over the Timor Sea, where aerosol–cloud interactions are likely to be sensitive to changes in CCN concentrations, perturbing cloud albedo and lifetime. Dry season fires in northern Australia are therefore potentially very important in cloud processes in this region.

Seasonal variations of triple oxygen isotopic compositions of atmospheric sulfate, nitrate, and ozone at Dumont d'Urville, coastal Antarctica

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Source: Atmos. Chem. Phys., 17, 3713-3727, 2017
<https://doi.org/10.5194/acp-17-3713-2017>

Triple oxygen isotopic compositions ($\Delta 17\text{O} = \delta 17\text{O} - 0.52 \times \delta 18\text{O}$) of atmospheric sulfate (SO_4^{2-}) and nitrate (NO_3^-) in the atmosphere reflect the relative contribution of oxidation pathways involved in their formation processes, which potentially provides information to reveal missing reactions in atmospheric chemistry models. However, there remain many theoretical assumptions for the controlling factors of $\Delta 17\text{O}(\text{SO}_4^{2-})$ and $\Delta 17\text{O}(\text{NO}_3^-)$ values in those model estimations. To test one of those assumption that $\Delta 17\text{O}$ values of ozone (O_3) have a flat value and do not influence the seasonality of $\Delta 17\text{O}(\text{SO}_4^{2-})$ and $\Delta 17\text{O}(\text{NO}_3^-)$ values, we performed the first simultaneous measurement of $\Delta 17\text{O}$ values of atmospheric sulfate, nitrate, and ozone collected at Dumont d'Urville (DDU) Station ($66^\circ 40' \text{S}$, $140^\circ 01' \text{E}$) throughout 2011. $\Delta 17\text{O}$ values of sulfate and nitrate exhibited seasonal variation characterized by minima in the austral summer and maxima in winter, within the ranges of 0.9–3.4 and 23.0–41.9 ‰, respectively. In contrast, $\Delta 17\text{O}$ values of ozone showed no significant seasonal variation, with values of 26 ± 1 ‰ throughout the year. These contrasting seasonal trends suggest that seasonality in $\Delta 17\text{O}(\text{SO}_4^{2-})$ and $\Delta 17\text{O}(\text{NO}_3^-)$ values is not the result of changes in $\Delta 17\text{O}(\text{O}_3)$, but of the changes in oxidation chemistry. The trends with summer minima and winter maxima for $\Delta 17\text{O}(\text{SO}_4^{2-})$ and $\Delta 17\text{O}(\text{NO}_3^-)$ values are caused by sunlight-driven changes in the relative contribution of O_3 oxidation to the oxidation by HOx , ROx , and H_2O_2 . In addition to that general trend, by comparing $\Delta 17\text{O}(\text{SO}_4^{2-})$ and $\Delta 17\text{O}(\text{NO}_3^-)$ values to ozone mixing ratios, we found that $\Delta 17\text{O}(\text{SO}_4^{2-})$ values observed in spring (September to November) were lower than in fall (March to May), while there was no significant spring and fall difference in $\Delta 17\text{O}(\text{NO}_3^-)$ values. The relatively lower sensitivity of $\Delta 17\text{O}(\text{SO}_4^{2-})$ values to the ozone mixing ratio in spring compared to fall is possibly explained by (i) the increased contribution of SO_2 oxidations by OH and H_2O_2 caused by NO_x emission from snowpack and/or (ii) SO_2 oxidation by hypohalous acids ($\text{HOX} = \text{HOCl} + \text{HOBr}$) in the aqueous phase.

Interannual variation, decadal trend, and future change in ozone outflow from East Asia

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Source: Atmos. Chem. Phys., 17, 3729-3747, 2017
<https://doi.org/10.5194/acp-17-3729-2017>

We examine the past and future changes in the O_3 outflow from East Asia using a global 3-D chemical transport model, GEOS-Chem. The simulations of Asian O_3 outflow for 1986–2006 are driven by the assimilated GEOS-4 meteorological fields, and those for 2000–2050 are driven by the

meteorological fields archived by the NASA Goddard Institute for Space Studies (GISS) general circulation model (GCM) 3 under the IPCC SRES A1B scenario. The evaluation of the model results against measurements shows that the GEOS-Chem model captures the seasonal cycles and interannual variations of tropospheric O₃ concentrations fairly well with high correlation coefficients of 0.82–0.93 at four ground-based sites and 0.55–0.88 at two ozonesonde sites where observations are available. The increasing trends in surface-layer O₃ concentrations in East Asia over the past 2 decades are captured by the model, although the modeled O₃ trends have low biases. Sensitivity studies are conducted to examine the respective impacts of meteorological parameters and emissions on the variations in the outflow flux of O₃. When both meteorological parameters and anthropogenic emissions varied from 1986–2006, the simulated Asian O₃ outflow fluxes exhibited a statistically insignificant decadal trend; however, they showed large interannual variations (IAVs) with seasonal values of 4–9 % for the absolute percent departure from the mean (APDM) and an annual APDM value of 3.3 %. The sensitivity simulations indicated that the large IAVs in O₃ outflow fluxes were mainly caused by variations in the meteorological conditions. The variations in meteorological parameters drove the IAVs in O₃ outflow fluxes by altering the O₃ concentrations over East Asia and by altering the zonal winds; the latter was identified to be the key factor, since the O₃ outflow was highly correlated with zonal winds from 1986–2006. The simulations of the 2000–2050 changes show that the annual outflow flux of O₃ will increase by 2.0, 7.9, and 12.2 % owing to climate change alone, emissions change alone, and changes in both climate and emissions, respectively. Therefore, climate change will aggravate the effects of the increases in anthropogenic emissions on future changes in the Asian O₃ outflow. Future climate change is predicted to greatly increase the Asian O₃ outflow in the spring and summer seasons as a result of the projected increases in zonal winds. The findings from the present study help us to understand the variations in tropospheric O₃ in the downwind regions of East Asia on different timescales and have important implications for long-term air quality planning in the regions downwind of China, such as Japan and the US.

Sensitivity of transatlantic dust transport to chemical aging and related atmospheric processes

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Source: *Atmos. Chem. Phys.*, 17, 3799–3821, 2017
<https://doi.org/10.5194/acp-17-3799-2017>

We present a sensitivity study on transatlantic dust transport, a process which has many implications for the atmosphere, the ocean and the climate. We investigate the impact of key processes that control the dust outflow, i.e., the emission flux, convection schemes and the chemical aging of mineral dust, by using the EMAC model following Abdelkader et al. (2015). To characterize the dust outflow over the Atlantic Ocean, we distinguish two geographic zones: (i) dust interactions within the Intertropical Convergence Zone (ITCZ), or the dust–ITCZ interaction zone (DIZ), and (ii) the adjacent dust transport over the Atlantic Ocean (DTA) zone. In the latter zone, the dust loading shows a steep and linear gradient westward over the Atlantic Ocean since particle sedimentation is the dominant removal process, whereas in the DIZ zone aerosol–cloud interactions, wet deposition and scavenging processes determine the extent of the dust outflow. Generally, the EMAC simulated

dust compares well with CALIPSO observations; however, our reference model configuration tends to overestimate the dust extinction at a lower elevation and underestimates it at a higher elevation. The aerosol optical depth (AOD) over the Caribbean responds to the dust emission flux only when the emitted dust mass is significantly increased over the source region in Africa by a factor of 10. These findings point to the dominant role of dust removal (especially wet deposition) in transatlantic dust transport. Experiments with different convection schemes have indeed revealed that the transatlantic dust transport is more sensitive to the convection scheme than to the dust emission flux parameterization.

To study the impact of dust chemical aging, we focus on a major dust outflow in July 2009. We use the calcium cation as a proxy for the overall chemical reactive dust fraction and consider the uptake of major inorganic acids (i.e., H_2SO_4 , HNO_3 and HCl) and their anions, i.e., sulfate (SO_4^{2-}), bisulfate (HSO_4^-), nitrate (NO_3^-) and chloride (Cl^-), on the surface of mineral particles. The subsequent neutralization reactions with the calcium cation form various salt compounds that cause the uptake of water vapor from the atmosphere, i.e., through the chemical aging of dust particles leading to an increase of 0.15 in the AOD under subsaturated conditions (July 2009 monthly mean). As a result of the radiative feedback on surface winds, dust emissions increased regionally. On the other hand, the aged dust particles, compared to the non-aged particles, are more efficiently removed by both wet and dry deposition due to the increased hygroscopicity and particle size (mainly due to water uptake). The enhanced removal of aged particles decreases the dust burden and lifetime, which indirectly reduces the dust AOD by 0.05 (monthly mean). Both processes can be significant (major dust outflow, July 2009), but the net effect depends on the region and level of dust chemical aging.

Nitrate transboundary heavy pollution over East Asia in winter

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Source: *Atmos. Chem. Phys.*, 17, 3823-3843, 2017
<https://doi.org/10.5194/acp-17-3823-2017>

High $\text{PM}_{2.5}$ concentrations of around $100 \mu\text{g m}^{-3}$ were observed twice during an intensive observation campaign in January 2015 at Fukuoka (33.52°N , 130.47°E) in western Japan. These events were analyzed comprehensively with a regional chemical transport model and synergetic ground-based observations with state-of-the-art measurement systems, which can capture the behavior of secondary inorganic aerosols (SO_4^{2-} , NO_3^- , and NH_4^+). The first episode of high $\text{PM}_{2.5}$ concentration was dominated by NO_3^- (type N) and the second episode by SO_4^{2-} (type S). The concentration of NH_4^+ (the counterion for SO_4^{2-} and NO_3^-) was high for both types. A sensitivity simulation in the chemical transport model showed that the dominant contribution was from transboundary air pollution for both types. To investigate the differences between these types further, the chemical transport model results were examined, and a backward trajectory analysis was used to provide additional information. During both types of episodes, high concentrations of NO_3^- were found above China, and an air mass that originated from northeast China reached Fukuoka. The travel time from the coastline of China to Fukuoka differed between types: it was 18 h for type N and 24 h for type S. The conversion ratio of SO_2 to SO_4^{2-} (Fs) was less than 0.1 for type N, but reached 0.3 for type S as the air mass approached Fukuoka. The higher Fs for type S was

related to the higher relative humidity and the concentration of H_2O_2 , which produces H_2O_2 , the most effective oxidant for the aqueous-phase production of SO_4^{2-} . Analyzing the gas ratio as an indicator of the sensitivity of NO_3^- to changes in SO_4^{2-} and NH_4^+ showed that the air mass over China was NH_3 -rich for type N, but almost NH_3 -neutral for type S. Thus, although the high concentration of NO_3^- above China gradually decreased during transport from China to Fukuoka, higher NO_3^- concentrations were maintained during transport owing to the lower SO_4^{2-} for type N. In contrast, for type S, the production of SO_4^{2-} led to the decomposition of NH_4NO_3 , and more SO_4^{2-} was transported. Notably, the type N transport pattern was limited to western Japan, especially the island of Kyushu. Transboundary air pollution dominated by SO_4^{2-} (type S) has been recognized as a major pattern of pollution over East Asia. However, our study confirms the importance of transboundary air pollution dominated by NO_3^- , which will help refine our understanding of transboundary heavy $\text{PM}_{2.5}$ pollution in winter over East Asia.

Upper tropospheric cloud systems derived from IR sounders: properties of cirrus anvils in the tropics

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Source: Atmos. Chem. Phys., 17, 3845-3859, 2017
<https://doi.org/10.5194/acp-17-3845-2017>

Representing about 30 % of the Earth's total cloud cover, upper tropospheric clouds play a crucial role in the climate system by modulating the Earth's energy budget and heat transport. When originating from convection, they often form organized systems. The high spectral resolution of the Atmospheric Infrared Sounder (AIRS) allows reliable cirrus identification, both from day and nighttime observations. Tropical upper tropospheric cloud systems have been analyzed by using a spatial composite technique on the retrieved cloud pressure of AIRS data. Cloud emissivity is used to distinguish convective core, cirrus and thin cirrus anvil within these systems. A comparison with simultaneous precipitation data from the Advanced Microwave Scanning Radiometer – Earth Observing System (AMSRE) shows that, for tropical upper tropospheric clouds, a cloud emissivity close to 1 is strongly linked to a high rain rate, leading to a proxy to identify convective cores. Combining AIRS cloud data with this cloud system approach, using physical variables, provides a new opportunity to relate the properties of the anvils, including also the thinner cirrus, to the convective cores. It also distinguishes convective cloud systems from isolated cirrus systems. Deep convective cloud systems, covering 15 % of the tropics, are further distinguished into single-core and multi-core systems. Though AIRS samples the tropics only twice per day, the evolution of longer-living convective systems can be still statistically captured, and we were able to select relatively mature single-core convective systems by using the fraction of convective core area within the cloud systems as a proxy for maturity. For these systems, we have demonstrated that the physical properties of the anvils are related to convective depth, indicated by the minimum retrieved cloud temperature within the convective core. Our analyses show that the size of the systems does in general increase with convective depth, though for similar convective depth oceanic convective cloud systems are slightly larger than continental ones, in agreement with other observations. In addition, our data reveal for the first time that the fraction of thin cirrus over the total anvil area increases with the convective depth similarly for oceanic and continental convective

systems. This has implications for the radiative feedbacks of anvils on convection which will be more closely studied in the future.

Global distribution of CO₂ in the upper troposphere and stratosphere

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Source: *Atmos. Chem. Phys.*, 17, 3861-3878, 2017
<https://doi.org/10.5194/acp-17-3861-2017>

In this study, we construct a new monthly zonal mean carbon dioxide (CO₂) distribution from the upper troposphere to the stratosphere over the 2000–2010 time period. This reconstructed CO₂ product is based on a Lagrangian backward trajectory model driven by ERA-Interim reanalysis meteorology and tropospheric CO₂ measurements. Comparisons of our CO₂ product to extratropical in situ measurements from aircraft transects and balloon profiles show remarkably good agreement. The main features of the CO₂ distribution include (1) relatively large mixing ratios in the tropical stratosphere; (2) seasonal variability in the extratropics, with relatively high mixing ratios in the summer and autumn hemisphere in the 15–20 km altitude layer; and (3) decreasing mixing ratios with increasing altitude from the upper troposphere to the middle stratosphere (~35 km). These features are consistent with expected variability due to the transport of long-lived trace gases by the stratospheric Brewer–Dobson circulation. The method used here to construct this CO₂ product is unique from other modelling efforts and should be useful for model and satellite validation in the upper troposphere and stratosphere as a prior for inversion modelling and to analyse features of stratosphere–troposphere exchange as well as the stratospheric circulation and its variability.

Lower tropospheric distributions of O₃ and aerosol over Raoyang, a rural site in the North China Plain

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Source: *Atmos. Chem. Phys.*, 17, 3891-3903, 2017
<https://doi.org/10.5194/acp-17-3891-2017>

The North China Plain (NCP) has become one of the most polluted regions in China, with the rapidly increasing economic growth in the past decades. High concentrations of ambient O₃ and aerosol have been observed at urban as well as rural sites in the NCP. Most of the in situ observations of air pollutants have been conducted near the ground so that current knowledge about the vertical distributions of tropospheric O₃ and aerosol over the NCP region is still limited. In this study, vertical profiles of O₃ and size-resolved aerosol concentrations below 2.5 km were measured in summer 2014 over a rural site in the NCP, using an unmanned aerial vehicle (UAV) equipped with miniature analyzers. In addition, vertical profiles of aerosol scattering property in the lower

troposphere and vertical profiles of O₃ below 1 km were also observed at the site using a lidar and tethered balloon, respectively. The depths of the mixed layer and residual layer were determined according to the vertical gradients of lidar particle extinction and aerosol number concentration. Average O₃ and size-resolved aerosol number concentration in both the mixed and residual layer were obtained from the data observed in seven UAV flights. The results show that during most of the flights the O₃ levels above the top of mixed layer were higher than those below. Such a positive gradient in the vertical distribution of O₃ makes the residual layer an important source of O₃ in the mixed layer, particularly during the morning when the top of mixed layer is rapidly elevated. In contrast to O₃, aerosol number concentration was normally higher in the mixed layer than in the residual layer, particularly in the early morning. Aerosol particles were overwhelmingly distributed in the size range < 1 µm, showing slight differences between the mixed and residual layers. Our measurements confirm that the lower troposphere over the rural area of the NCP is largely impacted by anthropogenic pollutants locally emitted or transported from urban areas. Compared with the historic O₃ vertical profiles over Beijing from the Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft (MOZAIC), a strong increase in O₃ can be found at all heights below 2.5 km in the decade from 2004 to 2014, with the largest enhancement of about 41.6 ppb. This indicates that the lower troposphere over the northern part of the NCP has experienced rapidly worsening photochemical pollution. This worsening trend in photochemical pollution deserves more attention in the future.

An investigation on the origin of regional springtime ozone episodes in the western Mediterranean

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Source: Atmos. Chem. Phys., 17, 3905-3928, 2017
<https://doi.org/10.5194/acp-17-3905-2017>

For the identification of regional springtime ozone episodes, rural European Monitoring and Evaluation Programme (EMEP) ozone measurements from countries surrounding the western Mediterranean (Spain, France, Switzerland, Italy, Malta) have been examined with emphasis on periods of high ozone-mixing ratios, according to the variation of the daily afternoon (12:00–18:00) ozone values. For two selected high ozone episodes in April and May 2008, composite NCEP/NCAR reanalysis maps of various meteorological parameters and/or their anomalies (geopotential height, specific humidity, vertical wind velocity omega, vector wind speed and temperature) at various tropospheric pressure levels have been examined together with the corresponding satellite Infrared Atmospheric Sounding Interferometer (IASI) ozone measurements (at 3 and 10 km), CHIMERE simulations, vertical ozone soundings and HYSPLIT back trajectories. The observations show that high ozone values are detected in several countries simultaneously over several days. Also, the examined spring ozone episodes over the western Mediterranean and in central Europe are linked to synoptic meteorological conditions very similar to those recently observed in summertime ozone episodes over the eastern Mediterranean (Kalabokas et al., 2013, 2015; Doche et al., 2014), where the transport of tropospheric ozone-rich air masses through atmospheric subsidence significantly influences the boundary layer and surface ozone-mixing ratios. In particular, the geographic areas with observed tropospheric subsidence seem to be the transition

regions between high-pressure and low-pressure systems. During the surface ozone episodes IASI satellite measurements show extended areas of high ozone in the lower- and upper-troposphere over the low-pressure system areas, adjacent to the anticyclones, which influence significantly the boundary layer and surface ozone-mixing ratios within the anticyclones by subsidence and advection in addition to the photochemically produced ozone there, resulting in exceedances of the 60 ppb standard.

Biomass burning and biogenic aerosols in northern Australia during the SAFIRED campaign

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Source: Atmos. Chem. Phys., 17, 3945-3961, 2017
<https://doi.org/10.5194/acp-17-3945-2017>

There is a lack of knowledge of how biomass burning aerosols in the tropics age, including those in the fire-prone Northern Territory in Australia. This paper reports chemical characterization of fresh and aged aerosols monitored during the 1-month-long SAFIRED (Savannah Fires in the Early Dry Season) field study, with an emphasis on the chemical signature and aging of organic aerosols. The campaign took place in June 2014 during the early dry season when the surface measurement site, the Australian Tropical Atmospheric Research Station (ATARS), located in the Northern Territory, was heavily influenced by thousands of wild and prescribed bushfires. ATARS was equipped with a wide suite of instrumentation for gaseous and aerosol characterization. A compact time-of-flight aerosol mass spectrometer was deployed to monitor aerosol chemical composition. Approximately 90 % of submicron non-refractory mass was composed of organic material. Ozone enhancement in biomass burning plumes indicated increased air mass photochemistry. The diversity in biomass burning emissions was illustrated through variability in chemical signature (e.g. wide range in f₄₄, from 0.06 to 0.18) for five intense fire events. The background particulate loading was characterized using positive matrix factorization (PMF). A PMF-resolved BBOA (biomass burning organic aerosol) factor comprised 24 % of the submicron non-refractory organic aerosol mass, confirming the significance of fire sources. A dominant PMF factor, OOA (oxygenated organic aerosol), made up 47 % of the sampled aerosol, illustrating the importance of aerosol aging in the Northern Territory. Biogenic isoprene-derived organic aerosol factor was the third significant fraction of the background aerosol (28 %).

Constraining sector-specific CO₂ and CH₄ emissions in the US

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Source: Atmos. Chem. Phys., 17, 3963-3985, 2017
<https://doi.org/10.5194/acp-17-3963-2017>

This review paper explores recent efforts to estimate state- and national-scale carbon dioxide (CO₂) and methane (CH₄) emissions from individual anthropogenic source sectors in the US. Nearly all state and national climate change regulations in the US target specific source sectors, and detailed monitoring of individual sectors presents a greater challenge than monitoring total emissions. We particularly focus on opportunities to synthesize disparate types of information on emissions, including emission inventory data and atmospheric greenhouse gas data.

We find that inventory estimates of sector-specific CO₂ emissions are sufficiently accurate for policy evaluation at the national scale but that uncertainties increase at state and local levels. CH₄ emission inventories are highly uncertain for all source sectors at all spatial scales, in part because of the complex, spatially variable relationships between economic activity and CH₄ emissions. In contrast to inventory estimates, top-down estimates use measurements of atmospheric mixing ratios to infer emissions at the surface; thus far, these efforts have had some success identifying urban CO₂ emissions and have successfully identified sector-specific CH₄ emissions in several opportunistic cases. We also describe a number of forward-looking opportunities that would aid efforts to estimate sector-specific emissions: fully combine existing top-down datasets, expand intensive aircraft measurement campaigns and measurements of secondary tracers, and improve the economic and demographic data (e.g., activity data) that drive emission inventories. These steps would better synthesize inventory and top-down data to support sector-specific emission reduction policies.

Satellite retrievals of dust aerosol over the Red Sea and the Persian Gulf (2005–2015)

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Source: Atmos. Chem. Phys., 17, 3987-4003, 2017
<https://doi.org/10.5194/acp-17-3987-2017>

The inter-annual variability of the dust aerosol presence over the Red Sea and the Persian Gulf is analysed over the period 2005–2015. Particular attention is paid to the variation in loading across the Red Sea, which has previously been shown to have a strong, seasonally dependent latitudinal gradient. Over the 11 years considered, the July mean 630 nm aerosol optical depth (AOD) derived from the Spinning Enhanced Visible and InfraRed Imager (SEVIRI) varies between 0.48 and 1.45 in the southern half of the Red Sea. In the north, the equivalent variation is between 0.22 and 0.66. The temporal and spatial pattern of variability captured by SEVIRI is also seen in AOD retrievals from the MODerate Imaging Spectroradiometer (MODIS), but there is a systematic offset between the two records. Comparisons of both sets of retrievals with ship- and land-based AERONET measurements show a high degree of correlation with biases of < 0.08. However, these comparisons typically only sample relatively low aerosol loadings. When both records are stratified by AOD retrievals from the Multi-angle Imaging SpectroRadiometer (MISR), opposing behaviour is revealed at high MISR AODs (> 1), with offsets of +0.19 for MODIS and –0.06 for SEVIRI. Similar behaviour is also seen over the Persian Gulf. Analysis of the scattering angles at which retrievals

from the SEVIRI and MODIS measurements are typically performed in these regions suggests that assumptions concerning particle sphericity may be responsible for the differences seen.

Volcanic ash modeling with the online NMMB-MONARCH-ASH v1.0 model: model description, case simulation, and evaluation

Alejandro Marti, Arnau Folch, Oriol Jorba, and Zavisla Janjic

Source: Atmos. Chem. Phys., 17, 4005-4030, 2017
<https://doi.org/10.5194/acp-17-4005-2017>

Traditionally, tephra transport and dispersal models have evolved decoupled (offline) from numerical weather prediction models. There is a concern that inconsistencies and shortcomings associated with this coupling strategy might lead to errors in the ash cloud forecast. Despite this concern and the significant progress in improving the accuracy of tephra dispersal models in the aftermath of the 2010 Eyjafjallajökull and 2011 Cordón Caulle eruptions, to date, no operational online dispersal model is available to forecast volcanic ash. Here, we describe and evaluate NMMB-MONARCH-ASH, a new online multi-scale meteorological and transport model that attempts to pioneer the forecast of volcanic aerosols at operational level. The model forecasts volcanic ash cloud trajectories, concentration of ash at relevant flight levels, and the expected deposit thickness for both regional and global configurations. Its online coupling approach improves the current state-of-the-art tephra dispersal models, especially in situations where meteorological conditions are changing rapidly in time, two-way feedbacks are significant, or distal ash cloud dispersal simulations are required. This work presents the model application for the first phases of the 2011 Cordón Caulle and 2001 Mount Etna eruptions. The computational efficiency of NMMB-MONARCH-ASH and its application results compare favorably with other long-range tephra dispersal models, supporting its operational implementation.

Remote sensing and modelling analysis of the extreme dust storm hitting the Middle East and eastern Mediterranean in September 2015

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Source: Atmos. Chem. Phys., 17, 4063-4079, 2017
<https://doi.org/10.5194/acp-17-4063-2017>

The extreme dust storm that affected the Middle East and the eastern Mediterranean in September 2015 resulted in record-breaking dust loads over Cyprus with aerosol optical depth exceeding 5.0 at 550 nm. We analyse this event using profiles from the European Aerosol Research Lidar Network (EARLINET) and the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO), geostationary observations from the Meteosat Second Generation (MSG) Spinning Enhanced Visible and Infrared Imager (SEVIRI), and high-resolution simulations from the

Regional Atmospheric Modeling System (RAMS). The analysis of modelling and remote sensing data reveals the main mechanisms that resulted in the generation and persistence of the dust cloud over the Middle East and Cyprus. A combination of meteorological and surface processes is found, including (a) the development of a thermal low in the area of Syria that results in unstable atmospheric conditions and dust mobilization in this area, (b) the convective activity over northern Iraq that triggers the formation of westward-moving haboobs that merge with the previously elevated dust layer, and (c) the changes in land use due to war in the areas of northern Iraq and Syria that enhance dust erodibility.

Evidence for renoxification in the tropical marine boundary layer

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Source: Atmos. Chem. Phys., 17, 4081-4092, 2017
<https://doi.org/10.5194/acp-17-4081-2017>

We present 2 years of NO_x observations from the Cape Verde Atmospheric Observatory located in the tropical Atlantic boundary layer. We find that NO_x mixing ratios peak around solar noon (at 20–30 pptV depending on season), which is counter to box model simulations that show a midday minimum due to OH conversion of NO₂ to HNO₃. Production of NO_x via decomposition of organic nitrogen species and the photolysis of HNO₃ appear insufficient to provide the observed noontime maximum. A rapid photolysis of nitrate aerosol to produce HONO and NO₂, however, is able to simulate the observed diurnal cycle. This would make it the dominant source of NO_x at this remote marine boundary layer site, overturning the previous paradigm according to which the transport of organic nitrogen species, such as PAN, is the dominant source. We show that observed mixing ratios (November–December 2015) of HONO at Cape Verde (~ 3.5 pptV peak at solar noon) are consistent with this route for NO_x production. Reactions between the nitrate radical and halogen hydroxides which have been postulated in the literature appear to improve the box model simulation of NO_x. This rapid conversion of aerosol phase nitrate to NO_x changes our perspective of the NO_x cycling chemistry in the tropical marine boundary layer, suggesting a more chemically complex environment than previously thought.

Wave modulation of the extratropical tropopause inversion layer

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Source: Atmos. Chem. Phys., 17, 4093-4114, 2017
<https://doi.org/10.5194/acp-17-4093-2017>

This study aims to quantify how much of the observed strength and variability in the zonal-mean extratropical tropopause inversion layer (TIL) comes from the modulation of the temperature field and its gradients around the tropopause by planetary- and synoptic-scale waves. By analyzing high-resolution observations, it also puts other TIL enhancing mechanisms into context.

Using gridded Global Positioning System radio occultation (GPS-RO) temperature profiles from the COSMIC mission (2007–2013), we are able to extract the extratropical wave signal by a simplified wavenumber–frequency domain filtering method and quantify the resulting TIL enhancement. By subtracting the extratropical wave signal, we show how much of the TIL is associated with other processes, at mid- and high latitudes, for both hemispheres and all seasons.

The transient and reversible modulation by planetary- and synoptic-scale waves is almost entirely responsible for the TIL in midlatitudes. This means that wave-mean flow interactions, inertia-gravity waves and the residual circulation are of minor importance for the strength and variability in the midlatitude TIL.

At polar regions, the extratropical wave modulation is dominant for the TIL strength as well, but there is also a clear fingerprint from sudden stratospheric warmings (SSWs) and final warmings in both hemispheres. Therefore, polar vortex breakups are partially responsible for the observed polar TIL strength in winter (if SSWs occur) and spring. Also, part of the polar summer TIL strength cannot be explained by extratropical wave modulation.

We suggest that our wave modulation mechanism integrates several TIL enhancing mechanisms proposed in previous literature while robustly disclosing the overall outcome of the different processes involved. By analyzing observations only, our study identifies which mechanisms dominate the extratropical TIL strength and their relative contribution. It remains to be determined, however, which roles the different planetary- and synoptic-scale wave types play within the total extratropical wave modulation of the TIL, as well as what causes the observed amplification of extratropical waves near the tropopause.

Day and night-time formation of organic nitrates at a forested mountain site in south-west Germany

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Source: *Atmos. Chem. Phys.*, 17, 4115–4130, 2017
<https://doi.org/10.5194/acp-17-4115-2017>

We report in situ measurements of total peroxy nitrates (Σ PNs) and total alkyl nitrates (Σ ANs) in a forested–urban location at the top of the Kleiner Feldberg mountain in south-west Germany. The data, obtained using thermal dissociation cavity ring-down spectroscopy (TD-CRDS) in August–September 2011 (PARADE campaign) and July 2015 (NOTOMO campaign), represent the first detailed study of Σ PNs and Σ ANs over continental Europe. We find that a significant fraction of NO_x (up to 75 %) is sequestered as organics nitrates at this site. Furthermore, we also show that the night-time production of alkyl nitrates by reaction of NO_3 with biogenic hydrocarbons is comparable to that from daytime OH-initiated oxidation pathways. The Σ ANs/ozone ratio obtained during PARADE was used to derive an approximate average yield of organic nitrates at noon from the OH initiated oxidation of volatile organic compounds (VOCs) of $\sim 7\%$ at this site in 2011, which is comparable with that obtained from an analysis of VOCs measured during the campaign. A

much lower AN yield, < 2 %, was observed in 2015, which may result from sampling air with different average air mass ages and thus different degrees of breakdown of assumptions used to derive the branching ratio, but it may also reflect a seasonal change in the VOC mixture at the site.

Field observations of volatile organic compound (VOC) exchange in red oaks

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Source: Atmos. Chem. Phys., 17, 4189-4207, 2017
<https://doi.org/10.5194/acp-17-4189-2017>

Volatile organic compounds (VOCs) emitted by forests strongly affect the chemical composition of the atmosphere. While the emission of isoprenoids has been largely characterized, forests also exchange many oxygenated VOCs (oVOCs), including methanol, acetone, methyl ethyl ketone (MEK), and acetaldehyde, which are less well understood. We monitored total branch-level exchange of VOCs of a strong isoprene emitter (*Quercus rubra* L.) in a mixed forest in New England, where canopy-level fluxes of VOCs had been previously measured. We report daily exchange of several oVOCs and investigated unknown sources and sinks, finding several novel insights. In particular, we found that emission of MEK is linked to uptake of methyl vinyl ketone (MVK), a product of isoprene oxidation. The link was confirmed by corollary experiments proving in vivo detoxification of MVK, which is harmful to plants. Comparison of MEK, MVK, and isoprene fluxes provided an indirect indication of within-plant isoprene oxidation. Furthermore, besides confirming bidirectional exchange of acetaldehyde, we also report for the first time direct evidence of benzaldehyde bidirectional exchange in forest plants. Net emission or deposition of benzaldehyde was found in different periods of measurements, indicating an unknown foliar sink that may influence atmospheric concentrations. Other VOCs, including methanol, acetone, and monoterpenes, showed clear daily emission trends but no deposition. Measured VOC emission and deposition rates were generally consistent with their ecosystem-scale flux measurements at a nearby site.

Microphysical sensitivity of coupled springtime Arctic stratocumulus to modelled primary ice over the ice pack, marginal ice, and ocean

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Source: Atmos. Chem. Phys., 17, 4209-4227, 2017
<https://doi.org/10.5194/acp-17-4209-2017>

This study uses large eddy simulations to test the sensitivity of single-layer mixed-phase stratocumulus to primary ice number concentrations in the European Arctic. Observations from the

Aerosol-Cloud Coupling and Climate Interactions in the Arctic (ACCACIA) campaign are considered for comparison with cloud microphysics modelled using the Large Eddy Model (LEM, UK Met. Office). We find that cloud structure is very sensitive to ice number concentrations, Nice, and small increases can cause persisting mixed-phase clouds to glaciata and break up.

Three key dependencies on Nice are identified from sensitivity simulations and comparisons with observations made over the sea ice pack, marginal ice zone (MIZ), and ocean. Over sea ice, we find deposition–condensation ice formation rates are overestimated, leading to cloud glaciation. When ice formation is limited to water-saturated conditions, we find microphysics comparable to aircraft observations over all surfaces considered. We show that warm supercooled ($-13\text{ }^{\circ}\text{C}$) mixed-phase clouds over the MIZ are simulated to reasonable accuracy when using both the DeMott et al.(2010) and Cooper(1986) primary ice nucleation parameterisations. Over the ocean, we find a strong sensitivity of Arctic stratus to Nice. The Cooper(1986) parameterisation performs poorly at the lower ambient temperatures, leading to a comparatively higher Nice(2.43 L^{-1} at the cloud-top temperature, approximately $-20\text{ }^{\circ}\text{C}$) and cloud glaciation. A small decrease in the predicted Nice (2.07 L^{-1} at $-20\text{ }^{\circ}\text{C}$), using the DeMott et al.(2010) parameterisation, causes mixed-phase conditions to persist for 24 h over the ocean. However, this representation leads to the formation of convective structures which reduce the cloud liquid water through snow precipitation, promoting cloud break-up through a depleted liquid phase. Decreasing the Nice further (0.54 L^{-1} , using a relationship derived from ACCACIA observations) allows mixed-phase conditions to be maintained for at least 24 h with more stability in the liquid and ice water paths. Sensitivity to Nice is also evident at low number concentrations, where $0.1 \times$ Nice predicted by the DeMott et al.(2010) parameterisation results in the formation of rainbands within the model; rainbands which also act to deplete the liquid water in the cloud and promote break-up.

Evaluation of the absorption Ångström exponents for traffic and wood burning in the Aethalometer-based source apportionment using radiocarbon measurements of ambient aerosol

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Source: Atmos. Chem. Phys., 17, 4229-4249, 2017
<https://doi.org/10.5194/acp-17-4229-2017>

Equivalent black carbon (EBC) measured by a multi-wavelength Aethalometer can be apportioned to traffic and wood burning. The method is based on the differences in the dependence of aerosol absorption on the wavelength of light used to investigate the sample, parameterized by the source-specific absorption Ångström exponent (α). While the spectral dependence (defined as α values) of the traffic-related EBC light absorption is low, wood smoke particles feature enhanced light absorption in the blue and near ultraviolet. Source apportionment results using this methodology are hence strongly dependent on the α values assumed for both types of emissions: traffic α_{TR} , and wood burning α_{WB} . Most studies use a single α_{TR} and α_{WB} pair in the Aethalometer model, derived from previous work. However, an accurate determination of the source specific α values is currently lacking and in some recent publications the applicability of the Aethalometer model was questioned.

Here we present an indirect methodology for the determination of α_{WB} and α_{TR} by comparing the source apportionment of EBC using the Aethalometer model with ^{14}C measurements of the EC fraction on 16 to 40 h filter samples from several locations and campaigns across Switzerland during 2005–2012, mainly in winter. The data obtained at eight stations with different source characteristics also enabled the evaluation of the performance and the uncertainties of the Aethalometer model in different environments. The best combination of α_{TR} and α_{WB} (0.9 and 1.68, respectively) was obtained by fitting the Aethalometer model outputs (calculated with the absorption coefficients at 470 and 950 nm) against the fossil fraction of EC (ECF/EC) derived from ^{14}C measurements. Aethalometer and ^{14}C source apportionment results are well correlated ($r = 0.81$) and the fitting residuals exhibit only a minor positive bias of 1.6 % and an average precision of 9.3 %. This indicates that the Aethalometer model reproduces reasonably well the ^{14}C results for all stations investigated in this study using our best estimate of a single α_{WB} and α_{TR} pair. Combining the EC, ^{14}C , and Aethalometer measurements further allowed assessing the dependence of the mass absorption cross section (MAC) of EBC on its source. Results indicate no significant difference in MAC at 880 nm between EBC originating from traffic or wood-burning emissions. Using ECF/EC as reference and constant a priori selected α_{TR} values, α_{WB} was also calculated for each individual data point. No clear station-to-station or season-to-season differences in α_{WB} were observed, but α_{TR} and α_{WB} values are interdependent. For example, an increase in α_{TR} by 0.1 results in a decrease in α_{WB} by 0.1. The fitting residuals of different α_{TR} and α_{WB} combinations depend on ECF/EC such that a good agreement cannot be obtained over the entire ECF/EC range using other α pairs. Additional combinations of $\alpha_{TR} = 0.8$, and 1.0 and $\alpha_{WB} = 1.8$ and 1.6, respectively, are possible but only for ECF/EC between ~ 40 and 85 %. Applying α values previously used in the literature such as α_{WB} of ~ 2 or any α_{WB} in combination with $\alpha_{TR} = 1.1$ to our data set results in large residuals. Therefore we recommend to use the best α combination as obtained here ($\alpha_{TR} = 0.9$ and $\alpha_{WB} = 1.68$) in future studies when no or only limited additional information like ^{14}C measurements are available. However, these results were obtained for locations impacted by black carbon (BC) mainly from traffic consisting of a modern car fleet and residential wood combustion with well-constrained combustion efficiencies. For regions of the world with different combustion conditions, additional BC sources, or fuels used, further investigations are needed.

Atmospheric trace metals measured at a regional background site (Welgegund) in South Africa

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Source: Atmos. Chem. Phys., 17, 4251-4263, 2017
<https://doi.org/10.5194/acp-17-4251-2017>

Atmospheric trace metals can cause a variety of health-related and environmental problems. Only a few studies on atmospheric trace metal concentrations have been conducted in South Africa. Therefore the aim of this study was to determine trace metal concentrations in aerosols collected at a regional background site, i.e. Welgegund, South Africa. PM₁, PM_{1-2.5} and PM_{2.5-10} samples

were collected for 13 months, and 31 atmospheric trace metal species were detected. Atmospheric iron (Fe) had the highest concentrations in all three size fractions, while calcium (Ca) was the second-most-abundant species. Chromium (Cr) and sodium (Na) concentrations were the third- and fourth-most-abundant species, respectively. The concentrations of the trace metal species in all three size ranges were similar, with the exception of Fe, which had higher concentrations in the PM₁ size fraction. With the exception of titanium (Ti), aluminium (Al) and manganese (Mg), 70 % or more of the trace metal species detected were in the smaller size fractions, which indicated the influence of industrial activities. However, the large influence of wind-blown dust was reflected by 30 % or more of trace metals being present in the PM_{2.5-10} size fraction. Comparison of trace metals determined at Welgegund to those in the western Bushveld Igneous Complex indicated that at both locations similar species were observed, with Fe being the most abundant. However, concentrations of these trace metal species were significantly higher in the western Bushveld Igneous Complex. Fe concentrations at the Vaal Triangle were similar to levels thereof at Welgegund, while concentrations of species associated with pyrometallurgical smelting were lower. Annual average Ni was 4 times higher, and annual average As was marginally higher than their respective European standard values, which could be attributed to regional influence of pyrometallurgical industries in the western Bushveld Igneous Complex. All three size fractions indicated elevated trace metal concentrations coinciding with the end of the dry season, which could partially be attributed to decreased wet removal and increases in wind generation of particulates. Principal component factor analysis (PCFA) revealed four meaningful factors in the PM₁ size fraction, i.e. crustal, pyrometallurgical-related and Au slimes dams. No meaningful factors were determined for the PM_{1-2.5} and PM_{2.5-10} size fractions, which was attributed to the large influence of wind-blown dust on atmospheric trace metals determined at Welgegund. Pollution roses confirmed the influence of wind-blown dust on trace metal concentrations measured at Welgegund, while the impact of industrial activities was also substantiated.

Carbonaceous aerosol source apportionment using the Aethalometer model – evaluation by radiocarbon and levoglucosan analysis at a rural background site in southern Sweden

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Source: Atmos. Chem. Phys., 17, 4265-4281, 2017
<https://doi.org/10.5194/acp-17-4265-2017>

With the present demand on fast and inexpensive aerosol source apportionment methods, the Aethalometer model was evaluated for a full seasonal cycle (June 2014–June 2015) at a rural atmospheric measurement station in southern Sweden by using radiocarbon and levoglucosan measurements. By utilizing differences in absorption of UV and IR, the Aethalometer model apportions carbon mass into wood burning (WB) and fossil fuel combustion (FF) aerosol. In this study, a small modification in the model in conjunction with carbon measurements from thermal-optical analysis allowed apportioned non-light-absorbing biogenic aerosol to vary in time. The absorption differences between WB and FF can be quantified by the absorption Ångström exponent (AAE). In this study AAE_{WB} was set to 1.81 and AAE_{FF} to 1.0. Our observations show that the AAE was elevated during winter (1.36 ± 0.07) compared to summer (1.12 ± 0.07). Quantified WB aerosol

showed good agreement with levoglucosan concentrations, both in terms of correlation ($R^2 = 0.70$) and in comparison to reference emission inventories. WB aerosol showed strong seasonal variation with high concentrations during winter ($0.65 \mu\text{g m}^{-3}$, 56 % of total carbon) and low concentrations during summer ($0.07 \mu\text{g m}^{-3}$, 6 % of total carbon). FF aerosol showed less seasonal dependence; however, black carbon (BC) FF showed clear diurnal patterns corresponding to traffic rush hour peaks. The presumed non-light-absorbing biogenic carbonaceous aerosol concentration was high during summer ($1.04 \mu\text{g m}^{-3}$, 72 % of total carbon) and low during winter ($0.13 \mu\text{g m}^{-3}$, 8 % of total carbon). Aethalometer model results were further compared to radiocarbon and levoglucosan source apportionment results. The comparison showed good agreement for apportioned mass of WB and biogenic carbonaceous aerosol, but discrepancies were found for FF aerosol mass. The Aethalometer model overestimated FF aerosol mass by a factor of 1.3 compared to radiocarbon and levoglucosan source apportionment. A performed sensitivity analysis suggests that this discrepancy can be explained by interference of non-light-absorbing biogenic carbon during winter. In summary, the Aethalometer model offers a cost-effective yet robust high-time-resolution source apportionment at rural background stations compared to a radiocarbon and levoglucosan alternative.

Chemical transport model simulations of organic aerosol in southern California: model evaluation and gasoline and diesel source contributions

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Source: Atmos. Chem. Phys., 17, 4305-4318, 2017
<https://doi.org/10.5194/acp-17-4305-2017>

Gasoline- and diesel-fueled engines are ubiquitous sources of air pollution in urban environments. They emit both primary particulate matter and precursor gases that react to form secondary particulate matter in the atmosphere. In this work, we updated the organic aerosol module and organic emissions inventory of a three-dimensional chemical transport model, the Community Multiscale Air Quality Model (CMAQ), using recent, experimentally derived inputs and parameterizations for mobile sources. The updated model included a revised volatile organic compound (VOC) speciation for mobile sources and secondary organic aerosol (SOA) formation from unspiciated intermediate volatility organic compounds (IVOCs). The updated model was used to simulate air quality in southern California during May and June 2010, when the California Research at the Nexus of Air Quality and Climate Change (CalNex) study was conducted. Compared to the Traditional version of CMAQ, which is commonly used for regulatory applications, the updated model did not significantly alter the predicted organic aerosol (OA) mass concentrations but did substantially improve predictions of OA sources and composition (e.g., POA-SOA split), as well as ambient IVOC concentrations. The updated model, despite substantial differences in emissions and chemistry, performed similar to a recently released research version of CMAQ (Woody et al., 2016) that did not include the updated VOC and IVOC emissions and SOA data. Mobile sources were predicted to contribute 30–40 % of the OA in southern California (half of which was SOA), making mobile sources the single largest source contributor to OA in southern California. The remainder of the OA was attributed to non-mobile anthropogenic sources (e.g., cooking, biomass burning) with biogenic sources contributing to less than 5 % to the total OA. Gasoline sources were predicted to contribute about 13 times more OA than diesel sources; this difference was driven by

differences in SOA production. Model predictions highlighted the need to better constrain multi-generational oxidation reactions in chemical transport models.

Source attribution of black carbon and its direct radiative forcing in China

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Source: *Atmos. Chem. Phys.*, 17, 4319-4336, 2017

<https://doi.org/10.5194/acp-17-4319-2017>

The source attributions for mass concentration, haze formation, transport and direct radiative forcing of black carbon (BC) in various regions of China are quantified in this study using the Community Earth System Model (CESM) with a source-tagging technique. Anthropogenic emissions are from the Community Emissions Data System that is newly developed for the Coupled Model Intercomparison Project Phase 6 (CMIP6). Over north China where the air quality is often poor, about 90 % of near-surface BC concentration is contributed by local emissions. Overall, 35 % of BC concentration over south China in winter can be attributed to emissions from north China, and 19 % comes from sources outside China in spring. For other regions in China, BC is largely contributed from nonlocal sources. We further investigated potential factors that contribute to the poor air quality in China. During polluted days, a net inflow of BC transported from nonlocal source regions associated with anomalous winds plays an important role in increasing local BC concentrations. BC-containing particles emitted from East Asia can also be transported across the Pacific. Our model results show that emissions from inside and outside China are equally important for the BC outflow from East Asia, while emissions from China account for 8 % of BC concentration and 29 % in column burden in the western United States in spring. Radiative forcing estimates show that 65 % of the annual mean BC direct radiative forcing (2.2 W m^{-2}) in China results from local emissions, and the remaining 35 % is contributed by emissions outside of China. Efficiency analysis shows that a reduction in BC emissions over eastern China could have a greater benefit for the regional air quality in China, especially in the winter haze season.

Influence of 2000–2050 climate change on particulate matter in the United States: results from a new statistical model

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Source: *Atmos. Chem. Phys.*, 17, 4355-4367, 2017

<https://doi.org/10.5194/acp-17-4355-2017>

We use a statistical model to investigate the effect of 2000–2050 climate change on fine particulate matter (PM_{2.5}) air quality across the contiguous United States. By applying observed relationships of PM_{2.5} and meteorology to the IPCC Coupled Model Intercomparison Project Phase 5 (CMIP5) archives, we bypass some of the uncertainties inherent in chemistry-climate models. Our approach

uses both the relationships between PM_{2.5} and local meteorology as well as the synoptic circulation patterns, defined as the singular value decomposition (SVD) pattern of the spatial correlations between PM_{2.5} and meteorological variables in the surrounding region. Using an ensemble of 19 global climate models (GCMs) under the RCP4.5 scenario, we project an increase of 0.4–1.4 $\mu\text{g m}^{-3}$ in annual mean PM_{2.5} in the eastern US and a decrease of 0.3–1.2 $\mu\text{g m}^{-3}$ in the Intermountain West by the 2050s, assuming present-day anthropogenic sources of PM_{2.5}. Mean summertime PM_{2.5} increases as much as 2–3 $\mu\text{g m}^{-3}$ in the eastern United States due to faster oxidation rates and greater mass of organic aerosols from biogenic emissions. Mean wintertime PM_{2.5} decreases by 0.3–3 $\mu\text{g m}^{-3}$ over most regions in the United States, likely due to the volatilization of ammonium nitrate. Our approach provides an efficient method to calculate the potential climate penalty on air quality across a range of models and scenarios. We find that current atmospheric chemistry models may underestimate or even fail to capture the strongly positive sensitivity of monthly mean PM_{2.5} to temperature in the eastern United States in summer, and they may underestimate future changes in PM_{2.5} in a warmer climate. In GEOS-Chem, the underestimate in monthly mean PM_{2.5}–temperature relationship in the east in summer is likely caused by overly strong negative sensitivity of monthly mean low cloud fraction to temperature in the assimilated meteorology ($\sim -0.04 \text{ K}^{-1}$) compared to the weak sensitivity implied by satellite observations ($\pm 0.01 \text{ K}^{-1}$). The strong negative dependence of low cloud cover on temperature in turn causes the modeled rates of sulfate aqueous oxidation to diminish too rapidly as temperatures rise, leading to the underestimate of sulfate–temperature slopes, especially in the south. Our work underscores the importance of evaluating the sensitivity of PM_{2.5} to its key controlling meteorological variables in climate-chemistry models on multiple timescales before they are applied to project future air quality.

Estimates of the organic aerosol volatility in a boreal forest using two independent methods

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Source: Atmos. Chem. Phys., 17, 4387-4399, 2017
<https://doi.org/10.5194/acp-17-4387-2017>

The volatility distribution of secondary organic aerosols that formed and had undergone aging – i.e., the particle mass fractions of semi-volatile, low-volatility and extremely low volatility organic compounds in the particle phase – was characterized in a boreal forest environment of Hyytiälä, southern Finland. This was done by interpreting field measurements using a volatility tandem differential mobility analyzer (VTDMA) with a kinetic evaporation model. The field measurements were performed during April and May 2014. On average, 40 % of the organics in particles were semi-volatile, 34 % were low-volatility organics and 26 % were extremely low volatility organics. The model was, however, very sensitive to the vaporization enthalpies assumed for the organics ($\Delta\text{H}_{\text{VAP}}$). The best agreement between the observed and modeled temperature dependence of the evaporation was obtained when effective vaporization enthalpy values of 80 kJ mol^{−1} were assumed. There are several potential reasons for the low effective enthalpy value, including molecular decomposition or dissociation that might occur in the particle phase upon heating,

mixture effects and compound-dependent uncertainties in the mass accommodation coefficient. In addition to the VTDMA-based analysis, semi-volatile and low-volatility organic mass fractions were independently determined by applying positive matrix factorization (PMF) to high-resolution aerosol mass spectrometer (HR-AMS) data. The factor separation was based on the oxygenation levels of organics, specifically the relative abundance of mass ions at m/z 43 (f43) and m/z 44 (f44). The mass fractions of these two organic groups were compared against the VTDMA-based results. In general, the best agreement between the VTDMA results and the PMF-derived mass fractions of organics was obtained when $\Delta H_{VAP} = 80 \text{ kJ mol}^{-1}$ was set for all organic groups in the model, with a linear correlation coefficient of around 0.4. However, this still indicates that only about 16 % (R^2) of the variation can be explained by the linear regression between the results from these two methods. The prospect of determining of extremely low volatility organic aerosols (ELVOAs) from AMS data using the PMF analysis should be assessed in future studies.

Impacts of coal burning on ambient PM_{2.5} pollution in China

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Source: Atmos. Chem. Phys., 17, 4477-4491, 2017
<https://doi.org/10.5194/acp-17-4477-2017>

High concentration of fine particles (PM_{2.5}), the primary concern about air quality in China, is believed to closely relate to China's large consumption of coal. In order to quantitatively identify the contributions of coal combustion in different sectors to ambient PM_{2.5}, we developed an emission inventory for the year 2013 using up-to-date information on energy consumption and emission controls, and we conducted standard and sensitivity simulations using the chemical transport model GEOS-Chem. According to the simulation, coal combustion contributes $22 \mu\text{g m}^{-3}$ (40 %) to the total PM_{2.5} concentration at national level (averaged in 74 major cities) and up to $37 \mu\text{g m}^{-3}$ (50 %) in the Sichuan Basin. Among major coal-burning sectors, industrial coal burning is the dominant contributor, with a national average contribution of $10 \mu\text{g m}^{-3}$ (17 %), followed by coal combustion in power plants and the domestic sector. The national average contribution due to coal combustion is estimated to be $18 \mu\text{g m}^{-3}$ (46 %) in summer and $28 \mu\text{g m}^{-3}$ (35 %) in winter. While the contribution of domestic coal burning shows an obvious reduction from winter to summer, contributions of coal combustion in power plants and the industrial sector remain at relatively constant levels throughout the year.

Probing into the aging dynamics of biomass burning aerosol by using satellite measurements of aerosol optical depth and carbon monoxide

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Source: *Atmos. Chem. Phys.*, 17, 4513-4537, 2017
<https://doi.org/10.5194/acp-17-4513-2017>

Carbonaceous aerosol released into the atmosphere from open biomass burning (BB) is known to undergo considerable chemical and physical transformations (aging). However, there is substantial controversy about the nature and observable effects of these transformations. A shortage of consistent observational evidence on BB aerosol aging processes under different environmental conditions and at various temporal scales hinders development of their adequate representations in chemistry transport models (CTMs). In this study, we obtain insights into the BB aerosol dynamics by using available satellite measurements of aerosol optical depth (AOD) and carbon monoxide (CO). The basic concept of our method is to consider AOD as a function of the BB aerosol photochemical age (that is, the time period characterizing the exposure of BB aerosol emissions to atmospheric oxidation reactions) predicted by means of model tracers. We evaluate the AOD enhancement ratio (ER) defined as the ratio of optical depth of actual BB aerosol with respect to that of a modeled aerosol tracer that is assumed to originate from the same fires as the real BB aerosol but that is not affected by any aging processes. To limit possible effects of model transport errors, the AOD measurements are normalized to CO column amounts that are also retrieved from satellite measurements. The method is applied to the analysis of the meso- and synoptic-scale evolution of aerosol in smoke plumes from major wildfires that occurred in Siberia in summer 2012. AOD and CO retrievals from MODIS and IASI measurements, respectively, are used in combination with simulations performed with the CHIMERE CTM. The analysis indicates that aging processes strongly affected the evolution of BB aerosol in the situation considered, especially in dense plumes (with spatial average PM_{2.5} concentration exceeding 100 $\mu\text{g m}^{-3}$). For such plumes, the ER is found to increase almost 2-fold on the scale of ~ 10 h of daytime aerosol evolution (after a few first hours of the evolution that are not resolved in our analysis). The robustness of this finding is corroborated by sensitivity tests and Monte Carlo experiments. Furthermore, a simulation using the volatility basis set framework suggests that a large part of the increase in the ER can be explained by atmospheric processing of semi-volatile organic compounds. Our results are consistent with findings of a number of earlier studies reporting considerable underestimation of AOD by CTMs in which BB aerosol aging processes have either been disregarded or simulated in a highly simplified way. In general, this study demonstrates the feasibility of using satellite measurements of AOD in biomass burning plumes in combination with aerosol tracer simulations for the investigation of BB aerosol evolution and validation of BB aerosol aging schemes in atmospheric models.

Constraining N₂O emissions since 1940 using firn air isotope measurements in both hemispheres

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Source: *Atmos. Chem. Phys.*, 17, 4539-4564, 2017
<https://doi.org/10.5194/acp-17-4539-2017>

N₂O is currently the third most important anthropogenic greenhouse gas in terms of radiative forcing and its atmospheric mole fraction is rising steadily. To quantify the growth rate and its causes over the past decades, we performed a multi-site reconstruction of the atmospheric N₂O mole fraction and isotopic composition using new and previously published firn air data collected from Greenland and Antarctica in combination with a firn diffusion and densification model. The multi-site reconstruction showed that while the global mean N₂O mole fraction increased from (290 ± 1) nmol mol⁻¹ in 1940 to (322 ± 1) nmol mol⁻¹ in 2008, the isotopic composition of atmospheric N₂O decreased by (-2.2 ± 0.2) ‰ for $\delta^{15}\text{N}_{\text{av}}$, (-1.0 ± 0.3) ‰ for $\delta^{18}\text{O}$, (-1.3 ± 0.6) ‰ for $\delta^{15}\text{N}_{\alpha}$, and (-2.8 ± 0.6) ‰ for $\delta^{15}\text{N}_{\beta}$ over the same period. The detailed temporal evolution of the mole fraction and isotopic composition derived from the firn air model was then used in a two-box atmospheric model (comprising a stratospheric box and a tropospheric box) to infer changes in the isotopic source signature over time. The precise value of the source strength depends on the choice of the N₂O lifetime, which we choose to fix at 123 years. The average isotopic composition over the investigated period is $\delta^{15}\text{N}_{\text{av}} = (-7.6 \pm 0.8)$ ‰ (vs. air-N₂), $\delta^{18}\text{O} = (32.2 \pm 0.2)$ ‰ (vs. Vienna Standard Mean Ocean Water – VSMOW) for $\delta^{18}\text{O}$, $\delta^{15}\text{N}_{\alpha} = (-3.0 \pm 1.9)$ ‰ and $\delta^{15}\text{N}_{\beta} = (-11.7 \pm 2.3)$ ‰. $\delta^{15}\text{N}_{\text{av}}$ and $\delta^{15}\text{N}_{\beta}$ show some temporal variability, while for the other signatures the error bars of the reconstruction are too large to retrieve reliable temporal changes. Possible processes that may explain trends in ¹⁵N are discussed. The ¹⁵N site preference ($= \delta^{15}\text{N}_{\alpha} - \delta^{15}\text{N}_{\beta}$) provides evidence of a shift in emissions from denitrification to nitrification, although the uncertainty envelopes are large.

A 15-year record of CO emissions constrained by MOPITT CO observations

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Source: *Atmos. Chem. Phys.*, 17, 4565-4583, 2017
<https://doi.org/10.5194/acp-17-4565-2017>

Long-term measurements from satellites and surface stations have demonstrated a decreasing trend of tropospheric carbon monoxide (CO) in the Northern Hemisphere over the past decade. Likely explanations for this decrease include changes in anthropogenic, fires, and/or biogenic emissions or changes in the primary chemical sink hydroxyl radical (OH). Using remotely sensed CO measurements from the Measurement of Pollution in the Troposphere (MOPITT) satellite instrument, in situ methyl chloroform (MCF) measurements from the World Data Centre for Greenhouse Gases (WDCGG) and the adjoint of the GEOS-Chem model, we estimate the change in global CO emissions from 2001 to 2015. We show that the loss rate of MCF varied by 0.2 % in the past 15 years, indicating that changes in global OH distributions do not explain the recent decrease in CO. Our two-step inversion approach for estimating CO emissions is intended to mitigate the effect of bias errors in the MOPITT data as well as model errors in transport and chemistry, which are the primary factors contributing to the uncertainties when quantifying CO emissions using these remotely sensed data. Our results confirm that the decreasing trend of tropospheric CO in the Northern Hemisphere is due to decreasing CO emissions from anthropogenic and biomass burning sources. In particular, we find decreasing CO emissions from the United States and China in the past

15 years, and unchanged anthropogenic CO emissions from Europe since 2008. We find decreasing trends of biomass burning CO emissions from boreal North America, boreal Asia and South America, but little change over Africa. In contrast to prior results, we find that a positive trend in CO emissions is likely for India and southeast Asia.

Changing trends and emissions of hydrochlorofluorocarbons (HCFCs) and their hydrofluorocarbon (HFCs) replacements

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Source: Atmos. Chem. Phys., 17, 4641-4655, 2017
<https://doi.org/10.5194/acp-17-4641-2017>

High-frequency, in situ global observations of HCFC-22 (CH₂ClF₂), HCFC-141b (CH₃CCl₂F), HCFC-142b (CH₃CClF₂) and HCFC-124 (CHClFCF₃) and their main HFC replacements, HFC-134a (CH₂FCF₃), HFC-125 (CHF₂CF₃), HFC-143a (CH₃CF₃) and HFC-32 (CH₂F₂), have been used to determine their changing global growth rates and emissions in response to the Montreal Protocol and its recent amendments. Global mean mole fractions of HCFC-22, -141b, and -142b have increased throughout the observation period, reaching 234, 24.3 and 22.4 pmol mol⁻¹, respectively, in 2015. HCFC-124 reached a maximum global mean mole fraction of 1.48 pmol mol⁻¹ in 2007 and has since declined by 23 % to 1.14 pmol mol⁻¹ in 2015. The HFCs all show increasing global mean mole fractions. In 2015 the global mean mole fractions (pmol mol⁻¹) were 83.3 (HFC-134a), 18.4 (HFC-125), 17.7 (HFC-143a) and 10.5 (HFC-32).

The 2007 adjustment to the Montreal Protocol required the accelerated phase-out of emissive uses of HCFCs with global production and consumption capped in 2013 to mitigate their environmental impact as both ozone-depleting substances and important greenhouse gases. We find that this change has coincided with a stabilisation, or moderate reduction, in global emissions of the four HCFCs with aggregated global emissions in 2015 of 449 ± 75 Gg yr⁻¹, in CO₂ equivalent units (CO₂ eq.) 0.76 ± 0.1 Gt yr⁻¹, compared with 483 ± 70 Gg yr⁻¹ (0.82 ± 0.1 Gt yr⁻¹ CO₂ eq.) in 2010 (uncertainties are 1σ throughout this paper). About 79 % of the total HCFC atmospheric burden in 2015 is HCFC-22, where global emissions appear to have been relatively similar since 2011, in spite of the 2013 cap on emissive uses. We attribute this to a probable increase in production and consumption of HCFC-22 in Montreal Protocol Article 5 (developing) countries and the continuing release of HCFC-22 from the large banks which dominate HCFC global emissions. Conversely, the four HFCs all show increasing mole fraction growth rates with aggregated global HFC emissions of 327 ± 70 Gg yr⁻¹ (0.65 ± 0.12 Gt yr⁻¹ CO₂ eq.) in 2015 compared to 240 ± 50 Gg yr⁻¹ (0.47 ± 0.08 Gt yr⁻¹ CO₂ eq.) in 2010. We also note that emissions of HFC-125 and HFC-32 appear to have increased more rapidly averaged over the 5-year period 2011–2015, compared to 2006–2010. As noted by Lunt et al. (2015) this may reflect a change to refrigerant blends, such as R-410A, which contain HFC-32 and -125 as a 50 : 50 blend.

Impact of typhoons on the composition of the upper troposphere within the Asian summer monsoon anticyclone: the SWOP campaign in Lhasa 2013

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Source: Atmos. Chem. Phys., 17, 4657-4672, 2017
<https://doi.org/10.5194/acp-17-4657-2017>

In the frame of the SWOP (sounding water vapour, ozone, and particle) campaign during the Asian summer monsoon (ASM), ozone and water vapour profiles were measured by balloon-borne sensors launched from Lhasa (29.66° N, 91.14° E, elevation 3650 m), China, in August 2013. In total, 24 soundings were launched, nearly half of which show strong variations in the relationship between ozone and water vapour in the tracer-tracer correlation in the upper troposphere and lower stratosphere (UTLS). For each sounding, 20-day backward trajectories were calculated using the trajectory module of the Chemical Lagrangian Model of the Stratosphere (CLaMS) to analyse these variations. The trajectory calculations demonstrate that three tropical cyclones (tropical storm Jebi, typhoons Utor and Trami), which occurred over the western Pacific Ocean during August 2013, had a considerable impact on the vertical distribution of ozone and water vapour by uplifting marine air masses to altitudes of the ASM anticyclone. Air parcels subsequently arrived at the observation site via two primary pathways: firstly via direct horizontal transport from the location of the typhoon to the station within approximately 3 days, and secondly via transport following the clockwise wind flow of the ASM within a timescale of 1 week. Furthermore, the interplay between the spatial position of the ASM anticyclone and tropical cyclones plays a key role in controlling the transport pathways of air parcels from the boundary layer of the western Pacific to Lhasa in horizontal and vertical transport. Moreover, the statistical analysis shows that the strongest impact by typhoons is found at altitudes between 14.5 and 17 km (365–375 K). Low ozone values (50–80 ppbv) were observed between 370 and 380 K due to the strong vertical transport within tropical cyclones.

OMI air-quality monitoring over the Middle East

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Source: Atmos. Chem. Phys., 17, 4687-4709, 2017
<https://doi.org/10.5194/acp-17-4687-2017>

Using Ozone Monitoring Instrument (OMI) trace gas vertical column observations of nitrogen dioxide (NO₂), formaldehyde (HCHO), sulfur dioxide (SO₂), and glyoxal (CHOCHO), we have conducted a robust and detailed time series analysis to assess changes in local air quality for over 1000 locations (focussing on urban, oil refinery, oil port, and power plant targets) over the Middle East for 2005–2014. Apart from NO₂, which is highest over urban locations, average tropospheric

column levels of these trace gases are highest over oil ports and refineries. The highest average pollution levels over urban settlements are typically in Bahrain, Kuwait, Qatar, and the United Arab Emirates.

We detect 278 statistically significant and real linear NO₂ trends in total. Over urban areas NO₂ increased by up to 12 % yr⁻¹, with only two locations showing a decreasing trend. Over oil refineries, oil ports, and power plants, NO₂ increased by about 2–9 % yr⁻¹. For HCHO, 70 significant and real trends were detected, with HCHO increasing by 2–7 % yr⁻¹ over urban settlements and power plants and by about 2–4 % yr⁻¹ over refineries and oil ports. Very few SO₂ trends were detected, which varied in direction and magnitude (23 increasing and 9 decreasing). Apart from two locations where CHOCHO is decreasing, we find that glyoxal tropospheric column levels are not changing over the Middle East. Hence, for many locations in the Middle East, OMI observes a degradation in air quality over 2005–2014. This study therefore demonstrates the capability of OMI to generate long-term air-quality monitoring at local scales over this region.

Long-term air concentrations, wet deposition, and scavenging ratios of inorganic ions, HNO₃, and SO₂ and assessment of aerosol and precipitation acidity at Canadian rural locations

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Source: Atmos. Chem. Phys., 17, 4711–4730, 2017
<https://doi.org/10.5194/acp-17-4711-2017>

This study analyzed long-term air concentrations and annual wet deposition of inorganic ions and aerosol and precipitation acidity at 31 Canadian sites from 1983 to 2011. Scavenging ratios of inorganic ions and relative contributions of particulate- and gas-phase species to NH₄⁺, NO₃⁻, and SO₄²⁻ wet deposition were determined. Geographical patterns of atmospheric Ca²⁺, Na⁺, Cl⁻, NH₄⁺, NO₃⁻, and SO₄²⁻ were similar to wet deposition and attributed to anthropogenic sources, sea-salt emissions, and agricultural emissions. Decreasing trends in atmospheric NH₄⁺ (1994–2010) and SO₄²⁻ (1983–2010) were prevalent. Atmospheric NO₃⁻ increased prior to 2001 and then declined afterwards. These results are consistent with SO₂, NO_x and NH₃ emission trends in Canada and the USA. Widespread declines in annual NO₃⁻ and SO₄²⁻ wet deposition ranged from 0.07 to 1.0 kg ha⁻¹ a⁻¹ (1984–2011). Acidic aerosols and precipitation impacted southern and eastern Canada more than western Canada; however, both trends have been decreasing since 1994. Scavenging ratios of particulate NH₄⁺, SO₄²⁻ and NO₃⁻ differed from literature values by 22 %, 44 %, and a factor of 6, respectively, because of the exclusion of gas scavenging in previous studies. Average gas and particle scavenging contributions to total wet deposition were estimated to be 72 % for HNO₃ and 28 % for particulate NO₃⁻, 37 % for SO₂ and 63 % for particulate SO₄²⁻, and 30 % for NH₃ and 70 % for particulate NH₄⁺.

Emission factors and light absorption properties of brown carbon from household coal combustion in China

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Source: *Atmos. Chem. Phys.*, 17, 4769-4780, 2017
<https://doi.org/10.5194/acp-17-4769-2017>

Brown carbon (BrC) draws increasing attention due to its effects on climate and other environmental factors. In China, household coal burned for heating and cooking purposes releases huge amounts of carbonaceous particles every year; however, BrC emissions have rarely been estimated in a persuasive manner due to the unavailable emission characteristics. Here, seven coals jointly covering geological maturity from low to high were burned in four typical stoves as both chunk and briquette styles. The optical integrating sphere (IS) method was applied to measure the emission factors (EFs) of BrC and black carbon (BC) via an iterative process using the different spectral dependence of light absorption for BrC and BC and using humic acid sodium salt (HASS) and carbon black (CarB) as reference materials. The following results have been found: (i) the average EFs of BrC for anthracite coal chunks and briquettes are 1.08 ± 0.80 and 1.52 ± 0.16 g kg⁻¹, respectively, and those for bituminous coal chunks and briquettes are 8.59 ± 2.70 and 4.01 ± 2.19 g kg⁻¹, respectively, reflecting a more significant decline in BrC EFs for bituminous coals than for anthracites due to briquetting. (ii) The BrC EF peaks at the middle of coal's geological maturity, displaying a bell-shaped curve between EF and volatile matter (Vdaf). (iii) The calculated BrC emissions from China's residential coal burning amounted to 592 Gg (1 Gg = 10⁹ g) in 2013, which is nearly half of China's total BC emissions. (iv) The absorption Ångström exponents (AAEs) of all coal briquettes are higher than those of coal chunks, indicating that the measure of coal briquetting increases the BrC/BC emission ratio and thus offsets some of the climate cooling effect of briquetting. (v) In the scenario of current household coal burning in China, solar light absorption by BrC (350–850 nm in this study) accounts for more than a quarter (0.265) of the total absorption. This implies the significance of BrC to climate modeling.

Consistent regional fluxes of CH₄ and CO₂ inferred from GOSAT proxy XCH₄ : XCO₂ retrievals, 2010–2014

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Source: *Atmos. Chem. Phys.*, 17, 4781-4797, 2017
<https://doi.org/10.5194/acp-17-4781-2017>

We use the GEOS-Chem global 3-D model of atmospheric chemistry and transport and an ensemble Kalman filter to simultaneously infer regional fluxes of methane (CH₄) and carbon dioxide (CO₂) directly from GOSAT retrievals of XCH₄:XCO₂, using sparse ground-based CH₄ and CO₂ mole fraction data to anchor the ratio. This work builds on the previously reported theory that takes into account that (1) these ratios are less prone to systematic error than either the full-physics data products or the proxy CH₄ data products; and (2) the resulting CH₄ and CO₂ fluxes are self-consistent. We show that a posteriori fluxes inferred from the GOSAT data generally outperform the fluxes inferred only from in situ data, as expected. GOSAT CH₄ and CO₂ fluxes are consistent with global growth rates for CO₂ and CH₄ reported by NOAA and have a range of independent data including new profile measurements (0–7 km) over the Amazon Basin that were collected specifically to help validate GOSAT over this geographical region. We find that large-scale multi-year annual a posteriori CO₂ fluxes inferred from GOSAT data are similar to those inferred from the in situ surface data but with smaller uncertainties, particularly over the tropics. GOSAT data are consistent with smaller peak-to-peak seasonal amplitudes of CO₂ than either the a priori or in situ inversion, particularly over the tropics and the southern extratropics. Over the northern extratropics, GOSAT data show larger uptake than the a priori but less than the in situ inversion, resulting in small net emissions over the year. We also find evidence that the carbon balance of tropical South America was perturbed following the droughts of 2010 and 2012 with net annual fluxes not returning to an approximate annual balance until 2013. In contrast, GOSAT data significantly changed the a priori spatial distribution of CH₄ emission with a 40 % increase over tropical South America and tropical Asia and a smaller decrease over Eurasia and temperate South America. We find no evidence from GOSAT that tropical South American CH₄ fluxes were dramatically affected by the two large-scale Amazon droughts. However, we find that GOSAT data are consistent with double seasonal peaks in Amazonian fluxes that are reproduced over the 5 years we studied: a small peak from January to April and a larger peak from June to October, which are likely due to superimposed emissions from different geographical regions.

Impacts of East Asian summer and winter monsoons on interannual variations of mass concentrations and direct radiative forcing of black carbon over eastern China

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Source: Atmos. Chem. Phys., 17, 4799–4816, 2017
<https://doi.org/10.5194/acp-17-4799-2017>

We applied a global three-dimensional chemical transport model (GEOS-Chem) to examine the impacts of the East Asian monsoon on the interannual variations of mass concentrations and direct radiative forcing (DRF) of black carbon (BC) over eastern China (110–125° E, 20–45° N). With emissions fixed at the year 2010 levels, model simulations were driven by the Goddard Earth Observing System (GEOS-4) meteorological fields for 1986–2006 and the Modern Era Retrospective-analysis for Research and Applications (MERRA) meteorological fields for 1980–2010. During the period of 1986–2006, simulated June–July–August (JJA) and December–January–February (DJF) surface BC concentrations were higher in MERRA than in GEOS-4 by 0.30 µg m⁻³ (44 %) and 0.77 µg m⁻³ (54 %), respectively, because of the generally weaker

precipitation in MERRA. We found that the strength of the East Asian summer monsoon (EASM; East Asian winter monsoon, EAWM) negatively correlated with simulated JJA (DJF) surface BC concentrations ($r = -0.7$ (-0.7) in GEOS-4 and -0.4 (-0.7) in MERRA), mainly by the changes in atmospheric circulation. Relative to the 5 strongest EASM years, simulated JJA surface BC concentrations in the 5 weakest monsoon years were higher over northern China ($110\text{--}125^\circ\text{E}$, $28\text{--}45^\circ\text{N}$) by $0.04\text{--}0.09\text{ }\mu\text{g m}^{-3}$ (3–11 %), but lower over southern China ($110\text{--}125^\circ\text{E}$, $20\text{--}27^\circ\text{N}$) by $0.03\text{--}0.04\text{ }\mu\text{g m}^{-3}$ (10–11 %). Compared to the 5 strongest EAWM years, simulated DJF surface BC concentrations in the 5 weakest monsoon years were higher by $0.13\text{--}0.15\text{ }\mu\text{g m}^{-3}$ (5–8 %) in northern China and by $0.04\text{--}0.10\text{ }\mu\text{g m}^{-3}$ (3–12 %) in southern China. The resulting JJA (DJF) mean all-sky DRF of BC at the top of the atmosphere was 0.04 W m^{-2} (3 %; 0.03 W m^{-2} , 2 %) higher in northern China but 0.06 W m^{-2} (14 %; 0.03 W m^{-2} , 3 %) lower in southern China. In the weakest monsoon years, the weaker vertical convection at the elevated altitudes led to the lower BC concentrations above 1–2 km in southern China, and therefore the lower BC DRF in the region. The differences in vertical profiles of BC between the weakest and strongest EASM years (1998–1997) and EAWM years (1990–1996) reached up to $-0.09\text{ }\mu\text{g m}^{-3}$ (–46 %) and $-0.08\text{ }\mu\text{g m}^{-3}$ (–11 %) at 1–2 km in eastern China.

Improving PM_{2.5} forecast over China by the joint adjustment of initial conditions and source emissions with an ensemble Kalman filter

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Source: Atmos. Chem. Phys., 17, 4837–4855, 2017
<https://doi.org/10.5194/acp-17-4837-2017>

In an attempt to improve the forecasting of atmospheric aerosols, the ensemble square root filter algorithm was extended to simultaneously optimize the chemical initial conditions (ICs) and emission input. The forecast model, which was expanded by combining the Weather Research and Forecasting with Chemistry (WRF-Chem) model and a forecast model of emission scaling factors, generated both chemical concentration fields and emission scaling factors. The forecast model of emission scaling factors was developed by using the ensemble concentration ratios of the WRF-Chem forecast chemical concentrations and also the time smoothing operator. Hourly surface fine particulate matter (PM_{2.5}) observations were assimilated in this system over China from 5 to 16 October 2014. A series of 48 h forecasts was then carried out with the optimized initial conditions and emissions on each day at 00:00 UTC and a control experiment was performed without data assimilation. In addition, we also performed an experiment of pure assimilation chemical ICs and the corresponding 48 h forecasts experiment for comparison. The results showed that the forecasts with the optimized initial conditions and emissions typically outperformed those from the control experiment. In the Yangtze River delta (YRD) and the Pearl River delta (PRD) regions, large reduction of the root-mean-square errors (RMSEs) was obtained for almost the entire 48 h forecast range attributed to assimilation. In particular, the relative reduction in RMSE due to assimilation was about 37.5 % at nighttime when WRF-Chem performed comparatively worse. In the Beijing–Tianjin–Hebei (JJI) region, relatively smaller improvements were achieved in the first 24 h forecast but then no improvements were achieved afterwards. Comparing to the forecasts with only the optimized ICs, the forecasts with the joint adjustment were always much better during the

night in the PRD and YRD regions. However, they were very similar during daytime in both regions. Also, they performed similarly for almost the entire 48 h forecast range in the JJJ region.

Tropospheric ozone maxima observed over the Arabian Sea during the pre-monsoon

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Source: Atmos. Chem. Phys., 17, 4915-4930, 2017
<https://doi.org/10.5194/acp-17-4915-2017>

An enhancement of the tropospheric ozone column (TOC) over Arabian Sea (AS) during the pre-monsoon season is reported in this study. The potential sources of the AS spring ozone pool are investigated by use of multiple data sets (e.g., SCIAMACHY Limb-Nadir-Matching TOC, OMI/MLS TOC, TES TOC, MACC reanalysis data, MOZART-4 model and HYSPLIT model). Three-quarters of the enhanced ozone concentrations are attributed to the 0–8 km height range. The main source of the ozone enhancement is considered to be caused by long-range transport of ozone pollutants from India (~ 50 % contributions to the lowest 4 km, ~ 20 % contributions to the 4–8 km height range), the Middle East, Africa and Europe (~ 30 % in total). In addition, the vertical pollution accumulation in the lower troposphere, especially at 4–8 km, was found to be important for the AS spring ozone pool formation. Local photochemistry, on the other hand, plays a negligible role in producing ozone at the 4–8 km height range. In the 0–4 km height range, ozone is quickly removed by wet deposition. The AS spring TOC maxima are influenced by the dynamical variations caused by the sea surface temperature (SST) anomaly during the El Niño period in 2005 and 2010 with a ~ 5 DU decrease.

Contributions of surface solar radiation and precipitation to the spatiotemporal patterns of surface and air warming in China from 1960 to 2003

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Source: Atmos. Chem. Phys., 17, 4931-4944, 2017
<https://doi.org/10.5194/acp-17-4931-2017>

Although global warming has been attributed to increases in atmospheric greenhouse gases, the mechanisms underlying spatiotemporal patterns of warming trends remain under debate. Herein, we analyzed surface and air warming observations recorded at 1977 stations in China from 1960 to 2003. Our results showed a significant spatial pattern for the warming of the daily maximum surface (Ts-max) and air (Ta-max) temperatures, and the pattern was stronger in northwest and northeast China and weaker or negative in South China and the North China Plain. These warming spatial patterns were attributed to surface shortwave solar radiation (Rs) and precipitation (P),

which play a key role in the surface energy budget. During the study period, R_s decreased by $-1.50 \pm 0.42 \text{ W m}^{-2} \text{ 10 yr}^{-1}$ in China, which reduced the trends of $T_s\text{-max}$ and $T_a\text{-max}$ by about 0.139 and $0.053 \text{ }^\circ\text{C 10 yr}^{-1}$, respectively. More importantly, the decreasing rates in South China and the North China Plain were stronger than those in other parts of China. The spatial contrasts in the trends of $T_s\text{-max}$ and $T_a\text{-max}$ in China were significantly reduced after adjusting for the effect of R_s and P . For example, after adjusting for the effect of R_s and P , the difference in the $T_s\text{-max}$ and $T_a\text{-max}$ values between the North China Plain and the Loess Plateau was reduced by 97.8 and 68.3 %, respectively; the seasonal contrast in $T_s\text{-max}$ and $T_a\text{-max}$ decreased by 45.0 and 17.2 %, respectively; and the daily contrast in the warming rates of the surface and air temperature decreased by 33.0 and 29.1 %, respectively. This study shows that the land energy budget plays an essential role in the identification of regional warming patterns.

Emissions of volatile organic compounds (VOCs) from concentrated animal feeding operations (CAFOs): chemical compositions and separation of sources

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Source: Atmos. Chem. Phys., 17, 4945-4956, 2017
<https://doi.org/10.5194/acp-17-4945-2017>

Concentrated animal feeding operations (CAFOs) emit a large number of volatile organic compounds (VOCs) to the atmosphere. In this study, we conducted mobile laboratory measurements of VOCs, methane (CH_4) and ammonia (NH_3) downwind of dairy cattle, beef cattle, sheep and chicken CAFO facilities in northeastern Colorado using a hydronium ion time-of-flight chemical-ionization mass spectrometer ($\text{H}_3\text{O}^+\text{ToF-CIMS}$), which can detect numerous VOCs. Regional measurements of CAFO emissions in northeastern Colorado were also performed using the NOAA WP-3D aircraft during the Shale Oil and Natural Gas Nexus (SONGNEX) campaign. Alcohols and carboxylic acids dominate VOC concentrations and the reactivity of the VOCs with hydroxyl (OH) radicals. Sulfur-containing and phenolic species provide the largest contributions to the odor activity values and the nitrate radical (NO_3) reactivity of VOC emissions, respectively. VOC compositions determined from mobile laboratory and aircraft measurements generally agree well with each other. The high time-resolution mobile measurements allow for the separation of the sources of VOCs from different parts of the operations occurring within the facilities. We show that the emissions of ethanol are primarily associated with feed storage and handling. Based on mobile laboratory measurements, we apply a multivariate regression analysis using NH_3 and ethanol as tracers to determine the relative importance of animal-related emissions (animal exhalation and waste) and feed-related emissions (feed storage and handling) for different VOC species. Feed storage and handling contribute significantly to emissions of alcohols, carbonyls, carboxylic acids and sulfur-containing species. Emissions of phenolic species and nitrogen-containing species are predominantly associated with animals and their waste.

Multi-pollutant emissions from the burning of major agricultural residues in China and the related health-economic effects

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Source: Atmos. Chem. Phys., 17, 4957-4988, 2017
<https://doi.org/10.5194/acp-17-4957-2017>

Multi-pollutants in smoke particulate matter (SPM) were identified and quantified for the biomass burning of five major agricultural residues (wheat, rice, corn, cotton, and soybean straw) in China by an aerosol chamber system combined with various measurement techniques. The primary emission factors (EFs) for PM_{1.0} and PM_{2.5} are 3.04–12.64 and 3.25–15.16 g kg⁻¹. Organic carbon (OC), elemental carbon (EC), water-soluble inorganics (WSIs), water-soluble organic acids (WSOAs), water-soluble amine salts (WSAs), trace mineral elements (THMs), polycyclic aromatic hydrocarbons (PAHs), and phenols in smoke PM_{1.0}/PM_{2.5} are 1.34–6.04/1.54–7.42, 0.58–2.08/0.61–2.18, 0.51–3.52/0.52–3.81, 0.13–0.64/0.14–0.77, (4.39–85.72/4.51–104.79) × 10⁻³, (11.8–51.1/14.0–131.6) × 10⁻³, (1.1–4.0/1.8–8.3) × 10⁻³, and (7.7–23.5/9.7–41.5) × 10⁻³ g kg⁻¹, respectively. Black carbon (BC) mainly exists in PM_{1.0}; heavy-metal-bearing particles favour residing in the range of smoke PM_{1.0}–2.5, which is also confirmed by individual particle analysis.

With respect to the five scenarios of burning activities, the average emissions and overall propagation of uncertainties at the 95 % confidence interval (CI) of SPM from agricultural open burning in China in 2012 were estimated to be 1005.7 (–24.6, 33.7 %), 901.4 (–24.4, 33.5 %), 432.4 (–24.2, 33.5 %), 134.2 (–24., 34.0 %), 249.8 (–25.4, 34.9 %), 25.1 (–33.3, 41.4 %), 5.8 (–30.1, 38.5 %), 8.7 (–26.6, 35.6 %), 0.5 (–26.0, 34.9 %), and 2.7 (–26.1, 35.1 %) Gg for PM_{2.5}, PM_{1.0}, OC, EC, WSI, WSOA, WSA, THM, PAHs, and phenols, respectively. The emissions were further spatio-temporally characterized using a geographic information system (GIS) in different regions in the summer and autumn post-harvest periods. It was found that less than 25 % of the total emissions were released during the summer harvest, which was mainly contributed by the North Plain and the centre of China, especially Henan, Shandong, and Anhui, which are the top three provinces regarding smoke particle emissions.

Flux concentrations of primarily emitted smoke PM_{2.5} that were calculated using the box-model method based on five versions of emission inventories all exceed the carcinogenic-risk permissible exposure limits (PEL). The health impacts and health-related economic losses from the smoke PM_{2.5} short-term exposure were assessed. The results show that China suffered from 7836 cases (95 % CI: 3232, 12362) of premature mortality and 7 267 237 cases (95 % CI: 2 961 487, 11 130 784) of chronic bronchitis in 2012, which led to losses of USD 8822.4 million (95 % CI: 3574.4, 13 034.2) or 0.1 % of the total GDP. We suggest that the percentage of open-burnt crop straw in the post-harvest period should be cut down by over 97 % to ensure a reduction in carcinogenicity risk, especially in the North Plain and the northeast, where the emissions should decrease at least by 94 % to meet the PEL. With such emission control, over 92 % of the mortality and morbidity attributed to agricultural fire smoke PM_{2.5} can be avoided in China.

Impact of temperature dependence on the possible contribution of organics to new particle formation in the atmosphere

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Source: *Atmos. Chem. Phys.*, 17, 4997-5005, 2017
<https://doi.org/10.5194/acp-17-4997-2017>

Secondary particles formed via new particle formation (NPF) dominate cloud condensation nuclei (CCN) abundance in most parts of the troposphere and are important for aerosol indirect radiative forcing (IRF). Laboratory measurements have shown that certain organic compounds can significantly enhance the binary nucleation of sulfuric acid and H₂O. According to our recent study comparing particle size distributions measured in nine forest areas in North America with those predicted by a global size-resolved aerosol model, current H₂SO₄-organics nucleation parameterizations appear to significantly overpredict NPF and particle number concentrations in summer. The lack of temperature dependence in the current H₂SO₄-organics nucleation parameterization has been suggested to be a possible reason for the observed overprediction. In this work, H₂SO₄-organics clustering thermodynamics from quantum chemical studies has been employed to develop a scheme to incorporate temperature dependence into H₂SO₄-organics nucleation parameterization. We show that temperature has a strong impact on H₂SO₄-organics nucleation rates and may reduce the nucleation rate by ~ 1 order of magnitude per 10 K of temperature increase. The particle number concentrations in summer over North America based on the revised scheme is a factor of more than 2 lower, which is in much better agreement with the observations. With the temperature-dependent H₂SO₄-organics nucleation parameterization, the summer CCN concentrations in the lower troposphere in the Northern Hemisphere are about 10–30 % lower compared to the temperature-independent parameterization. This study highlights the importance of the temperature effect and its impacts on NPF in the global modeling of aerosol number abundance.

Validation of OMI, GOME-2A and GOME-2B tropospheric NO₂, SO₂ and HCHO products using MAX-DOAS observations from 2011 to 2014 in Wuxi, China: investigation of the effects of priori profiles and aerosols on the satellite products

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Source: *Atmos. Chem. Phys.*, 17, 5007-5033, 2017
<https://doi.org/10.5194/acp-17-5007-2017>

Tropospheric vertical column densities (VCDs) of NO₂, SO₂ and HCHO derived from the Ozone Monitoring Instrument (OMI) on AURA and the Global Ozone Monitoring Experiment 2 aboard METOP-A (GOME-2A) and METOP-B (GOME-2B) are widely used to characterize the global

distributions, trends and dominating sources of these trace gases. They are also useful for the comparison with chemical transport models (CTMs). We use tropospheric VCDs and vertical profiles of NO₂, SO₂ and HCHO derived from MAX-DOAS measurements from 2011 to 2014 in Wuxi, China, to validate the corresponding products (daily and bi-monthly-averaged data) derived from OMI and GOME-2A/B by different scientific teams. Prior to the comparison, the spatial and temporal coincidence criteria for MAX-DOAS and satellite data are determined by a sensitivity study using different spatial and temporal averaging conditions. Cloud effects on both MAX-DOAS and satellite observations are also investigated. Our results indicate that the discrepancies between satellite and MAX-DOAS results increase with increasing effective cloud fraction and are dominated by the effects of clouds on the satellite products. In comparison with MAX-DOAS, we found a systematic underestimation of all SO₂ (40 to 57 %) and HCHO products (about 20 %), and an overestimation of the GOME-2A/B NO₂ products (about 30 %), but good consistency with the DOMINO version 2 NO₂ product. To better understand the reasons for these differences, we evaluated the a priori profile shapes used in the OMI retrievals (derived from CTM) by comparison with those derived from the MAX-DOAS observations. Significant differences are found for the SO₂ and HCHO profile shapes derived from the IMAGES model, whereas on average good agreement is found for the NO₂ profile shapes derived from the TM4 model. We also applied the MAX-DOAS profile shapes to the satellite retrievals and found that these modified satellite VCDs agree better with the MAX-DOAS VCDs than the VCDs from the original data sets by up to 10, 47 and 35 % for NO₂, SO₂ and HCHO, respectively. Furthermore, we investigated the effect of aerosols on the satellite retrievals. For OMI observations of NO₂, a systematic underestimation is found for large AOD, which is mainly attributed to effect of the aerosols on the cloud retrieval and the subsequent application of a cloud correction scheme (implicit aerosol correction). In contrast, the effect of aerosols on the clear-sky air mass factor (explicit aerosol correction) has a smaller effect. For SO₂ and HCHO observations selected in the same way, no clear aerosol effect is found, probably because for the considered data sets no cloud correction is applied (and also because of the larger scatter). From our findings we conclude that for satellite observations with cloud top pressure (CTP) > 900 hPa and effective cloud fraction (eCF) < 10 % the application of a clear-sky air mass factor might be a good option if accurate aerosol information is not available. Another finding of our study is that the ratio of morning-to-afternoon NO₂ VCDs can be considerably overestimated if results from different sensors and/or retrievals (e.g. OMI and GOME-2) are used, whereas fewer deviations for HCHO and SO₂ VCDs are found.

Relative importance of black carbon, brown carbon, and absorption enhancement from clear coatings in biomass burning emissions

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Source: *Atmos. Chem. Phys.*, 17, 5063-5078, 2017
<https://doi.org/10.5194/acp-17-5063-2017>

A wide range of globally significant biomass fuels were burned during the fourth Fire Lab at Missoula Experiment (FLAME-4). A multi-channel photoacoustic absorption spectrometer (PAS)

measured dry absorption at 405, 532, and 660 nm and thermally denuded (250 °C) absorption at 405 and 660 nm. Absorption coefficients were broken into contributions from black carbon (BC), brown carbon (BrC), and lensing following three different methodologies, with one extreme being a method that assumes the thermal denuder effectively removes organics and the other extreme being a method based on the assumption that black carbon (BC) has an Ångström exponent of unity. The methodologies employed provide ranges of potential importance of BrC to absorption but, on average, there was a difference of a factor of 2 in the ratio of the fraction of absorption attributable to BrC estimated by the two methods. BrC absorption at shorter visible wavelengths is of equal or greater importance to that of BC, with maximum contributions of up to 92 % of total aerosol absorption at 405 nm and up to 58 % of total absorption at 532 nm. Lensing is estimated to contribute a maximum of 30 % of total absorption, but typically contributes much less than this. Absorption enhancements and the estimated fraction of absorption from BrC show good correlation with the elemental-carbon-to-organic-carbon ratio (EC/OC) of emitted aerosols and weaker correlation with the modified combustion efficiency (MCE). Previous studies have shown that BrC grows darker (larger imaginary refractive index) as the ratio of black to organic aerosol (OA) mass increases. This study is consistent with those findings but also demonstrates that the fraction of total absorption attributable to BrC shows the opposite trend: increasing as the organic fraction of aerosol emissions increases and the EC/OC ratio decreases.

Real-time aerosol optical properties, morphology and mixing states under clear, haze and fog episodes in the summer of urban Beijing

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Source: Atmos. Chem. Phys., 17, 5079-5093, 2017
<https://doi.org/10.5194/acp-17-5079-2017>

Elucidating the relationship between characteristics of aerosol particles and optical absorption is important to deepen the understanding of atmospheric chemistry. Aerosol particles play significant roles in climate forcing via their optical absorption properties. However, the relationship between characteristics of aerosol particles and optical absorption remains poorly understood. Aerosol optical properties and morphologies were measured by a transmission electron microscope (TEM), cavity ring-down spectrometer (CRDS), a nephelometer and an Aethalometer in a urban site of Beijing from 24 May to 22 June. Five episodes were categorized according to the meteorological conditions and composition. The results showed that the clear episode (EP-2 and EP-4) featured as the low aerosol optical depth (AOD = 0.72) and fewer pollutants compared with haze (1.14) and fog (2.92) episodes and the particles are mostly externally mixed. The high Ångström exponent (> 2.0) suggests that coarse particles were scarcely observed in EP-2 due to the washout of a previous heavy rain, whereas they were widespread in EP-4 (Ångström exponent = 0.04), which had some mineral particles introduced from the north. In contrast, industry-induced haze (EP-1) and biomass-burning-induced haze (EP-5) were both affected by the south air mass. Compared with the EP-2 and EP-4, the AOD values and the size distribution of particles during EP-1 and EP-5 were much greater because of relatively high particle concentrations. All of the particles were classified into nine categories, i.e. S-rich, N-rich, mineral, K-rich, soot, tar ball, organic, metal and fly ash, on the basis of TEM analysis. In contrast to the EP-1, a large fraction of soot, which sticks to KCl, sulfate or nitrate particles, was detected during EP-5. Additionally, evident enhancement of light

absorption was observed during the EP-5, which was mainly ascribed to both black carbon (BC) acceleration and other absorbing substances. However, soot was found mostly internally mixed with sulfate and nitrate during a soot fog episode (EP-3), resulting in evident enhancement of light absorption. The larger size distribution was likely to be caused by both hygroscopic growth and collision between particles during the aging. About 28 % of particles were internally mixed during the foggy days, which favoured the light absorption. The comparison of all the episodes provides a deeper insight into how mixing states influence the aerosol extinction properties and also a clue as to how to control air pollution in the crop burning seasons.

Biophysical effects on the interannual variation in carbon dioxide exchange of an alpine meadow on the Tibetan Plateau

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Source: Atmos. Chem. Phys., 17, 5119-5129, 2017
<https://doi.org/10.5194/acp-17-5119-2017>

Eddy covariance measurements from 2012 to 2015 were used to investigate the interannual variation in carbon dioxide exchange and its control over an alpine meadow on the south-east margin of the Tibetan Plateau. The annual net ecosystem exchange (NEE) in the 4 years from 2012 to 2015 was -114.2 , -158.5 , -159.9 and -212.6 g C m⁻² yr⁻¹, and generally decreased with the mean annual air temperature (MAT). An exception occurred in 2014, which had the highest MAT. This was attributed to higher ecosystem respiration (RE) and similar gross primary production (GPP) in 2014 because the GPP increased with the MAT, but became saturated due to the limit in photosynthetic capacity. In the spring (March to May) of 2012, low air temperature (Ta) and drought events delayed grass germination and reduced GPP. In the late wet season (September to October) of 2012 and 2013, the low Ta in September and its negative effects on vegetation growth caused earlier grass senescence and significantly lower GPP. This indicates that the seasonal pattern of Ta has a substantial effect on the annual total GPP, which is consistent with results obtained using the homogeneity-of-slopes (HOS) model. The model results showed that the climatic seasonal variation explained 48.6 % of the GPP variability, while the percentages explained by climatic interannual variation and the ecosystem functional change were 9.7 and 10.6 %, respectively.

The Climatology of Australian Aerosol

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Source: Atmos. Chem. Phys., 17, 5131-5154, 2017
<https://doi.org/10.5194/acp-17-5131-2017>

Airborne particles or aerosols have long been recognised for their major contribution to uncertainty in climate change. In addition, aerosol amounts must be known for accurate atmospheric correction of

remotely sensed images, and are required to accurately gauge the available solar resource. However, despite great advances in surface networks and satellite retrievals over recent years, long-term continental-scale aerosol data sets are lacking. Here we present an aerosol assessment over Australia based on combined sun photometer measurements from the Bureau of Meteorology Radiation Network and CSIRO/AeroSpan. The measurements are continental in coverage, comprising 22 stations, and generally decadal in timescale, totalling 207 station-years. Monthly climatologies are given at all stations. Spectral decomposition shows that the time series can be represented as a weighted sum of sinusoids with periods of 12, 6 and 4 months, corresponding to the annual cycle and its second and third harmonics. Their relative amplitudes and phase relationships lead to sawtooth-like waveforms sharply rising to an austral spring peak, with a slower decline often including a secondary peak during the summer. The amplitude and phase of these periodic components show significant regional change across the continent. Fits based on this harmonic analysis are used to separate the periodic and episodic components of the aerosol time series. An exploratory classification of the aerosol types is undertaken based on (a) the relative periodic amplitudes of the Ångström exponent and aerosol optical depth, (b) the relative amplitudes of the 6- and 4-month harmonic components of the aerosol optical depth, and (c) the ratio of episodic to periodic variation in aerosol optical depth. It is shown that Australian aerosol can be broadly grouped into three classes: tropical, arid and temperate. Statistically significant decadal trends are found at 4 of the 22 stations. Despite the apparently small associated declining trends in mid-visible aerosol optical depth of between 0.001 and 0.002 per year, these trends are much larger than those projected to occur due to declining emissions of anthropogenic aerosols from the Northern Hemisphere. There is remarkable long-range coherence in the aerosol cycle across the continent, suggesting broadly similar source characteristics, including a possible role for intercontinental transport of biomass burning aerosol.

Investigation of the aerosol–cloud–rainfall association over the Indian summer monsoon region

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Pai

Source: Atmos. Chem. Phys., 17, 5185-5204, 2017
<https://doi.org/10.5194/acp-17-5185-2017>

Monsoonal rainfall is the primary source of surface water in India. Using 12 years of in situ and satellite observations, we examined the association of aerosol loading with cloud fraction, cloud top pressure, cloud top temperature, and daily surface rainfall over the Indian summer monsoon region (ISMR). Our results showed positive correlations between aerosol loading and cloud properties as well as rainfall. A decrease in outgoing longwave radiation and an increase in reflected shortwave radiation at the top of the atmosphere with an increase in aerosol loading further indicates a possible seminal role of aerosols in the deepening of cloud systems. Significant perturbation in liquid- and ice-phase microphysics was also evident over the ISMR. For the polluted cases, delay in the onset of collision–coalescence processes and an enhancement in the condensation efficiency allows for more condensate mass to be lifted up to the mixed colder phases. This results in the higher mass concentration of larger-sized ice-phase hydrometeors and, therefore, implies that the delayed rain processes eventually lead to more surface rainfall. A numerical simulation of a typical rainfall event case over the ISMR using a spectral bin microphysical scheme coupled with the Weather Research Forecasting (WRF-SBM) model was also

performed. Simulated microphysics also illustrated that the initial suppression of warm rain coupled with an increase in updraft velocity under high aerosol loading leads to enhanced super-cooled liquid droplets above freezing level and ice-phase hydrometeors, resulting in increased accumulated surface rainfall. Thus, both observational and numerical analysis suggest that high aerosol loading may induce cloud invigoration, thereby increasing surface rainfall over the ISMR. While the meteorological variability influences the strength of the observed positive association, our results suggest that the persistent aerosol-associated deepening of cloud systems and an intensification of surface rain amounts was applicable to all the meteorological sub-regimes over the ISMR. Hence, we believe that these results provide a step forward in our ability to address aerosol–cloud–rainfall associations based on satellite observations over the ISMR.

Direct radiative effect of carbonaceous aerosols from crop residue burning during the summer harvest season in East China

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Source: Atmos. Chem. Phys., 17, 5205-5219, 2017
<https://doi.org/10.5194/acp-17-5205-2017>

East China experiences extensive crop residue burnings in fields during harvest season. The direct radiative effect (DRE) of carbonaceous aerosols from crop residue burning in June 2013 in East China was investigated using the Weather Research and Forecasting Model coupled with Chemistry (WRF-Chem). Absorption of organic aerosol (OA) in the presence of brown carbon was considered using the parameterization of Saleh et al. (2014), in which the imaginary part of the OA refractive index is a function of wavelength and the ratio of black carbon (BC) and OA. The carbonaceous emissions from crop fires were estimated using the Moderate Resolution Imaging Spectroradiometer (MODIS) fire radiative power (FRP) product with a localized crop-burning-sourced BC-to-organic carbon (OC) ratio emission ratio of 0.27. Evaluation of the model results with in situ measurements of particulate matter with aerodynamic diameter less than 2.5 μm (PM_{2.5}) chemical composition, MODIS aerosol optical depth (AOD) detections and meteorological observations showed that this model was able to reproduce the magnitude, spatial variation and optical characteristics of carbonaceous aerosol pollution. The observed BC and OC peak concentrations at the site in Suixi, Anhui province, during the 2013 wheat burning season reached 55.3 $\mu\text{g m}^{-3}$ and 157.9 $\mu\text{g m}^{-3}$. WRF-Chem simulations reproduced these trends with a correlation coefficient of 0.74, estimating that crop residue burning contributed 86 and 90 % of peak BC and OC, respectively. The simulated hourly DRE from crop residue burning at the top of atmosphere (TOA) reached a maximum of +22.66 W m^{-2} at the Suixi site. On average, the simulations showed that the crop residue burning introduced a net positive DRE of +0.14 W m^{-2} at TOA throughout East China, with BC from this source as the main heating contributor (+0.79 W m^{-2}). The OA DRE from crop burning (−0.22 W m^{-2}) was a combined effect of the positive DRE of absorption (+0.21 W m^{-2}) and a stronger negative DRE of scattering (−0.43 W m^{-2}). Sensitivity tests showed that the DRE of OA absorption strongly depended on the imaginary part of the OA refractive index, the BC-to-OA emission ratio from crop residue burning and the assumed mixing state of the aerosol, whereby the volume mixing treatment resulted in a higher positive DRE compared to the core–shell treatment. The BC mixing

state and associated absorption enhancement during BC aging processes will be investigated in detail in future research.

Genesis of diamond dust, ice fog and thick cloud episodes observed and modelled above Dome C, Antarctica

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Source: Atmos. Chem. Phys., 17, 5221-5237, 2017
<https://doi.org/10.5194/acp-17-5221-2017>

Episodes of thick cloud and diamond dust/ice fog were observed during 15 March to 8 April 2011 and 4 to 5 March 2013 in the atmosphere above Dome C (Concordia station, Antarctica; 75°06' S, 123°21' E; 3233 m a.m.s.l.). The objectives of the paper are mainly to investigate the processes that cause these episodes based on observations and to verify whether operational models can evaluate them. The measurements were obtained from the following instruments: (1) a ground-based microwave radiometer (HAMSTRAD, H₂O Antarctica Microwave Stratospheric and Tropospheric Radiometers) installed at Dome C that provided vertical profiles of tropospheric temperature and absolute humidity every 7 min; (2) daily radiosoundings launched at 12:00 UTC at Dome C; (3) a tropospheric aerosol lidar that provides aerosol depolarization ratio along the vertical at Dome C; (4) down- and upward short- and long-wave radiations as provided by the Baseline Surface Radiation Network (BSRN) facilities; (5) an ICE-CAMERA to detect at an hourly rate the size of the ice crystal grains deposited at the surface of the camera; and (6) space-borne aerosol depolarization ratio from the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) lidar aboard the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) platform along orbits close to the Dome C station. The time evolution of the atmosphere has also been evaluated by considering the outputs from the mesoscale AROME and the global-scale ARPEGE meteorological models. Thick clouds are detected during the warm and wet periods (24–26 March 2011 and 4 March 2013) with high depolarization ratios (greater than 30 %) from the surface to 5–7 km above the ground associated with precipitation of ice particles and the presence of a supercooled liquid water (depolarization less than 10 %) clouds. Diamond dust and/or ice fog are detected during the cold and dry periods (5 April 2011 and 5 March 2013) with high depolarization ratios (greater than 30 %) in the planetary boundary layer to a maximum altitude of 100–300 m above the ground with little trace of precipitation. Considering 5-day back trajectories, we show that the thick cloud episodes are attributed to air masses with an oceanic origin whilst the diamond dust/ice fog episodes are attributed to air masses with continental origins. Although operational models can reproduce thick cloud episodes in the free troposphere, they cannot evaluate the diamond dust/ice fog episodes in the planetary boundary layer because they require to use more sophisticated cloud and aerosol microphysics schemes.

Urbanization-induced urban heat island and aerosol effects on climate extremes in the Yangtze River Delta region of China

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Source: Atmos. Chem. Phys., 17, 5439-5457, 2017
<https://doi.org/10.5194/acp-17-5439-2017>

The WRF-Chem model coupled with a single-layer urban canopy model (UCM) is integrated for 5 years at convection-permitting scale to investigate the individual and combined impacts of urbanization-induced changes in land cover and pollutant emissions on regional climate in the Yangtze River Delta (YRD) region in eastern China. Simulations with the urbanization effects reasonably reproduced the observed features of temperature and precipitation in the YRD region. Urbanization over the YRD induces an urban heat island (UHI) effect, which increases the surface temperature by 0.53 °C in summer and increases the annual heat wave days at a rate of 3.7 d yr⁻¹ in the major megacities in the YRD, accompanied by intensified heat stress. In winter, the near-surface air temperature increases by approximately 0.7 °C over commercial areas in the cities but decreases in the surrounding areas. Radiative effects of aerosols tend to cool the surface air by reducing net shortwave radiation at the surface. Compared to the more localized UHI effect, aerosol effects on solar radiation and temperature influence a much larger area, especially downwind of the city cluster in the YRD.

Results also show that the UHI increases the frequency of extreme summer precipitation by strengthening the convergence and updrafts over urbanized areas in the afternoon, which favor the development of deep convection. In contrast, the radiative forcing of aerosols results in a surface cooling and upper-atmospheric heating, which enhances atmospheric stability and suppresses convection. The combined effects of the UHI and aerosols on precipitation depend on synoptic conditions. Two rainfall events under two typical but different synoptic weather patterns are further analyzed. It is shown that the impact of urban land cover and aerosols on precipitation is not only determined by their influence on local convergence but also modulated by large-scale weather systems. For the case with a strong synoptic forcing associated with stronger winds and larger spatial convergence, the UHI and aerosol effects are relatively weak. When the synoptic forcing is weak, however, the UHI and aerosol effects on local convergence dominate. This suggests that synoptic forcing plays a significant role in modulating the urbanization-induced land-cover and aerosol effects on individual rainfall event. Hence precipitation changes due to urbanization effects may offset each other under different synoptic conditions, resulting in little changes in mean precipitation at longer timescales.

Secondary organic aerosol formation in biomass-burning plumes: theoretical analysis of lab studies and ambient plumes

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Source: Atmos. Chem. Phys., 17, 5459-5475, 2017
<https://doi.org/10.5194/acp-17-5459-2017>

Secondary organic aerosol (SOA) has been shown to form in biomass-burning emissions in laboratory and field studies. However, there is significant variability among studies in mass enhancement, which could be due to differences in fuels, fire conditions, dilution, and/or limitations of laboratory experiments and observations. This study focuses on understanding processes affecting biomass-burning SOA formation in laboratory smog-chamber experiments and in ambient plumes. Vapor wall losses have been demonstrated to be an important factor that can suppress SOA formation in laboratory studies of traditional SOA precursors; however, impacts of vapor wall losses on biomass-burning SOA have not yet been investigated. We use an aerosol-microphysical model that includes representations of volatility and oxidation chemistry to estimate the influence of vapor wall loss on SOA formation observed in the FLAME III smog-chamber studies. Our simulations with base-case assumptions for chemistry and wall loss predict a mean OA mass enhancement (the ratio of final to initial OA mass, corrected for particle-phase wall losses) of 1.8 across all experiments when vapor wall losses are modeled, roughly matching the mean observed enhancement during FLAME III. The mean OA enhancement increases to over 3 when vapor wall losses are turned off, implying that vapor wall losses reduce the apparent SOA formation. We find that this decrease in the apparent SOA formation due to vapor wall losses is robust across the ranges of uncertainties in the key model assumptions for wall-loss and mass-transfer coefficients and chemical mechanisms.

We then apply similar assumptions regarding SOA formation chemistry and physics to smoke emitted into the atmosphere. In ambient plumes, the plume dilution rate impacts the organic partitioning between the gas and particle phases, which may impact the potential for SOA to form as well as the rate of SOA formation. We add Gaussian dispersion to our aerosol-microphysical model to estimate how SOA formation may vary under different ambient-plume conditions (e.g., fire size, emission mass flux, atmospheric stability). Smoke from small fires, such as typical prescribed burns, dilutes rapidly, which drives evaporation of organic vapor from the particle phase, leading to more effective SOA formation. Emissions from large fires, such as intense wildfires, dilute slowly, suppressing OA evaporation and subsequent SOA formation in the near field. We also demonstrate that different approaches to the calculation of OA enhancement in ambient plumes can lead to different conclusions regarding SOA formation. OA mass enhancement ratios of around 1 calculated using an inert tracer, such as black carbon or CO, have traditionally been interpreted as exhibiting little or no SOA formation; however, we show that SOA formation may have greatly contributed to the mass in these plumes.

In comparison of laboratory and plume results, the possible inconsistency of OA enhancement between them could be in part attributed to the effect of chamber walls and plume dilution. Our results highlight that laboratory and field experiments that focus on the fuel and fire conditions also need to consider the effects of plume dilution or vapor losses to walls.

How can mountaintop CO₂ observations be used to constrain regional carbon fluxes?

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Source: Atmos. Chem. Phys., 17, 5561-5581, 2017
<https://doi.org/10.5194/acp-17-5561-2017>

Despite the need for researchers to understand terrestrial biospheric carbon fluxes to account for carbon cycle feedbacks and predict future CO₂ concentrations, knowledge of these fluxes at the regional scale remains poor. This is particularly true in mountainous areas, where complex meteorology and lack of observations lead to large uncertainties in carbon fluxes. Yet mountainous regions are often where significant forest cover and biomass are found – i.e., areas that have the potential to serve as carbon sinks. As CO₂ observations are carried out in mountainous areas, it is imperative that they are properly interpreted to yield information about carbon fluxes. In this paper, we present CO₂ observations at three sites in the mountains of the western US, along with atmospheric simulations that attempt to extract information about biospheric carbon fluxes from the CO₂ observations, with emphasis on the observed and simulated diurnal cycles of CO₂. We show that atmospheric models can systematically simulate the wrong diurnal cycle and significantly misinterpret the CO₂ observations, due to erroneous atmospheric flows as a result of terrain that is misrepresented in the model. This problem depends on the selected vertical level in the model and is exacerbated as the spatial resolution is degraded, and our results indicate that a fine grid spacing of ~4 km or less may be needed to simulate a realistic diurnal cycle of CO₂ for sites on top of the steep mountains examined here in the American Rockies. In the absence of higher resolution models, we recommend coarse-scale models to focus on assimilating afternoon CO₂ observations on mountaintop sites over the continent to avoid misrepresentations of nocturnal transport and influence.

Source-sector contributions to European ozone and fine PM in 2010 using AQMEII modeling data

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Source: Atmos. Chem. Phys., 17, 5643-5664, 2017
<https://doi.org/10.5194/acp-17-5643-2017>

Source apportionment modeling provides valuable information on the contributions of different source sectors and/or source regions to ozone (O₃) or fine particulate matter (PM_{2.5}) concentrations. This information can be useful in designing air quality management strategies and in understanding the potential benefits of reducing emissions from a particular source category. The Comprehensive Air quality Model with Extensions (CAMx) offers unique source attribution tools, called the Ozone and Particulate Source Apportionment Technology (OSAT/PSAT), which track source contributions. We present results from a CAMx source attribution modeling study for a summer month and a winter month using a recently evaluated European CAMx modeling database developed for Phase 3 of the Air Quality Model Evaluation International Initiative (AQMEII). The contributions of several source sectors (including model boundary conditions of chemical species representing transport of emissions from outside the modeling domain as well as initial conditions of these species) to O₃ or PM_{2.5} concentrations in Europe were calculated using OSAT and PSAT, respectively. A 1-week spin-up period was used to reduce the influence of initial conditions. Evaluation focused on 16 major cities and

on identifying source sectors that contributed above 5 %. Boundary conditions have a large impact on summer and winter ozone in Europe and on summer PM_{2.5}, but they are only a minor contributor to winter PM_{2.5}. Biogenic emissions are important for summer ozone and PM_{2.5}. The important anthropogenic sectors for summer ozone are transportation (both on-road and non-road), energy production and conversion, and industry. In two of the 16 cities, solvent and product also contributed above 5 % to summertime ozone. For summertime PM_{2.5}, the important anthropogenic source sectors are energy, transportation, industry, and agriculture. Residential wood combustion is an important anthropogenic sector in winter for PM_{2.5} over most of Europe, with larger contributions in central and eastern Europe and the Nordic cities. Other anthropogenic sectors with large contributions to wintertime PM_{2.5} include energy, transportation, and agriculture.

The constraint of CO₂ measurements made onboard passenger aircraft on surface–atmosphere fluxes: the impact of transport model errors in vertical mixing

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Source: Atmos. Chem. Phys., 17, 5665–5675, 2017
<https://doi.org/10.5194/acp-17-5665-2017>

Inaccurate representation of atmospheric processes by transport models is a dominant source of uncertainty in inverse analyses and can lead to large discrepancies in the retrieved flux estimates. We investigate the impact of uncertainties in vertical transport as simulated by atmospheric transport models on fluxes retrieved using vertical profiles from aircraft as an observational constraint. Our numerical experiments are based on synthetic data with realistic spatial and temporal sampling of aircraft measurements. The impact of such uncertainties on the flux retrieved using the ground-based network and those retrieved using the aircraft profiles are compared. We find that the posterior flux retrieved using aircraft profiles is less susceptible to errors in boundary layer height, compared to the ground-based network. This finding highlights a benefit of utilizing atmospheric observations made onboard aircraft over surface measurements for flux estimation using inverse methods. We further use synthetic vertical profiles of CO₂ in an inversion to estimate the potential of these measurements, which will be made available through the IAGOS (In-service Aircraft for a Global Observing System) project in the future, in constraining the regional carbon budget. Our results show that the regions of tropical Africa and temperate Eurasia, that are under-constrained by the existing surface-based network, will benefit the most from these measurements, with a reduction of posterior flux uncertainty of about 7 to 10 %.

The radiative role of ozone and water vapour in the annual temperature cycle in the tropical tropopause layer

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Source: Atmos. Chem. Phys., 17, 5677-5701, 2017
<https://doi.org/10.5194/acp-17-5677-2017>

The structure and amplitude of the radiative contributions of the annual cycles in ozone and water vapour to the prominent annual cycle in temperatures in the tropical tropopause layer (TTL) are considered. This is done initially through a seasonally evolving fixed dynamical heating (SEFDH) calculation. The annual cycle in ozone is found to drive significant temperature changes predominantly locally (in the vertical) and roughly in phase with the observed TTL annual cycle. In contrast, temperature changes driven by the annual cycle in water vapour are out of phase with the latter. The effects are weaker than those of ozone but still quantitatively significant, particularly near the cold point (100 to 90 hPa) where there are substantial non-local effects from variations in water vapour in lower layers of the TTL. The combined radiative heating effect of the annual cycles in ozone and water vapour maximizes above the cold point and is one factor contributing to the vertical structure of the amplitude of the annual cycle in lower-stratospheric temperatures, which has a relatively localized maximum around 70 hPa. Other important factors are identified here: radiative damping timescales, which are shown to maximize over a deep layer centred on the cold point; the vertical structure of the dynamical heating; and non-radiative processes in the upper troposphere that are inferred to impose a strong constraint on tropical temperature perturbations below 130 hPa. The latitudinal structure of the radiative contributions to the annual cycle in temperatures is found to be substantially modified when the SEFDH assumption is relaxed and the dynamical response, as represented by a zonally symmetric calculation, is taken into account. The effect of the dynamical response is to reduce the strong latitudinal gradients and inter-hemispheric asymmetry seen in the purely radiative SEFDH temperature response, while leaving the 20° N–20° S average response relatively unchanged. The net contribution of the annual ozone and water vapour cycles to the peak-to-peak amplitude in the annual cycle of TTL temperatures is found to be around 35 % of the observed 8 K at 70 hPa, 40 % of 6 K at 90 hPa, and 45 % of 3 K at 100 hPa. The primary sensitivity of the calculated magnitude of the temperature response is identified as the assumed annual mean ozone mixing ratio in the TTL.

Impact of intercontinental pollution transport on North American ozone air pollution: an HTAP phase 2 multi-model study

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Source: Atmos. Chem. Phys., 17, 5721-5750, 2017
<https://doi.org/10.5194/acp-17-5721-2017>

The recent update on the US National Ambient Air Quality Standards (NAAQS) of the ground-level ozone (O₃) can benefit from a better understanding of its source contributions in different US regions during recent years. In the Hemispheric Transport of Air Pollution experiment phase 1 (HTAP1), various global models were used to determine the O₃ source–receptor (SR) relationships among three continents in the Northern Hemisphere in 2001. In support of the HTAP phase 2 (HTAP2) experiment that studies more recent years and involves higher-resolution global models and regional models' participation, we

conduct a number of regional-scale Sulfur Transport and dEposition Model (STEM) air quality base and sensitivity simulations over North America during May–June 2010. STEM's top and lateral chemical boundary conditions were downscaled from three global chemical transport models' (i.e., GEOS-Chem, RAQMS, and ECMWF C-IFS) base and sensitivity simulations in which the East Asian (EAS) anthropogenic emissions were reduced by 20 %. The mean differences between STEM surface O₃ sensitivities to the emission changes and its corresponding boundary condition model's are smaller than those among its boundary condition models, in terms of the regional/period-mean (< 10 %) and the spatial distributions. An additional STEM simulation was performed in which the boundary conditions were downscaled from a RAQMS (Realtime Air Quality Modeling System) simulation without EAS anthropogenic emissions. The scalability of O₃ sensitivities to the size of the emission perturbation is spatially varying, and the full (i.e., based on a 100 % emission reduction) source contribution obtained from linearly scaling the North American mean O₃ sensitivities to a 20 % reduction in the EAS anthropogenic emissions may be underestimated by at least 10 %. The three boundary condition models' mean O₃ sensitivities to the 20 % EAS emission perturbations are ~8 % (May–June 2010)/~11 % (2010 annual) lower than those estimated by eight global models, and the multi-model ensemble estimates are higher than the HTAP1 reported 2001 conditions. GEOS-Chem sensitivities indicate that the EAS anthropogenic NO_x emissions matter more than the other EAS O₃ precursors to the North American O₃, qualitatively consistent with previous adjoint sensitivity calculations.

In addition to the analyses on large spatial–temporal scales relative to the HTAP1, we also show results on subcontinental and event scales that are more relevant to the US air quality management. The EAS pollution impacts are weaker during observed O₃ exceedances than on all days in most US regions except over some high-terrain western US rural/remote areas. Satellite O₃ (TES, JPL–IASI, and AIRS) and carbon monoxide (TES and AIRS) products, along with surface measurements and model calculations, show that during certain episodes stratospheric O₃ intrusions and the transported EAS pollution influenced O₃ in the western and the eastern US differently. Free-running (i.e., without chemical data assimilation) global models underpredicted the transported background O₃ during these episodes, posing difficulties for STEM to accurately simulate the surface O₃ and its source contribution. Although we effectively improved the modeled O₃ by incorporating satellite O₃ (OMI and MLS) and evaluated the quality of the HTAP2 emission inventory with the Royal Netherlands Meteorological Institute–Ozone Monitoring Instrument (KNMI–OMI) nitrogen dioxide, using observations to evaluate and improve O₃ source attribution still remains to be further explored.

Satellite-derived methane hotspot emission estimates using a fast data-driven method

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Source: Atmos. Chem. Phys., 17, 5751–5774, 2017
<https://doi.org/10.5194/acp-17-5751-2017>

Methane is an important atmospheric greenhouse gas and an adequate understanding of its emission sources is needed for climate change assessments, predictions, and the development and verification of emission mitigation strategies. Satellite retrievals of near-surface-sensitive column-averaged dry-air mole fractions of atmospheric methane, i.e. XCH₄, can be used to quantify methane emissions. Maps of time-averaged satellite-derived XCH₄ show regionally elevated methane over several methane source regions. In order to obtain methane emissions of these source regions we use a simple and fast data-driven method to estimate annual methane emissions and corresponding 1 σ uncertainties directly from maps of annually averaged satellite XCH₄. From theoretical considerations we expect that our method tends to underestimate emissions. When applying our method to high-resolution atmospheric methane simulations, we typically find agreement within the uncertainty range of our method (often 100 %) but also find that our method tends to underestimate emissions by typically about 40 %. To what extent these findings are model dependent needs to be assessed. We apply our method to an ensemble of satellite XCH₄ data products consisting of two products from SCIAMACHY/ENVISAT and two products from TANSO-FTS/GOSAT covering the time period 2003–2014. We obtain annual emissions of four source areas: Four Corners in the south-western USA, the southern part of Central Valley, California, Azerbaijan, and Turkmenistan. We find that our estimated emissions are in good agreement with independently derived estimates for Four Corners and Azerbaijan. For the Central Valley and Turkmenistan our estimated annual emissions are higher compared to the EDGAR v4.2 anthropogenic emission inventory. For Turkmenistan we find on average about 50 % higher emissions with our annual emission uncertainty estimates overlapping with the EDGAR emissions. For the region around Bakersfield in the Central Valley we find a factor of 5–8 higher emissions compared to EDGAR, albeit with large uncertainty. Major methane emission sources in this region are oil/gas and livestock. Our findings corroborate recently published studies based on aircraft and satellite measurements and new bottom-up estimates reporting significantly underestimated methane emissions of oil/gas and/or livestock in this area in EDGAR.

Observations of atmospheric chemical deposition to high Arctic snow

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Source: Atmos. Chem. Phys., 17, 5775-5788, 2017
<https://doi.org/10.5194/acp-17-5775-2017>

Rapidly rising temperatures and loss of snow and ice cover have demonstrated the unique vulnerability of the high Arctic to climate change. There are major uncertainties in modelling the chemical depositional and scavenging processes of Arctic snow. To that end, fresh snow samples collected on average every 4 days at Alert, Nunavut, from September 2014 to June 2015 were analyzed for black carbon, major ions, and metals, and their concentrations and fluxes were reported. Comparison with simultaneous measurements of atmospheric aerosol mass loadings yields effective deposition velocities that encompass all processes by which the atmospheric species are transferred to the snow. It is inferred from these values that dry deposition is the dominant removal mechanism for several

compounds over the winter while wet deposition increased in importance in the fall and spring, possibly due to enhanced scavenging by mixed-phase clouds. Black carbon aerosol was the least efficiently deposited species to the snow.

Clouds over the summertime Sahara: an evaluation of Met Office retrievals from Meteosat Second Generation using airborne remote sensing

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Source: Atmos. Chem. Phys., 17, 5789-5807, 2017
<https://doi.org/10.5194/acp-17-5789-2017>

Novel methods of cloud detection are applied to airborne remote sensing observations from the unique Fennec aircraft dataset, to evaluate the Met Office-derived products on cloud properties over the Sahara based on the Spinning Enhanced Visible and InfraRed Imager (SEVIRI) on-board the Meteosat Second Generation (MSG) satellite. Two cloud mask configurations are considered, as well as the retrievals of cloud-top height (CTH), and these products are compared to airborne cloud remote sensing products acquired during the Fennec campaign in June 2011 and June 2012. Most detected clouds (67 % of the total) have a horizontal extent that is smaller than a SEVIRI pixel (3 km × 3 km). We show that, when partially cloud-contaminated pixels are included, a match between the SEVIRI and aircraft datasets is found in 80 ± 8 % of the pixels. Moreover, under clear skies the datasets are shown to agree for more than 90 % of the pixels. The mean cloud field, derived from the satellite cloud mask acquired during the Fennec flights, shows that areas of high surface albedo and orography are preferred sites for Saharan cloud cover, consistent with published theories. Cloud-top height retrievals however show large discrepancies over the region, which are ascribed to limiting factors such as the cloud horizontal extent, the derived effective cloud amount, and the absorption by mineral dust. The results of the CTH analysis presented here may also have further-reaching implications for the techniques employed by other satellite applications facilities across the world.

Comparisons of ground-based tropospheric NO₂ MAX-DOAS measurements to satellite observations with the aid of an air quality model over the Thessaloniki area, Greece

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Source: Atmos. Chem. Phys., 17, 5829-5849, 2017
<https://doi.org/10.5194/acp-17-5829-2017>

One of the main issues arising from the comparison of ground-based and satellite measurements is the difference in spatial representativeness, which for locations with inhomogeneous spatial distribution of pollutants may lead to significant differences between the two data sets. In order to investigate the spatial variability of tropospheric NO₂ within a sub-satellite pixel, a campaign which lasted for about 6 months was held in the greater area of Thessaloniki, Greece. Three multi-axial differential optical absorption spectroscopy (MAX-DOAS) systems performed measurements of tropospheric NO₂ columns at different sites representative of urban, suburban and rural conditions. The direct comparison of these ground-based measurements with corresponding products from the Ozone Monitoring Instrument onboard NASA's Aura satellite (OMI/Aura) showed good agreement over the rural and suburban areas, while the comparison with the Global Ozone Monitoring Experiment-2 (GOME-2) onboard EUMETSAT's Meteorological Operational satellites' (MetOp-A and MetOp-B) observations is good only over the rural area. GOME-2A and GOME-2B sensors show an average underestimation of tropospheric NO₂ over the urban area of about $10.51 \pm 8.32 \times 10^{15}$ and $10.21 \pm 8.87 \times 10^{15}$ molecules cm⁻², respectively. The mean difference between ground-based and OMI observations is significantly lower ($6.60 \pm 5.71 \times 10^{15}$ molecules cm⁻²). The differences found in the comparisons of MAX-DOAS data with the different satellite sensors can be attributed to the higher spatial resolution of OMI, as well as the different overpass times and NO₂ retrieval algorithms of the satellites. OMI data were adjusted using factors calculated by an air quality modeling tool, consisting of the Weather Research and Forecasting (WRF) mesoscale meteorological model and the Comprehensive Air Quality Model with Extensions (CAMx) multiscale photochemical transport model. This approach resulted in significant improvement of the comparisons over the urban monitoring site. The average difference of OMI observations from MAX-DOAS measurements was reduced to $-1.68 \pm 5.01 \times 10^{15}$ molecules cm⁻².

Alteration of the size distributions and mixing states of black carbon through transport in the boundary layer in east Asia

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Source: Atmos. Chem. Phys., 17, 5851-5864, 2017
<https://doi.org/10.5194/acp-17-5851-2017>

Ground-based measurements of black carbon (BC) were performed near an industrial source region in the early summer of 2014 and at a remote island in Japan in the spring of 2015. Here, we report the temporal variations in the transport, size distributions, and mixing states of the BC-containing particles. These particles were characterized using a continuous soot monitoring system, a single particle soot photometer, and an aerosol chemical speciation monitor. The effects of aging on the growth of BC-containing particles were examined by comparing the ground-based observations between the near-source and remote island sites. Secondary formation of sulfate and organic aerosols strongly affected the increases in BC coating (i.e., enhancement of cloud condensation nuclei activity) with air mass aging from the source to the outflow regions. The effects of wet removal on BC microphysics were elucidated by classifying the continental outflow air masses depending on the enhancement ratios of BC to CO ($\Delta\text{BC}/\Delta\text{CO}$), which were used as an indicator of the transport efficiency of BC. It was found that $\Delta\text{BC}/\Delta\text{CO}$ ratios were controlled mainly by the wet removal during transport in the planetary boundary

layer (PBL) on the timescale of 1–2 days. The meteorological conditions and backward trajectory analyses suggested that air masses strongly affected by wet removal originated mainly from a region in southern China (20–35° N) in the spring of 2015. Removal of large and thickly coated BC-containing particles was detected in the air masses that were substantially affected by the wet removal in the PBL, as predicted by Köhler theory. The size and water solubility of BC-containing particles in the PBL can be altered by the wet removal as well as the condensation of non-BC materials.

Sensitivity of black carbon concentrations and climate impact to aging and scavenging in OsloCTM2–M7

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Source: Atmos. Chem. Phys., 17, 6003–6022, 2017
<https://doi.org/10.5194/acp-17-6003-2017>

Accurate representation of black carbon (BC) concentrations in climate models is a key prerequisite for understanding its net climate impact. BC aging and scavenging are treated very differently in current models. Here, we examine the sensitivity of three-dimensional (3-D), temporally resolved BC concentrations to perturbations to individual model processes in the chemistry transport model OsloCTM2–M7. The main goals are to identify processes related to aerosol aging and scavenging where additional observational constraints may most effectively improve model performance, in particular for BC vertical profiles, and to give an indication of how model uncertainties in the BC life cycle propagate into uncertainties in climate impacts. Coupling OsloCTM2 with the microphysical aerosol module M7 allows us to investigate aging processes in more detail than possible with a simpler bulk parameterization. Here we include, for the first time in this model, a treatment of condensation of nitric acid on BC. Using kernels, we also estimate the range of radiative forcing and global surface temperature responses that may result from perturbations to key tunable parameters in the model. We find that BC concentrations in OsloCTM2–M7 are particularly sensitive to convective scavenging and the inclusion of condensation by nitric acid. The largest changes are found at higher altitudes around the Equator and at low altitudes over the Arctic. Convective scavenging of hydrophobic BC, and the amount of sulfate required for BC aging, are found to be key parameters, potentially reducing bias against HIPPO Pole-to-Pole Observations (HIPPO) flight-based measurements by 60 to 90 %. Even for extensive tuning, however, the total impact on global-mean surface temperature is estimated to less than 0.04 K. Similar results are found when nitric acid is allowed to condense on the BC aerosols. We conclude, in line with previous studies, that a shorter atmospheric BC lifetime broadly improves the comparison with measurements over the Pacific. However, we also find that the model–measurement discrepancies can not be uniquely attributed to uncertainties in a single process or parameter. Model development therefore needs to be focused on improvements to individual processes, supported by a broad range of observational and experimental data, rather than tuning of individual, effective parameters such as the global BC lifetime.

Downward particle fluxes of biogenic matter and Saharan dust across the equatorial North Atlantic

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Source: Atmos. Chem. Phys., 17, 6023-6040, 2017
<https://doi.org/10.5194/acp-17-6023-2017>

Massive amounts of Saharan dust are blown from the coast of northern Africa across the Atlantic Ocean towards the Americas each year. This dust has, depending on its chemistry, direct and indirect effects on global climate which include reflection and absorption of solar radiation as well as transport and deposition of nutrients and metals fertilizing both ocean and land. To determine the temporal and spatial variability of Saharan dust transport and deposition and their marine environmental effects across the equatorial North Atlantic Ocean, we have set up a monitoring experiment using deep-ocean sediment traps as well as land-based dust collectors. The sediment traps were deployed at five ocean sites along a transatlantic transect between north-west Africa and the Caribbean along 12° N, in a downwind extension of the land-based dust collectors placed at 19° N on the Mauritanian coast in Iouik. In this paper, we lay out the setup of the monitoring experiment and present the particle fluxes from sediment trap sampling over 24 continuous and synchronized intervals from October 2012 through to November 2013. We establish the temporal distribution of the particle fluxes deposited in the Atlantic and compare chemical compositions with the land-based dust collectors propagating to the downwind sediment trap sites, and with satellite observations of Saharan dust outbreaks.

First-year results show that the total mass fluxes in the ocean are highest at the sampling sites in the east and west, closest to the African continent and the Caribbean, respectively. Element ratios reveal that the lithogenic particles deposited nearest to Africa are most similar in composition to the Saharan dust collected in Iouik. Downwind increasing Al, Fe and K contents suggest a downwind change in the mineralogical composition of Saharan dust and indicate an increasing contribution of clay minerals towards the west. In the westernmost Atlantic Ocean, admixture of re-suspended clay-sized sediments advected towards the deep sediment trap cannot be excluded. Seasonality is most prominent near both continents but generally weak, with mass fluxes dominated by calcium carbonate and clear seasonal maxima of biogenic silica towards the west. The monitoring experiment is now extended, with autonomous dust sampling buoys for better quantification of Saharan dust transport and deposition from source to sink and their impact on fertilization and carbon export to the deep ocean.

Ozone and haze pollution weakens net primary productivity in China

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Source: Atmos. Chem. Phys., 17, 6073-6089, 2017
<https://doi.org/10.5194/acp-17-6073-2017>

Atmospheric pollutants have both beneficial and detrimental effects on carbon uptake by land ecosystems. Surface ozone (O₃) damages leaf photosynthesis by oxidizing plant cells, while aerosols promote carbon uptake by increasing diffuse radiation and exert additional influences through concomitant perturbations to meteorology and hydrology. China is currently the world's largest emitter of both carbon dioxide and short-lived air pollutants. The land ecosystems of China are estimated to provide a carbon sink, but it remains unclear whether air pollution acts to inhibit or promote carbon uptake. Here, we employ Earth system modeling and multiple measurement datasets to assess the separate and combined effects of anthropogenic O₃ and aerosol pollution on net primary productivity (NPP) in China. In the present day, O₃ reduces annual NPP by 0.6 Pg C (14 %) with a range from 0.4 Pg C (low O₃ sensitivity) to 0.8 Pg C (high O₃ sensitivity). In contrast, aerosol direct effects increase NPP by 0.2 Pg C (5 %) through the combination of diffuse radiation fertilization, reduced canopy temperatures, and reduced evaporation leading to higher soil moisture. Consequently, the net effects of O₃ and aerosols decrease NPP by 0.4 Pg C (9 %) with a range from 0.2 Pg C (low O₃ sensitivity) to 0.6 Pg C (high O₃ sensitivity). However, precipitation inhibition from combined aerosol direct and indirect effects reduces annual NPP by 0.2 Pg C (4 %), leading to a net air pollution suppression of 0.8 Pg C (16 %) with a range from 0.6 Pg C (low O₃ sensitivity) to 1.0 Pg C (high O₃ sensitivity). Our results reveal strong dampening effects of air pollution on the land carbon uptake in China today. Following the current legislation emission scenario, this suppression will be further increased by the year 2030, mainly due to a continuing increase in surface O₃. However, the maximum technically feasible reduction scenario could drastically relieve the current level of NPP damage by 70 % in 2030, offering protection of this critical ecosystem service and the mitigation of long-term global warming.

Trace gas composition in the Asian summer monsoon anticyclone: a case study based on aircraft observations and model simulations

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Source: Atmos. Chem. Phys., 17, 6091-6111, 2017
<https://doi.org/10.5194/acp-17-6091-2017>

We present in situ measurements of the trace gas composition of the upper tropospheric (UT) Asian summer monsoon anticyclone (ASMA) performed with the High Altitude and Long Range Research Aircraft (HALO) in the frame of the Earth System Model Validation (ESMVal) campaign. Air masses with enhanced O₃ mixing ratios were encountered after entering the ASMA at its southern edge at about 150 hPa on 18 September 2012. This is in contrast to the presumption that the anticyclone's interior is dominated by recently uplifted air with low O₃ in the monsoon season. We also observed enhanced CO and HCl in the ASMA, which are tracers for boundary layer pollution and tropopause layer (TL) air or stratospheric in-mixing respectively. In addition, reactive nitrogen was enhanced in the ASMA. Along the HALO flight track across the ASMA boundary, strong gradients of these tracers separate anticyclonic from outside air.

Lagrangian trajectory calculations using HYSPLIT show that HALO sampled a filament of UT air three times, which included air masses uplifted from the lower or mid-troposphere north of the Bay of Bengal.

The trace gas gradients between UT and uplifted air masses were preserved during transport within a belt of streamlines fringing the central part of the anticyclone (fringe), but are smaller than the gradients across the ASMA boundary. Our data represent the first in situ observations across the southern part and downstream of the eastern ASMA flank. Back-trajectories starting at the flight track furthermore indicate that HALO transected the ASMA where it was just splitting into a Tibetan and an Iranian part. The O₃-rich filament is diverted from the fringe towards the interior of the original anticyclone, and is at least partially bound to become part of the new Iranian eddy.

A simulation with the ECHAM/MESSy Atmospheric Chemistry (EMAC) model is found to reproduce the observations reasonably well. It shows that O₃-rich air is entrained by the outer streamlines of the anticyclone at its eastern flank. Back-trajectories and increased HCl mixing ratios indicate that the entrained air originates in the stratospherically influenced TL. Photochemical ageing of air masses in the ASMA additionally increases O₃ in originally O₃-poor, but CO-rich air. Simulated monthly mean trace gas distributions show decreased O₃ in the ASMA centre only at the 100 hPa level in July and August, but at lower altitudes and in September the ASMA is dominated by increased O₃. The combination of entrainment from the tropopause region, photochemistry and dynamical instabilities can explain the in situ observations, and might have a larger impact on the highly variable trace gas composition of the anticyclone than previously thought.

New particle formation in the Svalbard region 2006–2015

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Source: Atmos. Chem. Phys., 17, 6153–6175, 2017
<https://doi.org/10.5194/acp-17-6153-2017>

Events of new particle formation (NPF) were analyzed in a 10-year data set of hourly particle size distributions recorded on Mt. Zeppelin, Spitsbergen, Svalbard. Three different types of NPF events were identified through objective search algorithms. The first and simplest algorithm utilizes short-term increases in particle concentrations below 25 nm (PCT (percentiles) events). The second one builds on the growth of the sub-50 nm diameter median (DGR (diameter growth) events) and is most closely related to the classical banana type of event. The third and most complex, multiple-size approach to identifying NPF events builds on a hypothesis suggesting the concurrent production of polymer gel particles at several sizes below ca. 60 nm (MEV (multi-size growth) events).

As a first and general conclusion, we can state that NPF events are a summer phenomenon and not related to Arctic haze, which is a late winter to early spring feature. The occurrence of NPF events appears to be somewhat sensitive to the available data on precipitation. The seasonal distribution of solar flux suggests some photochemical control that may affect marine biological processes generating particle precursors and/or atmospheric photochemical processes that generate condensable vapors from precursor gases. Notably, the seasonal distribution of the biogenic methanesulfonate (MSA) follows that of the solar flux although it peaks before the maxima in NPF occurrence.

A host of ancillary data and findings point to varying and rather complex marine biological source processes. The potential source regions for all types of new particle formation appear to be restricted to

the marginal-ice and open-water areas between northeastern Greenland and eastern Svalbard. Depending on conditions, yet to be clarified new particle formation may become visible as short bursts of particles around 20 nm (PCT events), longer events involving condensation growth (DGR events), or extended events with elevated concentrations of particles at several sizes below 100 nm (MEV events). The seasonal distribution of NPF events peaks later than that of MSA and DGR, and in particular than that of MEV events, which reach into late summer and early fall with open, warm, and biologically active waters around Svalbard. Consequently, a simple model to describe the seasonal distribution of the total number of NPF events can be based on solar flux and sea surface temperature, representing environmental conditions for marine biological activity and condensation sink, controlling the balance between new particle nucleation and their condensational growth. Based on the sparse knowledge about the seasonal cycle of gel-forming marine microorganisms and their controlling factors, we hypothesize that the seasonal distribution of DGR and, more so, MEV events reflect the seasonal cycle of the gel-forming phytoplankton.

Spatial, temporal and source contribution assessments of black carbon over the northern interior of South Africa

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Source: Atmos. Chem. Phys., 17, 6177-6196, 2017
<https://doi.org/10.5194/acp-17-6177-2017>

After carbon dioxide (CO₂), aerosol black carbon (BC) is considered to be the second most important contributor to global warming. This paper presents equivalent black carbon (eBC) (derived from an optical absorption method) data collected from three sites in the interior of South Africa where continuous measurements were conducted, i.e. Elandsfontein, Welgegund and Marikana, as well elemental carbon (EC) (determined by evolved carbon method) data at five sites where samples were collected once a month on a filter and analysed offline, i.e. Louis Trichardt, Skukuza, Vaal Triangle, Amersfoort and Botsalano.

Analyses of eBC and EC spatial mass concentration patterns across the eight sites indicate that the mass concentrations in the South African interior are in general higher than what has been reported for the developed world and that different sources are likely to influence different sites. The mean eBC or EC mass concentrations for the background sites (Welgegund, Louis Trichardt, Skukuza, Botsalano) and sites influenced by industrial activities and/or nearby settlements (Elandsfontein, Marikana, Vaal Triangle and Amersfoort) ranged between 0.7 and 1.1, and 1.3 and 1.4 µg m⁻³, respectively. Similar seasonal patterns were observed at all three sites where continuous measurement data were collected (Elandsfontein, Marikana and Welgegund), with the highest eBC mass concentrations measured from June to October, indicating contributions from household combustion in the cold winter months (June–August), as well as savannah and grassland fires during the dry season (May to mid-October). Diurnal patterns of eBC at Elandsfontein, Marikana and Welgegund indicated maximum concentrations in the early mornings and late evenings, and minima during daytime. From the patterns it could be deduced

that for Marikana and Welgegund, household combustion, as well as savannah and grassland fires, were the most significant sources, respectively.

Possible contributing sources were explored in greater detail for Elandsfontein, with five main sources being identified as coal-fired power stations, pyrometallurgical smelters, traffic, household combustion, as well as savannah and grassland fires. Industries on the Mpumalanga Highveld are often blamed for all forms of pollution, due to the NO₂ hotspot over this area that is attributed to NO_x emissions from industries and vehicle emissions from the Johannesburg–Pretoria megacity. However, a comparison of source strengths indicated that household combustion as well as savannah and grassland fires were the most significant sources of eBC, particularly during winter and spring months, while coal-fired power stations, pyrometallurgical smelters and traffic contribute to eBC mass concentration levels year round.

Temporal and spatial variability of ammonia in urban and agricultural regions of northern Colorado, United States

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Source: Atmos. Chem. Phys., 17, 6197-6213, 2017
<https://doi.org/10.5194/acp-17-6197-2017>

Concentrated agricultural activities and animal feeding operations in the northeastern plains of Colorado represent an important source of atmospheric ammonia (NH₃). The NH₃ from these sources contributes to regional fine particle formation and to nitrogen deposition to sensitive ecosystems in Rocky Mountain National Park (RMNP), located ~ 80 km to the west. In order to better understand temporal and spatial differences in NH₃ concentrations in this source region, weekly concentrations of NH₃ were measured at 14 locations during the summers of 2010 to 2015 using Radiello passive NH₃ samplers. Weekly (biweekly in 2015) average NH₃ concentrations ranged from 2.66 to 42.7 µg m⁻³, with the highest concentrations near large concentrated animal feeding operations (CAFOs). The annual summertime mean NH₃ concentrations were stable in this region from 2010 to 2015, providing a baseline against which concentration changes associated with future changes in regional NH₃ emissions can be assessed. Vertical profiles of NH₃ were also measured on the 300 m Boulder Atmospheric Observatory (BAO) tower throughout 2012. The highest NH₃ concentration along the vertical profile was always observed at the 10 m height (annual average concentration of 4.63 µg m⁻³), decreasing toward the surface (4.35 µg m⁻³) and toward higher altitudes (1.93 µg m⁻³). The NH₃ spatial distributions measured using the passive samplers are compared with NH₃ columns retrieved by the Infrared Atmospheric Sounding Interferometer (IASI) satellite and concentrations simulated by the Comprehensive Air Quality Model with Extensions (CAMx). The satellite comparison adds to a growing body of evidence that IASI column retrievals of NH₃ provide very useful insight into regional variability in atmospheric NH₃, in this case even in a region with strong local sources and sharp spatial gradients. The CAMx comparison indicates that the model does a reasonable job simulating NH₃ concentrations near sources but tends to underpredict concentrations at locations farther downwind. Excess NH₃ deposition by the model is hypothesized as a possible explanation for this trend.

Technical note: Boundary layer height determination from lidar for improving air pollution episode modeling: development of new algorithm and evaluation

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Source: Atmos. Chem. Phys., 17, 6215-6225, 2017
<https://doi.org/10.5194/acp-17-6215-2017>

Predicting air pollution events in the low atmosphere over megacities requires a thorough understanding of the tropospheric dynamics and chemical processes, involving, notably, continuous and accurate determination of the boundary layer height (BLH). Through intensive observations experimented over Beijing (China) and an exhaustive evaluation of existing algorithms applied to the BLH determination, persistent critical limitations are noticed, in particular during polluted episodes. Basically, under weak thermal convection with high aerosol loading, none of the retrieval algorithms is able to fully capture the diurnal cycle of the BLH due to insufficient vertical mixing of pollutants in the boundary layer associated with the impact of gravity waves on the tropospheric structure. Consequently, a new approach based on gravity wave theory (the cubic root gradient method: CRGM) is developed to overcome such weakness and accurately reproduce the fluctuations of the BLH under various atmospheric pollution conditions. Comprehensive evaluation of CRGM highlights its high performance in determining BLH from lidar. In comparison with the existing retrieval algorithms, CRGM potentially reduces related computational uncertainties and errors from BLH determination (strong increase of correlation coefficient from 0.44 to 0.91 and significant decreases of the root mean square error from 643 to 142 m). Such a newly developed technique is undoubtedly expected to contribute to improving the accuracy of air quality modeling and forecasting systems.

Impact of Saharan dust on North Atlantic marine stratocumulus clouds: importance of the semidirect effect

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Source: Atmos. Chem. Phys., 17, 6305-6322, 2017
<https://doi.org/10.5194/acp-17-6305-2017>

One component of aerosol–cloud interactions (ACI) involves dust and marine stratocumulus clouds (MSc). Few observational studies have focused on dust–MSc interactions, and thus this effect remains poorly quantified. We use observations from multiple sensors in the NASA A-Train satellite constellation from 2004 to 2012 to obtain estimates of the aerosol–cloud radiative effect, including its uncertainty, of dust aerosol influencing Atlantic MSc off the coast of northern Africa between 45° W and 15° E and between 0 and 35° N. To calculate the aerosol–cloud radiative effect, we use two methods following Quaas et al. (2008) (Method 1) and Chen et al. (2014) (Method 2). These two methods yield similar results of -1.5 ± 1.4 and -1.5 ± 1.6 W m⁻², respectively, for the annual mean aerosol–cloud radiative effect. Thus, Saharan dust modifies MSc in a way that acts to cool the planet. There is a strong seasonal

variation, with the aerosol–cloud radiative effect switching from significantly negative during the boreal summer to weakly positive during boreal winter. Method 1 (Method 2) yields -3.8 ± 2.5 (-4.3 ± 4.1) during summer and 1 ± 2.9 (0.6 ± 1) W m^{-2} during winter. In Method 1, the aerosol–cloud radiative effect can be decomposed into two terms, one representing the first aerosol indirect effect and the second representing the combination of the second aerosol indirect effect and the semidirect effect (i.e., changes in liquid water path and cloud fraction in response to changes in absorbing aerosols and local heating). The first aerosol indirect effect is relatively small, varying from -0.7 ± 0.6 in summer to 0.1 ± 0.5 W m^{-2} in winter. The second term, however, dominates the overall radiative effect, varying from -3.2 ± 2.5 in summer to 0.9 ± 2.9 W m^{-2} during winter. Studies show that the semidirect effect can result in a negative (i.e., absorbing aerosol lies above low clouds like MSc) or positive (i.e., absorbing aerosol lies within low clouds) aerosol–cloud radiative effect. The semipermanent MSc are low and confined within the boundary layer. CALIPSO shows that 61.8 ± 12.6 % of Saharan dust resides above North Atlantic MSc during summer for our study area. This is consistent with a relatively weak first aerosol indirect effect and also suggests the second aerosol indirect effect plus semidirect effect (the second term in Method 1) is dominated by the semidirect effect. In contrast, the percentage of Saharan dust above North Atlantic MSc in winter is 11.9 ± 10.9 %, which is much lower than in summer. CALIPSO also shows that 88.3 ± 8.5 % of dust resides below 2.2 km the winter average of MSc top height. During summer, however, there are two peaks, with 35.6 ± 13 % below 1.9 km (summer average of MSc top height) and 44.4 ± 9.2 % between 2 and 4 km. Because the aerosol–cloud radiative effect is positive during winter, and is also dominated by the second term, this again supports the importance of the semidirect effect. We conclude that Saharan dust–MSc interactions off the coast of northern Africa are likely dominated by the semidirect effect.

Heterogeneous uptake of ammonia and dimethylamine into sulfuric and oxalic acid particles

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Source: Atmos. Chem. Phys., 17, 6323–6339, 2017
<https://doi.org/10.5194/acp-17-6323-2017>

Heterogeneous uptake is one of the major mechanisms governing the amounts of short-chain alkylamines and ammonia (NH_3) in atmospheric particles. Molar ratios of aminium to ammonium ions detected in ambient aerosols often exceed typical gas phase ratios. The present study investigated the simultaneous uptake of dimethylamine (DMA) and NH_3 into sulfuric and oxalic acid particles at gaseous DMA/ NH_3 molar ratios of 0.1 and 0.5 at 10, 50 and 70 % relative humidity (RH). Single-gas uptake and co-uptake were conducted under identical conditions and compared. Results show that the particulate dimethyl-aminium/ammonium molar ratios (DMAH/NH_4) changed substantially during the uptake process, which was severely influenced by the extent of neutralisation and the particle phase state. In general, DMA uptake and NH_3 uptake into concentrated H_2SO_4 droplets were initially similarly efficient, yielding DMAH/NH_4 ratios that were similar to DMA/ NH_3 ratios. As the co-uptake continued, the DMAH/NH_4 gradually dropped due to a preferential uptake of NH_3 into partially neutralised acidic droplets. At 50 % RH, once the sulfate droplets were neutralised, the stronger base DMA displaced some of the ammonium absorbed earlier, leading to DMAH/NH_4 ratios up to four times higher than the

corresponding gas phase ratios. However, at 10 % RH, crystallisation of partially neutralised sulfate particles prevented further DMA uptake, while NH₃ uptake continued and displaced DMAH⁺, forming almost pure ammonium sulfate. Displacement of DMAH⁺ by NH₃ has also been observed in neutralised, solid oxalate particles. The results can explain why DMAH⁺/NH₄ ratios in ambient liquid aerosols can be larger than DMA/NH₃, despite an excess of NH₃ in the gas phase. An uptake of DMA to aerosols consisting of crystalline ammonium salts, however, is unlikely, even at comparable DMA and NH₃ gas phase concentrations.

Comparison of emissions inventories of anthropogenic air pollutants and greenhouse gases in China

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Source: Atmos. Chem. Phys., 17, 6393-6421, 2017
<https://doi.org/10.5194/acp-17-6393-2017>

Anthropogenic air pollutant emissions have been increasing rapidly in China, leading to worsening air quality. Modelers use emissions inventories to represent the temporal and spatial distribution of these emissions needed to estimate their impacts on regional and global air quality. However, large uncertainties exist in emissions estimates. Thus, assessing differences in these inventories is essential for the better understanding of air pollution over China. We compare five different emissions inventories estimating emissions of carbon dioxide (CO₂), carbon monoxide (CO), nitrogen oxides (NO_x), sulfur dioxide (SO₂), and particulate matter with an aerodynamic diameter of 10 µm or less (PM₁₀) from China. The emissions inventories analyzed in this paper include the Regional Emission inventory in ASia v2.1 (REAS), the Multi-resolution Emission Inventory for China (MEIC), the Emission Database for Global Atmospheric Research v4.2 (EDGAR), the inventory by Yu Zhao (ZHAO), and the Greenhouse Gas and Air Pollution Interactions and Synergies (GAINS). We focus on the period between 2000 and 2008, during which Chinese economic activities more than doubled. In addition to national totals, we also analyzed emissions from four source sectors (industry, transport, power, and residential) and within seven regions in China (East, North, Northeast, Central, Southwest, Northwest, and South) and found that large disagreements exist among the five inventories at disaggregated levels. These disagreements lead to differences of 67 µg m⁻³, 15 ppbv, and 470 ppbv for monthly mean PM₁₀, O₃, and CO, respectively, in modeled regional concentrations in China. We also find that all the inventory emissions estimates create a volatile organic compound (VOC)-limited environment and MEIC emissions lead to much lower O₃ mixing ratio in East and Central China compared to the simulations using REAS and EDGAR estimates, due to their low VOC emissions. Our results illustrate that a better understanding of Chinese emissions at more disaggregated levels is essential for finding effective mitigation measures for reducing national and regional air pollution in China.

Particulate emissions from large North American wildfires estimated using a new top-down method

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Source: *Atmos. Chem. Phys.*, 17, 6423-6438, 2017
<https://doi.org/10.5194/acp-17-6423-2017>

Particulate matter emissions from wildfires affect climate, weather and air quality. However, existing global and regional aerosol emission estimates differ by a factor of up to 4 between different methods. Using a novel approach, we estimate daily total particulate matter (TPM) emissions from large wildfires in North American boreal and temperate regions. Moderate Resolution Imaging Spectroradiometer (MODIS) fire location and aerosol optical thickness (AOT) data sets are coupled with HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) atmospheric dispersion simulations, attributing identified smoke plumes to sources. Unlike previous approaches, the method (i) combines information from both satellite and AERONET (AErosol RObotic NETwork) observations to take into account aerosol water uptake and plume specific mass extinction efficiency when converting smoke AOT to TPM, and (ii) does not depend on instantaneous emission rates observed during individual satellite overpasses, which do not sample night-time emissions. The method also allows multiple independent estimates for the same emission period from imagery taken on consecutive days.

Repeated fire-emitted AOT estimates for the same emission period over 2 to 3 days of plume evolution show increases in plume optical thickness by approximately 10 % for boreal events and by 40 % for temperate emissions. Inferred median water volume fractions for aged boreal and temperate smoke observations are 0.15 and 0.47 respectively, indicating that the increased AOT is partly explained by aerosol water uptake. TPM emission estimates for boreal events, which predominantly burn during daytime, agree closely with bottom-up Global Fire Emission Database (GFEDv4) and Global Fire Assimilation System (GFASv1.0) inventories, but are lower by approximately 30 % compared to Quick Fire Emission Dataset (QFEDv2) PM_{2.5}, and are higher by approximately a factor of 2 compared to Fire Energetics and Emissions Research (FEERv1) TPM estimates. The discrepancies are larger for temperate fires, which are characterized by lower median fire radiative power values and more significant night-time combustion. The TPM estimates for this study for the biome are lower than QFED PM_{2.5} by 35 %, and are larger by factors of 2.4, 3.2 and 4 compared with FEER, GFED and GFAS inventories respectively. A large underestimation of TPM emission by bottom-up GFED and GFAS indicates low biases in emission factors or consumed biomass estimates for temperate fires.

Surface ozone in the Southern Hemisphere: 20 years of data from a site with a unique setting in El Tololo, Chile

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Source: *Atmos. Chem. Phys.*, 17, 6477-6492, 2017
<https://doi.org/10.5194/acp-17-6477-2017>

The knowledge of surface ozone mole fractions and their global distribution is of utmost importance due to the impact of ozone on human health and ecosystems and the central role of ozone in controlling the oxidation capacity of the troposphere. The availability of long-term ozone records is far better in the Northern than in the Southern Hemisphere, and recent analyses of the seven accessible records in the Southern Hemisphere have shown inconclusive trends. Since late 1995, surface ozone is measured in situ at "El Tololo", a high-altitude (2200 m a.s.l.) and pristine station in Chile (30° S, 71° W). The dataset has been recently fully quality controlled and reprocessed. This study presents the observed ozone trends and annual cycles and identifies key processes driving these patterns. From 1995 to 2010, an overall positive trend of ~ 0.7 ppb decade⁻¹ is found. Strongest trends per season are observed in March and April. Highest mole fractions are observed in late spring (October) and show a strong correlation with ozone transported from the stratosphere down into the troposphere, as simulated with a model. Over the 20 years of observations, the springtime ozone maximum has shifted to earlier times in the year, which, again, is strongly correlated with a temporal shift in the occurrence of the maximum of simulated stratospheric ozone transport at the site. We conclude that background ozone at El Tololo is mainly driven by stratospheric intrusions rather than photochemical production from anthropogenic and biogenic precursors. The major footprint of the sampled air masses is located over the Pacific Ocean. Therefore, due to the negligible influence of local processes, the ozone record also allows studying the influence of El Niño and La Niña episodes on background ozone levels in South America. In agreement with previous studies, we find that, during La Niña conditions, ozone mole fractions reach higher levels than during El Niño conditions.

Efficient bulk mass accommodation and dissociation of N₂O₅ in neutral aqueous aerosol

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Source: *Atmos. Chem. Phys.*, 17, 6493-6502, 2017
<https://doi.org/10.5194/acp-17-6493-2017>

An isotope exchange experiment with the short-lived radioactive tracer ¹³N is used to trace N₂O₅ uptake into nitrate-containing aqueous aerosol particles. While uptake of ¹³N-labelled N₂O₅ to deliquesced Na₂SO₄ aerosol is consistent with previous studies, efficient exchange of labelled nitrate with the non-labelled nitrate pool was observed in the presence of aerosol-phase nitrate. The experiments provide direct evidence for efficient bulk mass accommodation of N₂O₅ into aqueous solution with $\alpha_b > 0.4$ at room temperature, as well as for the fast dissociation into nitronium and nitrate. While for experimental reasons this study is limited to non-acidic nitrate-containing aerosol, it is likely that bulk mass accommodation and dissociation are not limiting N₂O₅ uptake under wider ranges of conditions.

Near-road sampling of PM_{2.5}, BC, and fine-particle chemical components in Kathmandu Valley, Nepal

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Source: Atmos. Chem. Phys., 17, 6503-6516, 2017
<https://doi.org/10.5194/acp-17-6503-2017>

Semicontinuous PM_{2.5} and black carbon (BC) concentrations, and 24 h integrated PM_{2.5} filter samples were collected near roadways in the Kathmandu Valley, Nepal. Instruments were carried by a group of volunteer traffic police officers in the vicinity of six major roadway intersections in the Kathmandu Valley across two sampling periods in 2014. Daily PM_{2.5} filter samples were analyzed for water-soluble inorganic ions, elemental carbon (EC) and organic carbon (OC), and 24 elements. Mean PM_{2.5} and BC concentrations were 124.76 $\mu\text{g m}^{-3}$ and 16.74 $\mu\text{gC m}^{-3}$ during the drier spring sampling period, and 45.92 $\mu\text{g m}^{-3}$ and 13.46 $\mu\text{gC m}^{-3}$ during monsoonal sampling. Despite the lower monsoonal PM_{2.5} concentrations, BC and several elements were not significantly lower during the monsoon, which indicates an important contribution of vehicle-related emissions throughout both seasons in this region. During the monsoon, there was an enhanced contribution of chemical species (elements and water-soluble inorganic ions), except secondary inorganic ions, and BC to PM_{2.5} (crustal elements: 19 %; heavy metals: 5 %; and BC: 39 %) compared to those in spring (crustal elements: 9 %; heavy metals: 1 %; and BC: 18 %). Silica, calcium, aluminum, and iron were the most abundant elements during both spring and the monsoon, with total concentrations of 12.13 and 8.85 $\mu\text{g m}^{-3}$, respectively. PM_{2.5} and BC showed less spatial variation compared to that for individual chemical species.

Summer ozone in the northern Front Range metropolitan area: weekend-weekday effects, temperature dependences, and the impact of drought

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Source: Atmos. Chem. Phys., 17, 6517-6529, 2017
<https://doi.org/10.5194/acp-17-6517-2017>

Contrary to most regions in the US, ozone in the northern Front Range metropolitan area (NFRMA) of Colorado was either stagnant or increasing between 2000 and 2015, despite substantial reductions in NO_x emissions. We used available long-term ozone and NO_x data in the NFRMA to investigate these trends. Ozone increased from weekdays to weekends for a number of sites in the NFRMA with weekend reductions in NO₂ at two sites in downtown Denver, indicating that the region was in a NO_x-saturated ozone production regime. The stagnation and increases in ozone in the NFRMA are likely due to a combination of decreasing NO_x emissions in a NO_x-saturated environment and increased anthropogenic volatile organic compound (VOC) emissions in the NFRMA. Further investigation of the weekend-weekday effect showed that the region outside of the most heavily trafficked Denver area was transitioning to peak ozone production towards NO_x-limited chemistry. This transition implies that continued NO_x decreases will result in ozone being less sensitive to changes in either anthropogenic or

biogenic VOC reactivity in the NFRMA. In contrast to anthropogenic VOCs, biogenic VOCs are unlikely to have increased in the NFRMA between 2000 and 2015, but are temperature dependent and likely vary by drought year. Ozone in the NFRMA has a temperature dependence, albeit smaller than many other US locations, consistent with biogenic VOC contributions to ozone production in the region. We show that while ozone increased with temperature in the NFRMA, which is consistent with a NO_x-saturated regime coupled to temperature-dependent VOCs, this relationship is suppressed in drought years. We attribute this drought year suppression to decreased biogenic isoprene emissions due to long-term drought stress. Thus, while anthropogenic NO_x and VOCs likely dominate ozone production regimes in the NFRMA, biogenic VOCs may also impact regional ozone and its temperature dependence.

Glacier evolution in high-mountain Asia under stratospheric sulfate aerosol injection geoengineering

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Source: Atmos. Chem. Phys., 17, 6547-6564, 2017
<https://doi.org/10.5194/acp-17-6547-2017>

Geoengineering by stratospheric sulfate aerosol injection may help preserve mountain glaciers by reducing summer temperatures. We examine this hypothesis for the glaciers in high-mountain Asia using a glacier mass balance model driven by climate simulations from the Geoengineering Model Intercomparison Project (GeoMIP). The G3 and G4 schemes specify use of stratospheric sulfate aerosols to reduce the radiative forcing under the Representative Concentration Pathway (RCP) 4.5 scenario for the 50 years between 2020 and 2069, and for a further 20 years after termination of geoengineering. We estimate and compare glacier volume loss for every glacier in the region using a glacier model based on surface mass balance parameterization under climate projections from three Earth system models under G3, five models under G4, and six models under RCP4.5 and RCP8.5. The ensemble projections suggest that glacier shrinkage over the period 2010–2069 is equivalent to sea-level rise of 9.0 ± 1.6 mm (G3), 9.8 ± 4.3 mm (G4), 15.5 ± 2.3 mm (RCP4.5), and 18.5 ± 1.7 mm (RCP8.5). Although G3 keeps the average temperature from increasing in the geoengineering period, G3 only slows glacier shrinkage by about 50 % relative to losses from RCP8.5. Approximately 72 % of glaciated area remains at 2069 under G3, as compared with about 30 % for RCP8.5. The widely reported reduction in mean precipitation expected for solar geoengineering is unlikely to be as important as the temperature-driven shift from solid to liquid precipitation for forcing Himalayan glacier change. The termination of geoengineering at 2069 under G3 leads to temperature rise of about 1.3 °C over the period 2070–2089 relative to the period 2050–2069 and corresponding increase in annual mean glacier volume loss rate from 0.17 to 1.1 % yr⁻¹, which is higher than the 0.66 % yr⁻¹ under RCP8.5 during 2070–2089.

Regional background O₃ and NO_x in the Houston–Galveston–Brazoria (TX) region: a decadal-scale perspective

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Source: Atmos. Chem. Phys., 17, 6565-6581, 2017
<https://doi.org/10.5194/acp-17-6565-2017>

Ozone (O₃) in the lower troposphere is harmful to people and plants, particularly during summer, when photochemistry is most active and higher temperatures favor local chemistry. Local precursor emissions, such as those of volatile organic compounds (VOCs) and nitrogen oxides (NO_x), together with their chemistry contribute to the O₃ and NO_x mixing ratios in the Houston–Galveston–Brazoria (HGB) region. In addition to local emissions, chemistry and transport, larger-scale factors also contribute to local O₃ and NO_x. These additional contributions (often referred to as regional background) are not well quantified within the HGB region, impeding more efficient controls on precursor emissions to achieve compliance with the National Ambient Air Quality Standards for O₃. In this study, we estimate ground-level regional background O₃ and NO_x in the HGB region and quantify their decadal-scale trends.

We use four different approaches based on principal component analysis (PCA) to quantify background O₃ and NO_x. Three of these approaches consist of independent PCA on both O₃ and NO_x for both 1 and 8 h levels to compare our results with previous studies and to highlight the effect of both temporal and spatial scales. In the fourth approach, we co-varied O₃, NO_x and meteorology.

Our results show that the estimation of regional background O₃ has less inherent uncertainty when it was constrained by NO_x and meteorology, yielding a statistically significant temporal trend of -0.68 ± 0.27 ppb yr⁻¹. Likewise, the estimation of regional background NO_x trend constrained by O₃ and meteorology was -0.04 ± 0.02 ppb yr⁻¹ (upper bound) and -0.03 ± 0.01 ppb yr⁻¹ (lower bound). Our best estimates of the 17-year average of season-scale background O₃ and NO_x were 46.72 ± 2.08 ppb and 6.80 ± 0.13 ppb (upper bound) or 4.45 ± 0.08 ppb (lower bound), respectively. Average background O₃ is consistent with previous studies and between the approaches used in this study, although the approaches based on 8 h averages likely overestimate background O₃ compared to the hourly median approach by 7–9 ppb. Similarly, the upper bound of average background NO_x is consistent between approaches in this study (A–C) but overestimated compared to the hourly approach by 1 ppb, on average. We likely overestimate the upper-bound background NO_x due to instrument overdetection of NO_x and the 8 h averaging of NO_x and meteorology coinciding with MDA8 O₃.

Influence of urban pollution on the production of organic particulate matter from isoprene epoxydiols in central Amazonia

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Source: Atmos. Chem. Phys., 17, 6611-6629, 2017
<https://doi.org/10.5194/acp-17-6611-2017>

The atmospheric chemistry of isoprene contributes to the production of a substantial mass fraction of the particulate matter (PM) over tropical forests. Isoprene epoxydiols (IEPOX) produced in the gas phase by the oxidation of isoprene under HO₂-dominant conditions are subsequently taken up by particles,

thereby leading to production of secondary organic PM. The present study investigates possible perturbations to this pathway by urban pollution. The measurement site in central Amazonia was located 4 to 6 h downwind of Manaus, Brazil. Measurements took place from February through March 2014 of the wet season, as part of the GoAmazon2014/5 experiment. Mass spectra of organic PM collected with an Aerodyne Aerosol Mass Spectrometer were analyzed by positive-matrix factorization. One resolved statistical factor (IEPOX-SOA factor) was associated with PM production by the IEPOX pathway. The IEPOX-SOA factor loadings correlated with independently measured mass concentrations of tracers of IEPOX-derived PM, namely C5-alkene triols and 2-methyltetrols ($R=0.96$ and 0.78 , respectively). The factor loading, as well as the ratio of the loading to organic PM mass concentration, decreased under polluted compared to background conditions. For an increase in NO_y concentration from 0.5 to 2 ppb, the factor loading and decreased by two to three fold. Overall, sulfate concentration explained 37 % of the variability in the factor loading. After segregation of factor loading into subsets based on NO_y concentration, the sulfate concentration explained up to 75 % of the variability. Considering both factors, the data sets show that the suppressing effects of increased NO concentrations dominated over the enhancing effects of higher sulfate concentrations. The pollution from Manaus elevated NO_y concentrations more significantly than sulfate concentrations relative to background conditions. In this light, increased emissions of nitrogen oxides, as anticipated for some scenarios of Amazonian economic development, could significantly alter pathways of PM production that presently prevail over the tropical forest, implying changes to air quality and regional climate.

The relationship between lower-stratospheric ozone at southern high latitudes and sea surface temperature in the East Asian marginal seas in austral spring

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Source: Atmos. Chem. Phys., 17, 6705-6722, 2017
<https://doi.org/10.5194/acp-17-6705-2017>

Using satellite observations, reanalysis data, and model simulations, this study investigates the effect of sea surface temperature (SST) on interannual variations of lower-stratospheric ozone at southern high latitudes in austral spring. It is found that the SST variations across the East Asian marginal seas (5°S – 35°N , 100 – 140°E) rather than the tropical eastern Pacific Ocean, where ENSO occurs, have the most significant correlation with the southern high-latitude lower-stratospheric ozone changes in austral spring. Further analysis reveals that planetary waves originating over the marginal seas in austral spring can propagate towards southern middle to high latitudes via teleconnection pathway. The anomalous propagation and dissipation of ultra-long Rossby waves in the stratosphere strengthen/cool (weaken/warm) the southern polar vortex, which produces more (less) active chlorine and enhances (suppresses) ozone depletion in the southern high-latitude stratosphere on one the hand and impedes (favors) the transport of ozone from the southern middle-latitude stratosphere to high latitudes on the other. The model simulations also reveal that approximately 17 % of the decreasing trend in the southern high-latitude lower-stratospheric ozone observed over the past 5 decades may be associated with the increasing trend in SST over the East Asian marginal seas.

Secondary ozone peaks in the troposphere over the Himalayas

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Source: Atmos. Chem. Phys., 17, 6743-6757, 2017
<https://doi.org/10.5194/acp-17-6743-2017>

Layers with strongly enhanced ozone concentrations in the middle–upper troposphere, referred to as secondary ozone peaks (SOPs), have been observed in different regions of the world. Here we use the global ECHAM5/MESSy atmospheric chemistry model (EMAC) to (i) investigate the processes causing SOPs, (ii) explore both their frequency of occurrence and seasonality, and (iii) assess their effects on the tropospheric ozone budget over the Himalayas. The vertical profiles of potential vorticity (PV) and a stratospheric ozone tracer (O3s) in EMAC simulations, in conjunction with the structure of SOPs, suggest that SOPs over the Himalayas are formed by stratosphere-to-troposphere transport (STT) of ozone. The spatial distribution of O3s further shows that such effects are in general most pronounced in the northern part of India. Model simulated ozone distributions and backward air trajectories show that ozone rich air masses, associated with STT, originate as far as northern Africa and the North Atlantic Ocean, the Middle East, as well as in nearby regions in Afghanistan and Pakistan, and are rapidly (within 2–3 days) transported to the Himalayas. Analysis of a 15-year (2000–2014) EMAC simulation shows that the frequency of SOPs is highest during the pre-monsoon season (e.g. 11 % of the time in May), while no intense SOP events are found during the July–October period. The SOPs are estimated to enhance the tropospheric column ozone (TCO) over the central Himalayas by up to 21 %.

Measurement of PM and its chemical composition in real-world emissions from non-road and on-road diesel vehicles

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Source: Atmos. Chem. Phys., 17, 6779-6795, 2017
<https://doi.org/10.5194/acp-17-6779-2017>

With the rapid growth in the number of both non-road and on-road diesel vehicles, the adverse effects of particulate matter (PM) and its constituents on air quality and human health have attracted increasing attentions. However, studies on the characteristics of PM and its composition emitted from diesel vehicles are still scarce, especially under real-world driving conditions. In this study, six excavators and five trucks that provided a wide range of emission standards and operation modes were tested, and PM emissions and their constituents – including organic carbon (OC), elemental carbon (EC), water-soluble ions (WSIs), elements, and organic species like polycyclic aromatic hydrocarbons (PAHs), n-alkanes, and hopanes – as well as steranes were analyzed and characterized. The average emission factors for PM (EFPM) from excavator and truck emissions were 829 ± 806 and 498 ± 234 mg kg⁻¹ fuel, respectively. EFPM and PM constituents were significantly affected by fuel quality, operational mode, and emission standards. A significant correlation ($R^2 = 0.79$, $p < 0.01$) was found between EFPM for excavators and the sulfur contents in fuel. The highest average EFPM for working excavators was 904 ± 979 mg kg⁻¹ fuel as a higher engine load required in this mode. From pre-stage 1 to stage 2, the

average EFPM for excavators decreased by 58 %. For trucks, the average non-highway EFPM at $548 \pm 311 \text{ mg kg}^{-1}$ fuel was higher than the highway EFPM at $497 \pm 231 \text{ mg kg}^{-1}$ fuel. Moreover, the reduction rates were 63.5 and 65.6 % when switched from China II and III to China IV standards, respectively. Generally, the PM composition emitted from excavators was dominated by OC ($39.2 \pm 21.0 \%$) and EC ($33.3 \pm 25.9 \%$); PM from trucks was dominated by EC ($26.9 \pm 20.8 \%$), OC ($9.89 \pm 12 \%$), and WSIs ($4.67 \pm 5.74 \%$). The average OC/EC ratios for idling and working excavators were 3 to 4 times higher than those for moving excavators. Although the EFPM for excavators and trucks was reduced with the constraint of regulations, the element fractions for excavators increased from 0.49 % in pre-stage 1 to 3.03 % in stage 2, and the fraction of WSIs for the China IV truck was 5 times higher than the average value of all other-level trucks. Furthermore, as compared with other diesel vehicles, wide ranges were found for excavators of the ratios of benzo[a]anthracene / (benzo[a]anthracene + chrysene) (0.26–0.86), indeno[1,2,3-cd]pyrene / (indeno[1,2,3-cd]pyrene + benzo[ghi]perylene) (0.20–1.0), and fluoranthene / (fluoranthene + pyrene) (0.24–0.87), which might be a result of the complex characteristics of the excavator operation modes. A comparison of our results with those in the literature revealed that on-board measurement data more accurately reflect actual conditions. Although the fractions of the 16 priority PAHs in PM from the excavator and truck emissions were similar, the equivalent concentrations of total benzo[a]pyrene of excavators were 31 times than that for trucks, implying that more attention should be paid to non-road vehicle emissions.

Influence of biomass burning from South Asia at a high-altitude mountain receptor site in China

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Source: Atmos. Chem. Phys., 17, 6853-6864, 2017
<https://doi.org/10.5194/acp-17-6853-2017>

Highly time-resolved in situ measurements of airborne particles were conducted at Mt. Yulong (3410 m above sea level) on the southeastern edge of the Tibetan Plateau in China from 22 March to 14 April 2015. The detailed chemical composition was measured by a high-resolution time-of-flight aerosol mass spectrometer together with other online instruments. The average mass concentration of the submicron particles (PM₁) was $5.7 \pm 5.4 \mu\text{g m}^{-3}$ during the field campaign, ranging from 0.1 up to $33.3 \mu\text{g m}^{-3}$. Organic aerosol (OA) was the dominant component in PM₁, with a fraction of 68 %. Three OA factors, i.e., biomass burning organic aerosol (BBOA), biomass-burning-influenced oxygenated organic aerosol (OOA-BB) and oxygenated organic aerosol (OOA), were resolved using positive matrix factorization analysis. The two oxygenated OA factors accounted for 87 % of the total OA mass. Three biomass burning events were identified by examining the enhancement of black carbon concentrations and the f₆₀ (the ratio of the signal at m/z 60 from the mass spectrum to the total signal of OA). Back trajectories of air masses and satellite fire map data were integrated to identify the biomass burning locations and pollutant transport. The western air masses from South Asia with active biomass burning activities transported large amounts of air pollutants, resulting in elevated organic concentrations up to 4-fold higher than those of the background conditions. This study at Mt. Yulong characterizes the

tropospheric background aerosols of the Tibetan Plateau during pre-monsoon season and provides clear evidence that the southeastern edge of the Tibetan Plateau was affected by the transport of anthropogenic aerosols from South Asia.

Radiative and climate effects of stratospheric sulfur geoengineering using seasonally varying injection areas

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Source: Atmos. Chem. Phys., 17, 6957-6974, 2017
<https://doi.org/10.5194/acp-17-6957-2017>

Stratospheric sulfur injections have often been suggested as a cost-effective geoengineering method to prevent or slow down global warming. In geoengineering studies, these injections are commonly targeted to the Equator, where the yearly mean intensity of the solar radiation is the highest and from where the aerosols disperse globally due to the Brewer–Dobson Circulation. However, compensating for greenhouse gas-induced zonal warming by reducing solar radiation would require a relatively larger radiative forcing to the mid- and high latitudes and a lower forcing to the low latitudes than what is achieved by continuous equatorial injections. In this study we employ alternative aerosol injection scenarios to investigate if the resulting radiative forcing can be targeted to be zonally more uniform without decreasing the global the mean radiative forcing of stratospheric sulfur geoengineering. We used a global aerosol–climate model together with an Earth system model to study the radiative and climate effects of stratospheric sulfur injection scenarios with different injection areas. According to our simulations, varying the SO₂ injection area seasonally would result in a similar global mean cooling effect as injecting SO₂ to the Equator, but with a more uniform zonal distribution of shortwave radiative forcing. Compared to the case of equatorial injections, in the seasonally varying injection scenario where the maximum sulfur production from injected SO₂ followed the maximum of solar radiation, the shortwave radiative forcing decreased by 27 % over the Equator (the latitudes between 20° N and 20° S) and increased by 15 % over higher latitudes. Compared to the continuous injections to the Equator, in summer months the radiative forcing was increased by 17 and 14 % and in winter months decreased by 14 and 16 % in Northern and Southern hemispheres, respectively. However, these forcings do not translate into as large changes in temperatures. The changes in forcing would only lead to 0.05 K warmer winters and 0.05 K cooler summers in the Northern Hemisphere, which is roughly 3 % of the cooling resulting from solar radiation management scenarios studied here.

Quantifying pollution transport from the Asian monsoon anticyclone into the lower stratosphere

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Source: Atmos. Chem. Phys., 17, 7055-7066, 2017
<https://doi.org/10.5194/acp-17-7055-2017>

Pollution transport from the surface to the stratosphere within the Asian monsoon circulation may cause harmful effects on stratospheric chemistry and climate. Here, we investigate air mass transport from the monsoon anticyclone into the stratosphere using a Lagrangian chemistry transport model. We show how two main transport pathways from the anticyclone emerge: (i) into the tropical stratosphere (tropical pipe), and (ii) into the Northern Hemisphere (NH) extratropical lower stratosphere. Maximum anticyclone air mass fractions reach around 5 % in the tropical pipe and 15 % in the extratropical lowermost stratosphere over the course of a year. The anticyclone air mass fraction correlates well with satellite hydrogen cyanide (HCN) and carbon monoxide (CO) observations, confirming that pollution is transported deep into the tropical stratosphere from the Asian monsoon anticyclone. Cross-tropopause transport occurs in a vertical chimney, but with the pollutants transported quasi-horizontally along isentropes above the tropopause into the tropics and NH.

Characterization of fresh and aged organic aerosol emissions from meat charbroiling

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Source: Atmos. Chem. Phys., 17, 7143-7155, 2017
<https://doi.org/10.5194/acp-17-7143-2017>

Cooking emissions can be a significant source of fine particulate matter in urban areas. In this study the aerosol- and gas-phase emissions from meat charbroiling were characterized. Greek souvlakia with pork were cooked using a commercial charbroiler and a fraction of the emissions were introduced into a smog chamber where after a characterization phase they were exposed to UV illumination and oxidants. The particulate and gas phases were characterized by a high-resolution time-of-flight aerosol mass spectrometer (HR-ToF-AMS) and a proton-transfer-reaction mass spectrometer (PTR-MS) correspondingly. More than 99 % of the aerosol emitted was composed of organic compounds, while black carbon (BC) contributed 0.3 % and the inorganic species less than 0.5 % of the total aerosol mass. The initial O : C ratio was approximately 0.09 and increased up to 0.30 after a few hours of chemical aging (exposures of 1010 molecules cm⁻³ s for OH and 100 ppb h for ozone). The initial and aged AMS spectra differed considerably ($\theta = 27^\circ$). Ambient measurements were also conducted during Fat Thursday in Patras, Greece, when traditionally meat is charbroiled everywhere in the city. Positive matrix factorization (PMF) revealed that cooking organic aerosol (COA) reached up to 85 % of the total OA from 10:00 to 12:00 LST that day. The ambient COA factor in two major Greek cities had a mass spectrum during spring and summer similar to the aged meat charbroiling emissions. In contrast, the ambient COA factor during winter resembled strongly the fresh laboratory meat charbroiling emissions.

Formation of secondary organic aerosols from gas-phase emissions of heated cooking oils

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Source: Atmos. Chem. Phys., 17, 7333-7344, 2017
<https://doi.org/10.5194/acp-17-7333-2017>

Cooking emissions can potentially contribute to secondary organic aerosol (SOA) but remain poorly understood. In this study, formation of SOA from gas-phase emissions of five heated vegetable oils (i.e., corn, canola, sunflower, peanut and olive oils) was investigated in a potential aerosol mass (PAM) chamber. Experiments were conducted at 19–20 °C and 65–70 % relative humidity (RH). The characterization instruments included a scanning mobility particle sizer (SMPS) and a high-resolution time-of-flight aerosol mass spectrometer (HR-TOF-AMS). The efficiency of SOA production, in ascending order, was peanut oil, olive oil, canola oil, corn oil and sunflower oil. The major SOA precursors from heated cooking oils were related to the content of monounsaturated fat and omega-6 fatty acids in cooking oils. The average production rate of SOA, after aging at an OH exposure of 1.7×10^{11} molecules cm^{-3} s, was $1.35 \pm 0.30 \mu\text{g min}^{-1}$, 3 orders of magnitude lower compared with emission rates of fine particulate matter (PM_{2.5}) from heated cooking oils in previous studies. The mass spectra of cooking SOA highly resemble field-derived COA (cooking-related organic aerosol) in ambient air, with R₂ ranging from 0.74 to 0.88. The average carbon oxidation state (OS_C) of SOA was –1.51 to –0.81, falling in the range between ambient hydrocarbon-like organic aerosol (HOA) and semi-volatile oxygenated organic aerosol (SV-OOA), indicating that SOA in these experiments was lightly oxidized.

Diagnostic methods for atmospheric inversions of long-lived greenhouse gases

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Source: Atmos. Chem. Phys., 17, 7405-7421, 2017
<https://doi.org/10.5194/acp-17-7405-2017>

The ability to predict the trajectory of climate change requires a clear understanding of the emissions and uptake (i.e., surface fluxes) of long-lived greenhouse gases (GHGs). Furthermore, the development of climate policies is driving a need to constrain the budgets of anthropogenic GHG emissions. Inverse problems that couple atmospheric observations of GHG concentrations with an atmospheric chemistry and transport model have increasingly been used to gain insights into surface fluxes. Given the inherent technical challenges associated with their solution, it is imperative that objective approaches exist for the evaluation of such inverse problems. Because direct observation of fluxes at compatible spatiotemporal scales is rarely possible, diagnostics tools must rely on indirect measures. Here we review diagnostics that have been implemented in recent studies and discuss their use in informing adjustments to model setup. We group the diagnostics along a continuum starting with those that are most closely related to the scientific question being targeted, and ending with those most closely tied to the statistical and computational setup of the inversion. We thus begin with diagnostics based on

assessments against independent information (e.g., unused atmospheric observations, large-scale scientific constraints), followed by statistical diagnostics of inversion results, diagnostics based on sensitivity tests, and analyses of robustness (e.g., tests focusing on the chemistry and transport model, the atmospheric observations, or the statistical and computational framework), and close with the use of synthetic data experiments (i.e., observing system simulation experiments, OSSEs). We find that existing diagnostics provide a crucial toolbox for evaluating and improving flux estimates but, not surprisingly, cannot overcome the fundamental challenges associated with limited atmospheric observations or the lack of direct flux measurements at compatible scales. As atmospheric inversions are increasingly expected to contribute to national reporting of GHG emissions, the need for developing and implementing robust and transparent evaluation approaches will only grow.

Aerosol effects on the development of cumulus clouds over the Tibetan Plateau

Xu Zhou, Naifang Bei, Hongli Liu, Junji Cao, Li Xing, Wenfang Lei, Luisa T. Molina, and Guohui Li

Source: Atmos. Chem. Phys., 17, 7423-7434, 2017
<https://doi.org/10.5194/acp-17-7423-2017>

The aerosol–cloud interaction over the Tibetan Plateau has been investigated using a cloud-resolving weather research and forecasting model with a two-moment bulk microphysical scheme including aerosol effects on cloud condensation nuclei and ice nuclei. Two types of cumulus clouds with a similar convective available potential energy, occurring over the Tibetan Plateau (Cu-TP) and North China Plain (Cu-NCP) in August 2014, are simulated to explore the response of convective clouds to aerosols. A set of aerosol profiles is used in the simulations, with the surface aerosol number concentration varying from 20 to 9000 cm⁻³ and the sulfate mass concentration varying from 0.02 to 9.0 µg cm⁻³. Increasing aerosol concentrations generally enhances the cloud core updraft and maximum updraft, intensifying convections in Cu-TP and Cu-NCP. However, the core updraft is much stronger in Cu-TP than Cu-NCP, because of the early occurrence of the glaciation process in Cu-TP that is triggered at an elevation above 4000 m. The precipitation increases steadily with aerosol concentrations in Cu-NCP, caused by the suppression of the warm rain but occurrence of efficient mix-phased precipitation due to the reduced cloud droplet size. The precipitation in Cu-TP also increases with aerosol concentrations, but the precipitation enhancement is not substantial compared to that in Cu-NCP with high aerosol concentrations. The aerosol-induced intensification of convections in Cu-TP not only facilitates the precipitation but also transports more ice-phase hydrometeors into the upper troposphere to decrease the precipitation efficiency. Considering the very clean atmosphere over the Tibetan Plateau, elevated aerosol concentrations can remarkably enhance convections due to its specific topography, which not only warms the middle troposphere to influence the Asian summer monsoon but also delivers hydrometeors into the upper troposphere to allow more water vapor to travel into the lower stratosphere.

Methane emissions from dairies in the Los Angeles Basin

Camille Viatte, Thomas Lauvaux, Jacob K. Hedelius, Harrison Parker, Jia Chen, Taylor Jones, Jonathan E. Franklin, Aijun J. Deng, Brian Gaudet, Kristal Verhulst, Riley Duren, Debra Wunch, Coleen Roehl, Manvendra K. Dubey, Steve Wofsy, and Paul O. Wennberg

Source: *Atmos. Chem. Phys.*, 17, 7509-7528, 2017
<https://doi.org/10.5194/acp-17-7509-2017>

We estimate the amount of methane (CH₄) emitted by the largest dairies in the southern California region by combining measurements from four mobile solar-viewing ground-based spectrometers (EM27/SUN), in situ isotopic ¹³/₁₂CH₄ measurements from a CRDS analyzer (Picarro), and a high-resolution atmospheric transport simulation with a Weather Research and Forecasting model in large-eddy simulation mode (WRF-LES).

The remote sensing spectrometers measure the total column-averaged dry-air mole fractions of CH₄ and CO₂ (XCH₄ and XCO₂) in the near infrared region, providing information on total emissions of the dairies at Chino. Differences measured between the four EM27/SUN ranged from 0.2 to 22 ppb (part per billion) and from 0.7 to 3 ppm (part per million) for XCH₄ and XCO₂, respectively. To assess the fluxes of the dairies, these differential measurements are used in conjunction with the local atmospheric dynamics from wind measurements at two local airports and from the WRF-LES simulations at 111 m resolution.

Our top-down CH₄ emissions derived using the Fourier transform spectrometers (FTS) observations of 1.4 to 4.8 ppt s⁻¹ are in the low end of previous top-down estimates, consistent with reductions of the dairy farms and urbanization in the domain. However, the wide range of inferred fluxes points to the challenges posed by the heterogeneity of the sources and meteorology. Inverse modeling from WRF-LES is utilized to resolve the spatial distribution of CH₄ emissions in the domain. Both the model and the measurements indicate heterogeneous emissions, with contributions from anthropogenic and biogenic sources at Chino. A Bayesian inversion and a Monte Carlo approach are used to provide the CH₄ emissions of 2.2 to 3.5 ppt s⁻¹ at Chino.

Effect of mid-term drought on *Quercus pubescens* BVOCs' emission seasonality and their dependency on light and/or temperature

Amélie Saunier, Elena Ormeño, Christophe Boissard, Henri Wortham, Brice Temime-Roussel, Caroline Lecareux, Alexandre Armengaud, and Catherine Fernandez

Source: *Atmos. Chem. Phys.*, 17, 7555-7566, 2017
<https://doi.org/10.5194/acp-17-7555-2017>

Biogenic volatile organic compounds (BVOCs) emitted by plants represent a large source of carbon compounds released into the atmosphere, where they account for precursors of tropospheric ozone and

secondary organic aerosols. Being directly involved in air pollution and indirectly in climate change, understanding what factors drive BVOC emissions is a prerequisite for modeling their emissions and predict air pollution. The main algorithms currently used to model BVOC emissions are mainly light and/or temperature dependent. Additional factors such as seasonality and drought also influence isoprene emissions, especially in the Mediterranean region, which is characterized by a rather long drought period in summer. These factors are increasingly included in models but only for the principal studied BVOC, namely isoprene, but there are still some discrepancies in estimations of emissions. In this study, the main BVOCs emitted by *Quercus pubescens* – isoprene, methanol, acetone, acetaldehyde, formaldehyde, MACR, MVK and ISOPOOH (these three last compounds detected under the same m/z) – were monitored with a PTR-ToF-MS over an entire seasonal cycle during both in situ natural and amplified drought, which is expected with climate change. Amplified drought impacted all studied BVOCs by reducing emissions in spring and summer while increasing emissions in autumn. All six BVOCs monitored showed daytime light and temperature dependencies while three BVOCs (methanol, acetone and formaldehyde) also showed emissions during the night despite the absence of light under constant temperature. Moreover, methanol and acetaldehyde burst in the early morning and formaldehyde deposition and uptake were also punctually observed, which were not assessed by the classical temperature and light models.

Size-resolved chemical composition, effective density, and optical properties of biomass burning particles

Jinghao Zhai, Xiaohui Lu, Ling Li, Qi Zhang, Ci Zhang, Hong Chen, Xin Yang, and Jianmin Chen

Source: Atmos. Chem. Phys., 17, 7481-7493, 2017
<https://doi.org/10.5194/acp-17-7481-2017>

Biomass burning aerosol has an important impact on the global radiative budget. A better understanding of the correlations between the mixing states of biomass burning particles and their optical properties is the goal of a number of current studies. In this work, the effective density, chemical composition, and optical properties of rice straw burning particles in the size range of 50–400 nm were measured using a suite of online methods. We found that the major components of particles produced by burning rice straw included black carbon (BC), organic carbon (OC), and potassium salts, but the mixing states of particles were strongly size dependent. Particles of 50 nm had the smallest effective density (1.16 g cm^{-3}) due to a relatively large proportion of aggregate BC. The average effective densities of 100–400 nm particles ranged from 1.35 to 1.51 g cm^{-3} with OC and inorganic salts as dominant components. Both density distribution and single-particle mass spectrometry showed more complex mixing states in larger particles. Upon heating, the separation of the effective density distribution modes confirmed the external mixing state of less-volatile BC or soot and potassium salts. The size-resolved optical properties of biomass burning particles were investigated at two wavelengths ($\lambda = 450$ and 530 nm). The single-scattering albedo (SSA) showed the lowest value for 50 nm particles (0.741 ± 0.007 and 0.889 ± 0.006) because of the larger proportion of BC content. Brown carbon played an important role for the SSA of 100–400 nm particles. The Ångström absorption exponent (AAE) values for all particles were above 1.6, indicating the significant presence of brown carbon in all sizes.

Concurrent measurements in our work provide a basis for discussing the physicochemical properties of biomass burning aerosol and its effects on the global climate and atmospheric environment.

Methane emissions from dairies in the Los Angeles Basin

Camille Viatte, Thomas Lauvaux, Jacob K. Hedelius, Harrison Parker, Jia Chen, Taylor Jones, Jonathan E. Franklin, Aijun J. Deng, Brian Gaudet, Kristal Verhulst, Riley Duren, Debra Wunch, Coleen Roehl, Manvendra K. Dubey, Steve Wofsy, and Paul O. Wennberg

Source: Atmos. Chem. Phys., 17, 7509-7528, 2017
<https://doi.org/10.5194/acp-17-7509-2017>

We estimate the amount of methane (CH₄) emitted by the largest dairies in the southern California region by combining measurements from four mobile solar-viewing ground-based spectrometers (EM27/SUN), in situ isotopic ¹³/₁₂CH₄ measurements from a CRDS analyzer (Picarro), and a high-resolution atmospheric transport simulation with a Weather Research and Forecasting model in large-eddy simulation mode (WRF-LES).

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Particle size dependence of biogenic secondary organic aerosol molecular composition

Peijun Tu and Murray V. Johnston

Source: Atmos. Chem. Phys., 17, 7593-7603, 2017
<https://doi.org/10.5194/acp-17-7593-2017>

Formation of secondary organic aerosol (SOA) is initiated by the oxidation of volatile organic compounds (VOCs) in the gas phase whose products subsequently partition to the particle phase. Non-volatile molecules have a negligible evaporation rate and grow particles at their condensation rate. Semi-volatile molecules have a significant evaporation rate and grow particles at a much slower rate than their condensation rate. Particle phase chemistry may enhance particle growth if it transforms partitioned semi-volatile molecules into non-volatile products. In principle, changes in molecular composition as a function of particle size allow non-volatile molecules that have condensed from the gas phase (a surface-limited process) to be distinguished from those produced by particle phase reaction (a volume-limited process). In this work, SOA was produced by β -pinene ozonolysis in a flow tube reactor. Aerosol exiting the reactor was size-selected with a differential mobility analyzer, and individual particle sizes between 35 and 110 nm in diameter were characterized by on- and offline mass spectrometry. Both the average oxygen-to-carbon (O/C) ratio and carbon oxidation state (OSc) were found to decrease with increasing particle size, while the relative signal intensity of oligomers increased with increasing particle size. These results are consistent with oligomer formation primarily in the particle phase (accretion reactions, which become more favored as the volume-to-surface-area ratio of the particle increases). Analysis of a series of polydisperse SOA samples showed similar dependencies: as the mass loading increased (and average volume-to-surface-area ratio increased), the average O/C ratio and OSc decreased, while the relative intensity of oligomer ions increased. The results illustrate the potential impact that particle phase chemistry can have on biogenic SOA formation and the particle size range where this chemistry becomes important.

Meteorological context of the onset and end of the rainy season in Central Amazonia during the GoAmazon2014/5

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Source: Atmos. Chem. Phys., 17, 7671-7681, 2017
<https://doi.org/10.5194/acp-17-7671-2017>

The onset and demise of the rainy season in Amazonia are assessed in this study using meteorological data from the GoAmazon experiment, with a focus on the 2014–2015 rainy season. In addition, global reanalyses are also used to identify changes in circulation leading to the establishment of the rainy season in the region. Our results show that the onset occurred in January 2015, 2–3 pentads later than normal, and the rainy season during the austral summer of 2015 contained several periods with consecutive dry days in both Manacapuru and Manaus, which are not common for the wet season, and resulted in below-normal precipitation. The onset of the rainy season has been strongly associated with changes in large-scale weather conditions in the region due to the effect of the Madden–Julian Oscillation (MJO). Regional thermodynamic indices and the height of the boundary layer did not present a significant difference between the onset and demise of the wet season of 2015. This suggests that local changes, such as those in the regional thermodynamic characteristics, may not have influenced its

onset. Thus, variability of the large-scale circulation was responsible for regional convection and rainfall changes in Amazonia during the austral summer of 2014–2015.

Speciation of anthropogenic emissions of non-methane volatile organic compounds: a global gridded data set for 1970–2012

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Source: Atmos. Chem. Phys., 17, 7683–7701, 2017
<https://doi.org/10.5194/acp-17-7683-2017>

Non-methane volatile organic compounds (NMVOCs) include a large number of chemical species which differ significantly in their chemical characteristics and thus in their impacts on ozone and secondary organic aerosol formation. It is important that chemical transport models (CTMs) simulate the chemical transformation of the different NMVOC species in the troposphere consistently. In most emission inventories, however, only total NMVOC emissions are reported, which need to be decomposed into classes to fit the requirements of CTMs. For instance, the Emissions Database for Global Atmospheric Research (EDGAR) provides spatially resolved global anthropogenic emissions of total NMVOCs. In this study the EDGAR NMVOC inventory was revised and extended in time and in sectors. Moreover the new version of NMVOC emission data in the EDGAR database were disaggregated on a detailed sector resolution to individual species or species groups, thus enhancing the usability of the NMVOC emission data by the modelling community. Region- and source-specific speciation profiles of NMVOC species or species groups are compiled and mapped to EDGAR processes (detailed resolution of sectors), with corresponding quality codes specifying the quality of the mapping. Individual NMVOC species in different profiles are aggregated to 25 species groups, in line with the common classification of the Global Emissions Initiative (GEIA). Global annual grid maps with a resolution of $0.1^\circ \times 0.1^\circ$ for the period 1970–2012 are produced by sector and species. Furthermore, trends in NMVOC composition are analysed, taking road transport and residential sources in Germany and the United Kingdom (UK) as examples.

Enhanced toxicity of aerosol in fog conditions in the Po Valley, Italy

Stefano Decesari, Mohammad Hossein Sowlat, Sina Hasheminassab, Silvia Sandrini, Stefania Gilardoni, Maria Cristina Facchini, Sandro Fuzzi, and Constantinos Sioutas

Source: Atmos. Chem. Phys., 17, 7721–7731, 2017
<https://doi.org/10.5194/acp-17-7721-2017>

While numerous studies have demonstrated the association between outdoor exposure to atmospheric particulate matter (PM) and adverse health effects, the actual chemical species responsible for PM toxicological properties remain a subject of investigation. We provide here reactive oxygen species (ROS) activity data for PM samples collected at a rural site in the Po Valley, Italy, during the fog season (i.e., November–March). We show that the intrinsic ROS activity of Po Valley PM, which is mainly composed of biomass burning and secondary aerosols, is comparable to that of traffic-related particles in urban areas. The airborne concentration of PM components responsible for the ROS activity decreases in fog conditions, when water-soluble species are scavenged within the droplets. Due to this partitioning effect of fog, the measured ROS activity of fog water was contributed mainly by water-soluble organic carbon (WSOC) and secondary inorganic ions rather than by transition metals. We found that the intrinsic ROS activity of fog droplets is even greater (> 2.5 times) than that of the PM on which droplets are formed, indicating that redox-active compounds are not only scavenged from the particulate phase, but are also produced within the droplets. Therefore, even if fog formation exerts a scavenging effect on PM mass and redox-active compounds, the aqueous-phase formation of reactive secondary organic compounds can eventually enhance ROS activity of PM when fog evaporates. These findings, based on a case study during a field campaign in November 2015, indicate that a significant portion of airborne toxicity in the Po Valley is largely produced by environmental conditions (fog formation and fog processing) and not simply by the emission and transport of pollutants.

Improved provincial emission inventory and speciation profiles of anthropogenic non-methane volatile organic compounds: a case study for Jiangsu, China

Yu Zhao, Pan Mao, Yaduan Zhou, Yang Yang, Jie Zhang, Shekou Wang, Yanping Dong, Fangjian Xie, Yiyong Yu, and Wenqing Li

Source: Atmos. Chem. Phys., 17, 7733-7756, 2017
<https://doi.org/10.5194/acp-17-7733-2017>

Non-methane volatile organic compounds (NMVOCs) are the key precursors of ozone (O₃) and secondary organic aerosol (SOA) formation. Accurate estimation of their emissions plays a crucial role in air quality simulation and policy making. We developed a high-resolution anthropogenic NMVOC emission inventory for Jiangsu in eastern China from 2005 to 2014, based on detailed information of individual local sources and field measurements of source profiles of the chemical industry. A total of 56 NMVOCs samples were collected in nine chemical plants and were then analyzed with a gas chromatography – mass spectrometry system (GC-MS). Source profiles of stack emissions from synthetic rubber, acetate fiber, polyether, vinyl acetate and ethylene production, and those of fugitive emissions from ethylene, butanol and octanol, propylene epoxide, polyethylene and glycol production were obtained. Various manufacturing technologies and raw materials led to discrepancies in source profiles between our domestic field tests and foreign results for synthetic rubber and ethylene production. The provincial NMVOC emissions were calculated to increase from 1774 Gg in 2005 to 2507 Gg in 2014, and relatively large emission densities were found in cities along the Yangtze River with developed economies and industries. The estimates were larger than those from most other available inventories, due mainly to the complete inclusion of emission sources and to the elevated activity levels from plant-by-plant investigation in this work. Industrial processes and solvent use were the largest contributing

sectors, and their emissions were estimated to increase, respectively, from 461 to 958 and from 38 to 966 Gg. Alkanes, aromatics and oxygenated VOCs (OVOCs) were the most important species, accounting for 25.9–29.9, 20.8–23.2 and 18.2–21.0 % to annual total emissions, respectively. Quantified with a Monte Carlo simulation, the uncertainties of annual NMVOC emissions vary slightly through the years, and the result for 2014 was –41 to +93 %, expressed as 95 % confidence intervals (CI). Reduced uncertainty was achieved compared to previous national and regional inventories, attributed partly to the detailed classification of emission sources and to the use of information at plant level in this work. Discrepancies in emission estimation were explored for the chemical and refinery sectors with various data sources and methods. Compared with the Multi-resolution Emission Inventory for China (MEIC), the spatial distribution of emissions in this work were more influenced by the locations of large point sources, and smaller emissions were found in urban area for developed cities in southern Jiangsu. In addition, discrepancies were found between this work and MEIC in the speciation of NMVOC emissions under the atmospheric chemistry mechanisms CB05 and SAPRC99. The difference in species OLE1 resulted mainly from the updated source profile of building paint use and the differences in other species from the varied sector contributions to emissions in the two inventories. The Community Multi-scale Air Quality (CMAQ) model simulation was applied to evaluate the two inventories, and better performance (indicated by daily 1 h maximum O₃ concentrations in Nanjing) were found for January, April and October 2012 when the provincial inventory was used.

Secondary inorganic aerosols in Europe: sources and the significant influence of biogenic VOC emissions, especially on ammonium nitrate

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Source: Atmos. Chem. Phys., 17, 7757–7773, 2017
<https://doi.org/10.5194/acp-17-7757-2017>

Contributions of various anthropogenic sources to the secondary inorganic aerosol (SIA) in Europe as well as the role of biogenic emissions on SIA formation were investigated using the three-dimensional regional model CAMx (comprehensive air quality model with extensions). Simulations were carried out for two periods of EMEP field campaigns, February–March 2009 and June 2006, which are representative of cold and warm seasons, respectively. Biogenic volatile organic compounds (BVOCs) are known mainly as precursors of ozone and secondary organic aerosol (SOA), but their role on inorganic aerosol formation has not attracted much attention so far. In this study, we showed the importance of the chemical reactions of BVOCs and how they affect the oxidant concentrations, leading to significant changes, especially in the formation of ammonium nitrate. A sensitivity test with doubled BVOC emissions in Europe during the warm season showed a large increase in secondary organic aerosol (SOA) concentrations (by about a factor of two), while particulate inorganic nitrate concentrations decreased by up to 35 %, leading to a better agreement between the model results and measurements. Sulfate concentrations decreased as well; the change, however, was smaller. The changes in inorganic nitrate and sulfate concentrations occurred at different locations in Europe, indicating the importance of precursor gases and biogenic emission types for the negative correlation between BVOCs and SIA. Further analysis of the data suggested that reactions of the additional terpenes with nitrate radicals at night were responsible for the decline in inorganic nitrate formation, whereas oxidation of BVOCs with

OH radicals led to a decrease in sulfate. Source apportionment results suggest that the main anthropogenic source of precursors leading to formation of particulate inorganic nitrate is road transport (SNAP7; see Table 1 for a description of the categories), whereas combustion in energy and transformation industries (SNAP1) was the most important contributor to sulfate particulate mass. Emissions from international shipping were also found to be very important for both nitrate and sulfate formation in Europe. In addition, we also examined contributions from the geographical source regions to SIA concentrations in the most densely populated region of Switzerland, the Swiss Plateau.

Measurement of scattering and absorption properties of dust aerosol in a Gobi farmland region of northwestern China – a potential anthropogenic influence

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Source: Atmos. Chem. Phys., 17, 7775-7792, 2017
<https://doi.org/10.5194/acp-17-7775-2017>

We conducted a comprehensive field campaign to explore the optical characteristics of mineral dust in Dunhuang farmland near the Gobi Desert of northwest China during spring of 2012. The day-to-day and diurnal variations of dust aerosol showed prominent features throughout the experiment, primarily attributable to frequent dust events and local anthropogenic emissions. The overall average mass concentrations of the particulate matter with an aerodynamic diameter less than 10 μm (PM₁₀), light scattering coefficient (σ_{sp} , 670), absorption coefficient (σ_{ap} , 670), and single-scattering albedo (SSA₆₇₀) were $113 \pm 169 \mu\text{g m}^{-3}$, $53.3 \pm 74.8 \text{ Mm}^{-1}$, $3.2 \pm 2.4 \text{ Mm}^{-1}$, and 0.913 ± 0.05 , respectively, which were comparable to the background levels in the southern United States but smaller than those in the eastern and other northwestern Chinese cities. The anthropogenic dust produced by agricultural cultivations (e.g., land planning, plowing, and disking) exerted a significant superimposed effect on high dust concentrations in Dunhuang farmland prior to the growing season (i.e., from 1 April to 10 May). Strong south valley wind and vertical mixing in daytime scavenged the pollution, and the weak northeast mountain wind and stable inversion layer at night favorably accumulated the air pollutants near the surface. In the afternoon (13:00–18:00 LT, local time), mean SSA₆₇₀ was 0.945 ± 0.04 predominantly from dust particles, whereas finer particles and lower SSA₆₇₀ values (~ 0.90 – 0.92) were measured at night, suggesting the potential influence by the mixed dust pollutants. During a typical biomass burning event on 4 April 2012, σ_{ap} , 670 increased from ~ 2.0 to 4.75 Mm^{-1} and SSA₆₇₀ changed from ~ 0.90 to ~ 0.83 , implying remarkable modification of aerosol absorptive properties induced by human activities. The findings of this study would help to advance an in-depth understanding of the interaction among dust aerosol, atmospheric chemistry, and climate change in a desert source region.

Air stagnation in China (1985–2014): climatological mean features and trends

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Source: *Atmos. Chem. Phys.*, 17, 7793–7805, 2017
<https://doi.org/10.5194/acp-17-7793-2017>

Air stagnation is an important meteorological measure of unfavorable air pollution conditions, but little is known about it in China. We conducted a comprehensive investigation of air stagnation in China from January 1985 to December 2014 based on sounding and surface observations from 81 stations. The stagnation criteria were revised to account for the large topographical diversity in the country. It is found that the annual mean of air stagnation occurrences is closely related to general topography and climate features. Two basins in the northwest and southwest of China, the Tarim and Sichuan basins, exhibit the most frequent stagnation occurrence (50 % of days per year), whereas two plateaus (the Qinghai–Tibet Plateau and the Inner Mongolian plateau) and the eastern coastal areas experience the least (20 % of days per year). Over the whole country, air stagnation is at a maximum in summer and a minimum in winter, except for Urumchi, a major city in northwestern China where stagnation maintains a rather constant value year round with a minimum in spring. There is a nationwide positive trend in stagnation occurrence during 1985–2014, with the strongest increasing centers over Shandong Peninsula in eastern China and southern Shaanxi in central China. Changes in air stagnation occurrences are dependent on three components (upper- and lower-air winds and precipitation-free days). This shows that the behavior of upper-air wind speeds is the main driver of the spatial distribution and trends in air stagnation, followed by near-surface winds and dry days, which contribute the least.

Estimating the size of a methane emission point source at different scales: from local to landscape

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Source: *Atmos. Chem. Phys.*, 17, 7839–7851, 2017
<https://doi.org/10.5194/acp-17-7839-2017>

High methane (CH₄) mixing ratios (up to 4 ppm) have occurred sporadically at our measurement site in Haddenham, Cambridgeshire, since July 2012. Isotopic measurements and back trajectories show that the source is the Waterbeach Waste Management Park 7 km SE of Haddenham. To investigate this further, measurements were made on 30 June and 1 July 2015 at other locations nearer to the source. Landfill emissions have been estimated using three different approaches at different scales; near source using the WindTrax inversion dispersion model, middle distance using a Gaussian plume (GP) model and at the landscape scale using the Numerical Atmospheric Modelling Environment (NAME) Inversion Technique for Emission Modelling (InTEM) inversion. The emission estimates derived using the WindTrax and Gaussian plume (GP) approaches agree well for the period of intense observations. Applying the Gaussian plume approach to all periods of elevated measurements seen at Haddenham produces year-

round and monthly landfill emission estimates with an estimated annual emission of 11.6 Gg CH₄ yr⁻¹. The monthly emission estimates are highest in winter (2160 kg h⁻¹ in February) and lowest in summer (620 kg h⁻¹ in July). These data identify the effects of environmental conditions on landfill CH₄ production and highlight the importance of year-round measurements to capture seasonal variability in CH₄ emission.

Optical, physical and chemical properties of aerosols transported to a coastal site in the western Mediterranean: a focus on primary marine aerosols

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Source: Atmos. Chem. Phys., 17, 7891-7915, 2017
<https://doi.org/10.5194/acp-17-7891-2017>

As part of the ChArMEx-ADRI-MED campaign (summer 2013), ground-based in situ observations were conducted at the Ersa site (northern tip of Corsica; 533 m a.s.l.) to characterise the optical, physical and chemical properties of aerosols. During the observation period, a major influence of primary marine aerosols was detected (22–26 June), with a mass concentration reaching up to 6.5 µg m⁻³ and representing more than 40 % of the total PM₁₀ mass concentration. Its relatively low ratio of chloride to sodium (average of 0.57) indicates a fairly aged sea salt aerosol at Ersa. In this work, an original data set, obtained from online real-time instruments (ATOFMS, PILS-IC) has been used to characterise the ageing of primary marine aerosols (PMAs). During this PMA period, the mixing of fresh and aged PMAs was found to originate from both local and regional (Gulf of Lion) emissions, according to local wind measurements and FLEXPART back trajectories. Two different aerosol regimes have been identified: a dust outbreak (dust) originating from Algeria/Tunisia, and a pollution period with aerosols originating from eastern Europe, which includes anthropogenic and biomass burning sources (BBP). The optical, physical and chemical properties of the observed aerosols, as well as their local shortwave (SW) direct radiative effect (DRE) in clear-sky conditions, are compared for these three periods in order to assess the importance of the direct radiative impact of PMAs compared to other sources above the western Mediterranean Basin.

As expected, AERONET retrievals indicate a relatively low local SW DRF during the PMA period with mean values of -11 ± 4 at the surface and -8 ± 3 W m⁻² at the top of the atmosphere (TOA). In comparison, our results indicate that the dust outbreak observed at our site during the campaign, although of moderate intensity (AOD of 0.3–0.4 at 440 nm and column-integrated SSA of 0.90–0.95), induced a local instantaneous SW DRF that is nearly 3 times the effect calculated during the PMA period, with maximum values up to -40 W m⁻² at the surface. A similar range of values were found for the BBP period to those during the dust period (SW DRF at the surface and TOA of -23 ± 6 and -15 ± 4 W m⁻² respectively).

The multiple sources of measurements at Ersa allowed the detection of a PMA-dominant period and

their characterisation in terms of ageing, origin, transport, optical and physical properties and direct climatic impact.

Aerosol–landscape–cloud interaction: signatures of topography effect on cloud droplet formation

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Source: Atmos. Chem. Phys., 17, 7955–7964, 2017
<https://doi.org/10.5194/acp-17-7955-2017>

Long-term in situ measurements of aerosol–cloud interactions are usually performed in measurement stations residing on hills, mountains, or high towers. In such conditions, the surface topography of the surrounding area can affect the measured cloud droplet distributions by increasing turbulence or causing orographic flows and thus the observations might not be representative for a larger scale. The objective of this work is to analyse, how the local topography affects the observations at Puijo measurement station, which is located in the 75 m high Puijo tower, which itself stands on a 150 m high hill. The analysis of the measurement data shows that the observed cloud droplet number concentration mainly depends on the cloud condensation nuclei (CCN) concentration. However, when the wind direction aligns with the direction of the steepest slope of the hill, a clear topography effect is observed. This finding was further analysed by simulating 3-D flow fields around the station and by performing trajectory ensemble modelling of aerosol- and wind-dependent cloud droplet formation. The results showed that in typical conditions, with geostrophic winds of about 10 m s^{-1} , the hill can cause updrafts of up to 1 m s^{-1} in the air parcels arriving at the station. This is enough to produce in-cloud supersaturations (SSs) higher than typically found at the cloud base of $\sim 0.2 \%$, and thus additional cloud droplets may form inside the cloud. In the observations, this is seen in the form of a bimodal cloud droplet size distribution. The effect is strongest with high winds across the steepest slope of the hill and with low liquid water contents, and its relative importance quickly decreases as these conditions are relaxed. We therefore conclude that, after careful screening for wind speed and liquid water content, the observations at Puijo measurement station can be considered representative for clouds in a boreal environment.

Size distribution and source of black carbon aerosol in urban Beijing during winter haze episodes

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Source: Atmos. Chem. Phys., 17, 7965–7975, 2017
<https://doi.org/10.5194/acp-17-7965-2017>

Black carbon (BC) has important impact on climate and environment due to its light absorption ability, which greatly depends on its physicochemical properties including morphology, size and mixing state. The size distribution of the refractory BC (rBC) was investigated in urban Beijing in the late winter of 2014, during which there were frequent haze events, through analysis of measurements obtained using a single-particle soot photometer (SP2). By assuming void-free rBC with a density of 1.8 g cm^{-3} , the mass of the rBC showed an approximately lognormal distribution as a function of the volume-equivalent diameter (VED), with a peak diameter of 213 nm. Larger VED values of the rBC were observed during polluted periods than on clean days, implying an alteration in the rBC sources, as the size distribution of the rBC from a certain source was relative stable, and VED of an individual rBC varied little once it was emitted into the atmosphere. The potential source contribution function analysis showed that air masses from the south to east of the observation site brought higher rBC loadings with more thick coatings and larger core sizes. The mean VED of the rBC presented a significant linear correlation with the number fraction of thickly coated rBC, extrapolating to be $\sim 150 \text{ nm}$ for the completely non-coated or thinly coated rBC. It was considered as the typical mean VED of the rBC from local traffic sources in this study. Local traffic was estimated to contribute 35 to 100 % of the hourly rBC mass concentration with a mean of 59 % during the campaign. Lower local traffic contributions were observed during polluted periods, suggesting increasing contributions from other sources (e.g., coal combustion and biomass burning) to the rBC. Thus, the heavy pollution in Beijing was greatly influenced by other sources in addition to the local traffic.

Contributions of mobile, stationary and biogenic sources to air pollution in the Amazon rainforest: a numerical study with the WRF-Chem model

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Source: Atmos. Chem. Phys., 17, 7977-7995, 2017
<https://doi.org/10.5194/acp-17-7977-2017>

This paper evaluates the contributions of the emissions from mobile, stationary and biogenic sources on air pollution in the Amazon rainforest by using the Weather Research and Forecasting with Chemistry (WRF-Chem) model. The analyzed air pollutants were CO, NO_x, SO₂, O₃, PM_{2.5}, PM₁₀ and volatile organic compounds (VOCs). Five scenarios were defined in order to evaluate the emissions by biogenic, mobile and stationary sources, as well as a future scenario to assess the potential air quality impact of doubled anthropogenic emissions. The stationary sources explain the highest concentrations for all air pollutants evaluated, except for CO, for which the mobile sources are predominant. The anthropogenic sources considered resulted an increasing in the spatial peak-temporal average concentrations of pollutants in 3 to 2780 times in relation to those with only biogenic sources. The future scenario showed an increase in the range of 3 to 62 % in average concentrations and 45 to 109 % in peak concentrations depending on the pollutant. In addition, the spatial distributions of the scenarios has shown that the air pollution plume from the city of Manaus is predominantly transported west and southwest, and it can reach hundreds of kilometers in length.

Contribution of different processes to changes in tropical lower-stratospheric water vapor in chemistry–climate models

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Source: Atmos. Chem. Phys., 17, 8031-8044, 2017
<https://doi.org/10.5194/acp-17-8031-2017>

Variations in tropical lower-stratospheric humidity influence both the chemistry and climate of the atmosphere. We analyze tropical lower-stratospheric water vapor in 21st century simulations from 12 state-of-the-art chemistry–climate models (CCMs), using a linear regression model to determine the factors driving the trends and variability. Within CCMs, warming of the troposphere primarily drives the long-term trend in stratospheric humidity. This is partially offset in most CCMs by an increase in the strength of the Brewer–Dobson circulation, which tends to cool the tropical tropopause layer (TTL). We also apply the regression model to individual decades from the 21st century CCM runs and compare them to a regression of a decade of observations. Many of the CCMs, but not all, compare well with these observations, lending credibility to their predictions. One notable deficiency is that most CCMs underestimate the impact of the quasi-biennial oscillation on lower-stratospheric water vapor. Our analysis provides a new and potentially superior way to evaluate model trends in lower-stratospheric humidity.

Intercomparison of meteorological analyses and trajectories in the Antarctic lower stratosphere with Concordiasi superpressure balloon observations

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Source: Atmos. Chem. Phys., 17, 8045-8061, 2017
<https://doi.org/10.5194/acp-17-8045-2017>

In this study we compared temperatures and horizontal winds of meteorological analyses in the Antarctic lower stratosphere, a region of the atmosphere that is of major interest regarding chemistry and dynamics of the polar vortex. The study covers the European Centre for Medium-Range Weather Forecasts (ECMWF) operational analysis, the ERA-Interim reanalysis, the Modern-Era Retrospective analysis for Research and Applications version 1 and 2 (MERRA and MERRA-2), and the National Centers for Environmental Prediction and National Center for Atmospheric Research (NCEP/NCAR) reanalysis. The comparison was performed with respect to long-duration observations from 19 superpressure balloon flights during the Concordiasi field campaign in September 2010 to January 2011. Most of the balloon measurements were conducted at altitudes of 17–18.5 km and latitudes of 60–85° S. We found that large-scale state temperatures of the analyses have a mean precision of 0.5–1.4 K and a warm bias of 0.4–2.1 K with respect to the balloon data. Zonal and meridional winds have a mean precision of 0.9–2.3 m s⁻¹ and a bias below ± 0.5 m s⁻¹. Standard deviations related to small-scale fluctuations due to

gravity waves are reproduced at levels of 15–60 % for temperature and 30–60 % for the horizontal winds. Considering the fact that the balloon observations have been assimilated into all analyses, except for NCEP/NCAR, notable differences found here indicate that other observations, the forecast models, and the data assimilation procedures have a significant impact on the analyses as well. We also used the balloon observations to evaluate trajectory calculations with our new Lagrangian transport model Massive-Parallel Trajectory Calculations (MPTRAC), where vertical motions of simulated trajectories were nudged to pressure measurements of the balloons. We found relative horizontal transport deviations of 4–12 % and error growth rates of 60–170 km day⁻¹ for 15-day trajectories. Dispersion simulations revealed some difficulties with the representation of subgrid-scale wind fluctuations in MPTRAC, as the spread of air parcels simulated with different analyses was not consistent. However, although case studies suggest that the accuracy of trajectory calculations is influenced by meteorological complexity, diffusion generally does not contribute significantly to transport deviations in our analysis. Overall, evaluation results are satisfactory and compare well to earlier studies using superpressure balloon observations.

Effect of volcanic aerosol on stratospheric NO₂ and N₂O₅ from 2002–2014 as measured by Odin-OSIRIS and Envisat-MIPAS

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Source: Atmos. Chem. Phys., 17, 8063–8080, 2017
<https://doi.org/10.5194/acp-17-8063-2017>

Following the large volcanic eruptions of Pinatubo in 1991 and El Chichón in 1982, decreases in stratospheric NO₂ associated with enhanced aerosol were observed. The Optical Spectrograph and Infrared Imaging Spectrometer (OSIRIS) measured the widespread enhancements of stratospheric aerosol following seven volcanic eruptions between 2002 and 2014, although the magnitudes of these eruptions were all much smaller than the Pinatubo and El Chichón eruptions. In order to isolate and quantify the relationship between volcanic aerosol and NO₂, NO₂ anomalies were calculated using measurements from OSIRIS and the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS). In the tropics, variability due to the quasi-biennial oscillation was subtracted from the time series. OSIRIS profile measurements indicate that the strongest anticorrelations between NO₂ and volcanic aerosol extinction were for the 5 km layer starting ~3 km above the climatological mean tropopause at the given latitude. OSIRIS stratospheric NO₂ partial columns in this layer were found to be smaller than background NO₂ levels during these aerosol enhancements by up to ~60 % with typical Pearson correlation coefficients of $R \sim -0.7$. MIPAS also observed decreases in NO₂ partial columns during periods affected by volcanic aerosol, with percent differences of up to ~25 % relative to background levels. An even stronger anticorrelation was observed between OSIRIS aerosol optical depth and MIPAS N₂O₅ partial columns, with $R \sim -0.9$, although no link with MIPAS HNO₃ was observed. The variation in OSIRIS NO₂ with increasing aerosol was found to be consistent with simulations from a photochemical box model within the estimated model uncertainty.

Source apportionment of NMVOCs in the Kathmandu Valley during the SusKat-ABC international field campaign using positive matrix factorization

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Source: Atmos. Chem. Phys., 17, 8129-8156, 2017
<https://doi.org/10.5194/acp-17-8129-2017>

A positive matrix factorization model (US EPA PMF version 5.0) was applied for the source apportionment of the dataset of 37 non-methane volatile organic compounds (NMVOCs) measured from 19 December 2012 to 30 January 2013 during the SusKat-ABC international air pollution measurement campaign using a proton-transfer-reaction time-of-flight mass spectrometer in the Kathmandu Valley. In all, eight source categories were identified with the PMF model using the new constrained model operation mode. Unresolved industrial emissions and traffic source factors were the major contributors to the total measured NMVOC mass loading (17.9 and 16.8 %, respectively) followed by mixed industrial emissions (14.0 %), while the remainder of the source was split approximately evenly between residential biofuel use and waste disposal (10.9 %), solvent evaporation (10.8 %), biomass co-fired brick kilns (10.4 %), biogenic emissions (10.0 %) and mixed daytime factor (9.2 %). Conditional probability function (CPF) analyses were performed to identify the physical locations associated with different sources. Source contributions to individual NMVOCs showed that biomass co-fired brick kilns significantly contribute to the elevated concentrations of several health relevant NMVOCs such as benzene. Despite the highly polluted conditions, biogenic emissions had the largest contribution (24.2 %) to the total daytime ozone production potential, even in winter, followed by solvent evaporation (20.2 %), traffic (15.0 %) and unresolved industrial emissions (14.3 %). Secondary organic aerosol (SOA) production had approximately equal contributions from biomass co-fired brick kilns (28.9 %) and traffic (28.2 %). Comparison of PMF results based on the in situ data versus REAS v2.1 and EDGAR v4.2 emission inventories showed that both the inventories underestimate the contribution of traffic and do not take the contribution of brick kilns into account. In addition, the REAS inventory overestimates the contribution of residential biofuel use and underestimates the contribution of solvent use and industrial sources in the Kathmandu Valley. The quantitative source apportionment of major NMVOC sources in the Kathmandu Valley based on this study will aid in improving hitherto largely unvalidated bottom-up NMVOC emission inventories, enabling more focused mitigation measures and improved parameterizations in chemical transport models.

Investigation of the mixing layer height derived from ceilometer measurements in the Kathmandu Valley and implications for local air quality

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Source: Atmos. Chem. Phys., 17, 8157-8176, 2017
<https://doi.org/10.5194/acp-17-8157-2017>

In this study 1 year of ceilometer measurements taken in the Kathmandu Valley, Nepal, in the framework of the SusKat project (A Sustainable Atmosphere for the Kathmandu Valley) were analysed to investigate the diurnal variation of the mixing layer height (MLH) and its dependency on the meteorological conditions. In addition, the impact of the MLH on the temporal variation and the magnitude of the measured black carbon concentrations are analysed for each season. Based on the assumption that black carbon aerosols are vertically well mixed within the mixing layer and the finding that the mixing layer varies only little during night time and morning hours, black carbon emission fluxes are estimated for these hours and per month. Even though this method is relatively simple, it can give an observationally based first estimate of the black carbon emissions in this region, especially illuminating the seasonal cycle of the emission fluxes.

The monthly minimum median MLH values typically range between 150 and 200 m during night and early morning hours, the monthly maximum median values between 625 m in July and 1460 m in March. Seasonal differences are not only found in the absolute MLHs, but also in the duration of the typical daytime maximum ranging between 2 and 3 h in January and 6–7 h in May. During the monsoon season a diurnal cycle has been observed with the smallest amplitude (typically between 400 and 500 m), with the lowest daytime mixing height of all seasons (maximum monthly median values typically between 600 and 800 m), and also the highest night-time and early morning mixing height of all seasons (minimum monthly median values typically between 200 and 220 m). These characteristics can mainly be explained with the frequently present clouds and the associated reduction in incoming solar radiation and outgoing longwave radiation.

In general, the black carbon concentrations show a clear anticorrelation with MLH measurements, although this relation is less pronounced in the monsoon season. The daily evolution of the black carbon diurnal cycle differs between the seasons, partly due to the different meteorological conditions including the MLH. Other important reasons are the different main emission sources and their diurnal variations in the individual seasons. The estimation of the black carbon emission flux for the morning hours show a clear seasonal cycle with maximum values in December to April. Compared to the emission flux values provided by different emission databases for this region, the estimated values here are considerably higher. Several possible sources of uncertainty are considered, and even the absolute lower bound of the emissions based on our methodology is higher than in most emissions datasets, providing strong evidence that the black carbon emissions for this region have likely been underestimated in modelling studies thus far.

The boundary condition for vertical velocity and its interdependence with surface gas exchange

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Source: Atmos. Chem. Phys., 17, 8177-8187, 2017
<https://doi.org/10.5194/acp-17-8177-2017>

The law of conservation of linear momentum is applied to surface gas exchanges, employing scale analysis to diagnose the vertical velocity (w) in the boundary layer. Net upward momentum in the surface layer is forced by evaporation (E) and defines non-zero vertical motion, with a magnitude defined by the ratio of E to the air density, as $w = E/\rho$. This is true even right down at the surface where the boundary condition is $w|_0 = E/\rho|_0$ (where $w|_0$ and $\rho|_0$ represent the vertical velocity and density of air at the surface). This Stefan flow velocity implies upward transport of a non-diffusive nature that is a general feature of the troposphere but is of particular importance at the surface, where it assists molecular diffusion with upward gas migration (of H_2O , for example) but opposes that of downward-diffusing species like CO_2 during daytime. The definition of flux–gradient relationships (eddy diffusivities) requires rectification to exclude non-diffusive transport, which does not depend on scalar gradients. At the microscopic scale, the role of non-diffusive transport in the process of evaporation from inside a narrow tube – with vapour transport into an overlying, horizontal airstream – was described long ago in classical mechanics and is routinely accounted for by chemical engineers, but has been neglected by scientists studying stomatal conductance. Correctly accounting for non-diffusive transport through stomata, which can appreciably reduce net CO_2 transport and marginally boost that of water vapour, should improve characterisations of ecosystem and plant functioning.

Observation- and model-based estimates of particulate dry nitrogen deposition to the oceans

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Source: *Atmos. Chem. Phys.*, 17, 8189–8210, 2017
<https://doi.org/10.5194/acp-17-8189-2017>

Anthropogenic nitrogen (N) emissions to the atmosphere have increased significantly the deposition of nitrate (NO_3^-) and ammonium (NH_4^+) to the surface waters of the open ocean, with potential impacts on marine productivity and the global carbon cycle. Global-scale understanding of the impacts of N deposition to the oceans is reliant on our ability to produce and validate models of nitrogen emission, atmospheric chemistry, transport and deposition. In this work, ~2900 observations of aerosol NO_3^- and NH_4^+ concentrations, acquired from sampling aboard ships in the period 1995–2012, are used to assess the performance of modelled N concentration and deposition fields over the remote ocean. Three ocean regions (the eastern tropical North Atlantic, the northern Indian Ocean and northwest Pacific) were selected, in which the density and distribution of observational data were considered sufficient to provide effective comparison to model products. All of these study regions are affected by transport and deposition of mineral dust, which alters the deposition of N, due to uptake of nitrogen oxides (NO_x) on mineral surfaces.

Assessment of the impacts of atmospheric N deposition on the ocean requires atmospheric chemical transport models to report deposition fluxes; however, these fluxes cannot be measured over the ocean. Modelling studies such as the Atmospheric Chemistry and Climate Model Intercomparison

Project (ACCMIP), which only report deposition flux, are therefore very difficult to validate for dry deposition. Here, the available observational data were averaged over a $5^{\circ} \times 5^{\circ}$ grid and compared to ACCMIP dry deposition fluxes (ModDep) of oxidised N (NO_y) and reduced N (NH_x) and to the following parameters from the Tracer Model 4 of the Environmental Chemical Processes Laboratory (TM4): ModDep for NO_y, NH_x and particulate NO₃⁻ and NH₄⁺, and surface-level particulate NO₃⁻ and NH₄⁺ concentrations. As a model ensemble, ACCMIP can be expected to be more robust than TM4, while TM4 gives access to speciated parameters (NO₃⁻ and NH₄⁺) that are more relevant to the observed parameters and which are not available in ACCMIP. Dry deposition fluxes (CalDep) were calculated from the observed concentrations using estimates of dry deposition velocities. Model–observation ratios (RA, n), weighted by grid-cell area and number of observations, were used to assess the performance of the models. Comparison in the three study regions suggests that TM4 overestimates NO₃⁻ concentrations (RA, n = 1.4–2.9) and underestimates NH₄⁺ concentrations (RA, n = 0.5–0.7), with spatial distributions in the tropical Atlantic and northern Indian Ocean not being reproduced by the model. In the case of NH₄⁺ in the Indian Ocean, this discrepancy was probably due to seasonal biases in the sampling. Similar patterns were observed in the various comparisons of CalDep to ModDep (RA, n = 0.6–2.6 for NO₃⁻, 0.6–3.1 for NH₄⁺). Values of RA, n for NH_x CalDep–ModDep comparisons were approximately double the corresponding values for NH₄⁺ CalDep–ModDep comparisons due to the significant fraction of gas-phase NH₃ deposition incorporated in the TM4 and ACCMIP NH_x model products. All of the comparisons suffered due to the scarcity of observational data and the large uncertainty in dry deposition velocities used to derive deposition fluxes from concentrations. These uncertainties have been a major limitation on estimates of the flux of material to the oceans for several decades. Recommendations are made for improvements in N deposition estimation through changes in observations, modelling and model–observation comparison procedures. Validation of modelled dry deposition requires effective comparisons to observable aerosol-phase species' concentrations, and this cannot be achieved if model products only report dry deposition flux over the ocean.

Estimating daily surface NO₂ concentrations from satellite data – a case study over Hong Kong using land use regression models

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Source: Atmos. Chem. Phys., 17, 8211–8230, 2017
<https://doi.org/10.5194/acp-17-8211-2017>

Land use regression (LUR) models have been used in epidemiology to determine the fine-scale spatial variation in air pollutants such as nitrogen dioxide (NO₂) in cities and larger regions. However, they are often limited in their temporal resolution, which may potentially be rectified by employing the synoptic coverage provided by satellite measurements. In this work a mixed-effects LUR model is developed to model daily surface NO₂ concentrations over the Hong Kong SAR during the period 2005–2015. In situ measurements from the Hong Kong Air Quality Monitoring Network, along with tropospheric vertical column density (VCD) data from the OMI, GOME-2A, and SCIAMACHY satellite instruments were combined with fine-scale land use parameters to provide the spatiotemporal information necessary to predict daily surface concentrations. Cross-validation with the in situ data shows that the mixed-effects LUR model using OMI data has a high predictive power (adj. R² = 0.84), especially when compared with surface concentrations derived using the MACC-II reanalysis model dataset (adj. R² = 0.11). Time series

analysis shows no statistically significant trend in NO₂ concentrations during 2005–2015, despite a reported decline in NO_x emissions. This study demonstrates the utility in combining satellite data with LUR models to derive daily maps of ambient surface NO₂ for use in exposure studies.

Long-term change in the source contribution to surface ozone over Japan

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Source: Atmos. Chem. Phys., 17, 8231-8246, 2017
<https://doi.org/10.5194/acp-17-8231-2017>

The relative contributions of various source regions to the long-term (1980–2005) increasing trend in surface ozone (O₃) over Japan were estimated by a series of tracer-tagging simulations using a global chemical transport model. The model simulated the observed increasing trend in surface O₃, including its seasonal variation and geographical features, in Japan well and demonstrated the relative roles of different source regions in forming this trend. Most of the increasing trend in surface O₃ over Japan (~ 97 %) that was simulated was explained as the sum of trends in contributions of different regions to photochemical O₃ production. The increasing trend in O₃ produced in China accounted for 36 % of the total increasing trend and those in the other northeast Asian regions (the Korean Peninsula, coastal regions in East Asia, and Japan) each accounted for about 12–15 %. Furthermore, the contributions of O₃ created in the entire free troposphere and in western, southern, and southeastern Asian regions also increased, and their increasing trends accounted for 16 and 7 % of the total trend, respectively. The impact of interannual variations in climate, in methane concentration, and in emission of O₃ precursors from different source regions on the relative contributions of O₃ created in each region estimated above was also investigated. The variation of climate and the increase in methane concentration together caused the increase of photochemical O₃ production in several regions, and represented about 19 % of the total increasing trend in surface O₃ over Japan. The increase in emission of O₃ precursors in China caused an increase of photochemical O₃ production not only in China itself but also in the other northeast Asian regions and accounted for about 46 % of the total increase in surface O₃ over Japan. Similarly, the relative impact of O₃ precursor emission changes in the Korean Peninsula and Japan were estimated as about 16 and 4 % of the total increasing trend, respectively. The O₃ precursor emission change in regions other than northeast Asia caused increases in surface O₃ over Japan mainly through increasing photochemical O₃ production in western, southern, and southeast Asia and the free troposphere and accounted for about 16 % of the total.

Changes to the chemical state of the Northern Hemisphere atmosphere during the second half of the twentieth century

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Source: Atmos. Chem. Phys., 17, 8269-8283, 2017
<https://doi.org/10.5194/acp-17-8269-2017>

The NO_x (NO and NO₂) and HO_x (OH and HO₂) budgets of the atmosphere exert a major influence on atmospheric composition, controlling removal of primary pollutants and formation of a wide range of secondary products, including ozone, that can influence human health and climate. However, there remain large uncertainties in the changes to these budgets over recent decades. Due to their short atmospheric lifetimes, NO_x and HO_x are highly variable in space and time, and so the measurements of these species are of limited value for examining long-term, large-scale changes to their budgets. Here, we take an alternative approach by examining long-term atmospheric trends of alkyl nitrates, the production efficiency of which is dependent on the atmospheric [NO]/[HO₂] ratio. We derive long-term trends in the alkyl nitrates from measurements in firn air from the NEEM site, Greenland. Their mixing ratios increased by a factor of 3–5 between the 1970s and 1990s. This was followed by a steep decline to the sampling date of 2008. Moreover, we examine how the trends in the alkyl nitrates compare to similarly derived trends in their parent alkanes (i.e. the alkanes which, when oxidised in the presence of NO_x, lead to the formation of the alkyl nitrates). The ratios of the alkyl nitrates to their parent alkanes increased from around 1970 to the late 1990s. This is consistent with large changes to the [NO]/[HO₂] ratio in the Northern Hemisphere atmosphere during this period. Alternatively, they could represent changes to concentrations of the hydroxyl radical, OH, or to the transport time of the air masses from source regions to the Arctic.

Evaluation of ACCMIP ozone simulations and ozonesonde sampling biases using a satellite-based multi-constituent chemical reanalysis

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Source: Atmos. Chem. Phys., 17, 8285-8312, 2017
<https://doi.org/10.5194/acp-17-8285-2017>

The Atmospheric Chemistry Climate Model Intercomparison Project (ACCMIP) ensemble ozone simulations for the present day from the 2000 decade simulation results are evaluated by a state-of-the-art multi-constituent atmospheric chemical reanalysis that ingests multiple satellite data including the Tropospheric Emission Spectrometer (TES), the Microwave Limb Sounder (MLS), the Ozone Monitoring Instrument (OMI), and the Measurement of Pollution in the Troposphere (MOPITT) for 2005–2009. Validation of the chemical reanalysis against global ozonesondes shows good agreement throughout the free troposphere and lower stratosphere for both seasonal and year-to-year variations, with an annual mean bias of less than 0.9 ppb in the middle and upper troposphere at the tropics and mid-latitudes. The reanalysis provides comprehensive spatiotemporal evaluation of chemistry-model performance that complements direct ozonesonde comparisons, which are shown to suffer from significant sampling bias. The reanalysis reveals that the ACCMIP ensemble mean overestimates ozone in the northern extratropics by 6–11 ppb while underestimating by up to 18 ppb in the southern tropics over the Atlantic in the lower troposphere. Most models underestimate the spatial variability of the annual mean lower tropospheric concentrations in the extratropics of both hemispheres by up to 70 %. The ensemble mean also overestimates the seasonal amplitude by 25–70 % in the northern extratropics and overestimates the inter-hemispheric gradient by about 30 % in the lower and middle troposphere. A

part of the discrepancies can be attributed to the 5-year reanalysis data for the decadal model simulations. However, these differences are less evident with the current sonde network. To estimate ozonesonde sampling biases, we computed model bias separately for global coverage and the ozonesonde network. The ozonesonde sampling bias in the evaluated model bias for the seasonal mean concentration relative to global coverage is 40–50 % over the western Pacific and east Indian Ocean and reaches 110 % over the equatorial Americas and up to 80 % for the global tropics. In contrast, the ozonesonde sampling bias is typically smaller than 30 % for the Arctic regions in the lower and middle troposphere. These systematic biases have implications for ozone radiative forcing and the response of chemistry to climate that can be further quantified as the satellite observational record extends to multiple decades.

Carbon dioxide and methane measurements from the Los Angeles Megacity Carbon Project – Part 1: calibration, urban enhancements, and uncertainty estimates

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Source: Atmos. Chem. Phys., 17, 8313–8341, 2017
<https://doi.org/10.5194/acp-17-8313-2017>

We report continuous surface observations of carbon dioxide (CO₂) and methane (CH₄) from the Los Angeles (LA) Megacity Carbon Project during 2015. We devised a calibration strategy, methods for selection of background air masses, calculation of urban enhancements, and a detailed algorithm for estimating uncertainties in urban-scale CO₂ and CH₄ measurements. These methods are essential for understanding carbon fluxes from the LA megacity and other complex urban environments globally. We estimate background mole fractions entering LA using observations from four extra-urban sites including two marine sites located south of LA in La Jolla (LJO) and offshore on San Clemente Island (SCI), one continental site located in Victorville (VIC), in the high desert northeast of LA, and one continental/mid-troposphere site located on Mount Wilson (MWO) in the San Gabriel Mountains. We find that a local marine background can be established to within ~ 1 ppm CO₂ and ~ 10 ppb CH₄ using these local measurement sites. Overall, atmospheric carbon dioxide and methane levels are highly variable across Los Angeles. Urban and suburban sites show moderate to large CO₂ and CH₄ enhancements relative to a marine background estimate. The USC (University of Southern California) site near downtown LA exhibits median hourly enhancements of ~ 20 ppm CO₂ and ~ 150 ppb CH₄ during 2015 as well as ~ 15 ppm CO₂ and ~ 80 ppb CH₄ during mid-afternoon hours (12:00–16:00 LT, local time), which is the typical period of focus for flux inversions. The estimated measurement uncertainty is typically better than 0.1 ppm CO₂ and 1 ppb CH₄ based on the repeated standard gas measurements from the LA sites during the last 2 years, similar to Andrews et al. (2014). The largest component of the measurement uncertainty is due to the single-point calibration method; however, the uncertainty in the background mole fraction is much larger than the measurement uncertainty. The background uncertainty for the marine background estimate is ~ 10 and ~ 15 % of the median mid-afternoon enhancement near downtown LA for CO₂ and CH₄, respectively. Overall, analytical and background uncertainties are small relative to the local CO₂ and CH₄ enhancements;

however, our results suggest that reducing the uncertainty to less than 5 % of the median mid-afternoon enhancement will require detailed assessment of the impact of meteorology on background conditions.

Understanding the primary emissions and secondary formation of gaseous organic acids in the oil sands region of Alberta, Canada

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Source: Atmos. Chem. Phys., 17, 8411-8427, 2017
<https://doi.org/10.5194/acp-17-8411-2017>

Organic acids are known to be emitted from combustion processes and are key photochemical products of biogenic and anthropogenic precursors. Despite their multiple environmental impacts, such as on acid deposition and human–ecosystem health, little is known regarding their emission magnitudes or detailed chemical formation mechanisms. In the current work, airborne measurements of 18 gas-phase low-molecular-weight organic acids were made in the summer of 2013 over the oil sands region of Alberta, Canada, an area of intense unconventional oil extraction. The data from these measurements were used in conjunction with emission retrieval algorithms to derive the total and speciated primary organic acid emission rates, as well as secondary formation rates downwind of oil sands operations. The results of the analysis indicate that approximately 12 t day^{−1} of low-molecular-weight organic acids, dominated by C1–C5 acids, were emitted directly from off-road diesel vehicles within open pit mines. Although there are no specific reporting requirements for primary organic acids, the measured emissions were similar in magnitude to primary oxygenated hydrocarbon emissions, for which there are reporting thresholds, measured previously (≈ 20 t day^{−1}). Conversely, photochemical production of gaseous organic acids significantly exceeded the primary sources, with formation rates of up to ≈ 184 t day^{−1} downwind of the oil sands facilities. The formation and evolution of organic acids from a Lagrangian flight were modelled with a box model, incorporating a detailed hydrocarbon reaction mechanism extracted from the Master Chemical Mechanism (v3.3). Despite evidence of significant secondary organic acid formation, the explicit chemical box model largely underestimated their formation in the oil sands plumes, accounting for 39, 46, 26, and 23 % of the measured formic, acetic, acrylic, and propionic acids respectively and with little contributions from biogenic VOC precursors. The model results, together with an examination of the carbon mass balance between the organic acids formed and the primary VOCs emitted from oil sands operations, suggest the existence of significant missing secondary sources and precursor emissions related to oil sands and/or an incomplete mechanistic and quantitative understanding of how they are processed in the atmosphere.

Global O₃–CO correlations in a chemistry and transport model during July–August: evaluation with TES satellite observations and sensitivity to input meteorological data and emissions

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Source: Atmos. Chem. Phys., 17, 8429–8452, 2017
<https://doi.org/10.5194/acp-17-8429-2017>

We examine the capability of the Global Modeling Initiative (GMI) chemistry and transport model to reproduce global mid-tropospheric (618 hPa) ozone–carbon monoxide (O₃–CO) correlations determined by the measurements from the Tropospheric Emission Spectrometer (TES) aboard NASA's Aura satellite during boreal summer (July–August). The model is driven by three meteorological data sets (finite-volume General Circulation Model (fvGCM) with sea surface temperature for 1995, Goddard Earth Observing System Data Assimilation System Version 4 (GEOS-4 DAS) for 2005, and Modern-Era Retrospective Analysis for Research and Applications (MERRA) for 2005), allowing us to examine the sensitivity of model O₃–CO correlations to input meteorological data. Model simulations of radionuclide tracers (²²²Rn, ²¹⁰Pb, and ⁷Be) are used to illustrate the differences in transport-related processes among the meteorological data sets. Simulated O₃ values are evaluated with climatological profiles from ozonesonde measurements and satellite tropospheric O₃ columns. Despite the fact that the three simulations show significantly different global and regional distributions of O₃ and CO concentrations, they show similar patterns of O₃–CO correlations on a global scale. All model simulations sampled along the TES orbit track capture the observed positive O₃–CO correlations in the Northern Hemisphere midlatitude continental outflow and the Southern Hemisphere subtropics. While all simulations show strong negative correlations over the Tibetan Plateau, northern Africa, the subtropical eastern North Pacific, and the Caribbean, TES O₃ and CO concentrations at 618 hPa only show weak negative correlations over much narrower areas (i.e., the Tibetan Plateau and northern Africa). Discrepancies in regional O₃–CO correlation patterns in the three simulations may be attributed to differences in convective transport, stratospheric influence, and subsidence, among other processes. To understand how various emissions drive global O₃–CO correlation patterns, we examine the sensitivity of GMI/MERRA model-calculated O₃ and CO concentrations and their correlations to emission types (fossil fuel, biomass burning, biogenic, and lightning NO_x emissions). Fossil fuel and biomass burning emissions are mainly responsible for the strong positive O₃–CO correlations over continental outflow regions in both hemispheres. Biogenic emissions have a relatively smaller impact on O₃–CO correlations than other emissions but are largely responsible for the negative correlations over the tropical eastern Pacific, reflecting the fact that O₃ is consumed and CO generated during the atmospheric oxidation process of isoprene under low-NO_x conditions. We find that lightning NO_x emissions degrade both positive correlations at mid- and high latitudes and negative correlations in the tropics because ozone production downwind of lightning NO_x emissions is not directly related to the emission and transport of CO. Our study concludes that O₃–CO correlations may be used effectively to constrain the sources of regional tropospheric O₃ in global 3-D models, especially for those regions where convective transport of pollution plays an important role.

Uncertainties of fluxes and $^{13}\text{C} / ^{12}\text{C}$ ratios of atmospheric reactive-gas emissions

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Source: Atmos. Chem. Phys., 17, 8525-8552, 2017
<https://doi.org/10.5194/acp-17-8525-2017>

We provide a comprehensive review of the proxy data on the $^{13}\text{C}/^{12}\text{C}$ ratios and uncertainties of emissions of reactive carbonaceous compounds into the atmosphere, with a focus on CO sources. Based on an evaluated set-up of the EMAC model, we derive the isotope-resolved data set of its emission inventory for the 1997–2005 period. Additionally, we revisit the calculus required for the correct derivation of uncertainties associated with isotope ratios of emission fluxes. The resulting $\delta^{13}\text{C}$ of overall surface CO emission in 2000 of $-(25.2 \pm 0.7) \text{‰}$ is in line with previous bottom-up estimates and is less uncertain by a factor of 2. In contrast to this, we find that uncertainties of the respective inverse modelling estimates may be substantially larger due to the correlated nature of their derivation. We reckon the $\delta^{13}\text{C}$ values of surface emissions of higher hydrocarbons to be within -24 to -27‰ (uncertainty typically below $\pm 1 \text{‰}$), with an exception of isoprene and methanol emissions being close to -30 and -60‰ , respectively. The isotope signature of ethane surface emission coincides with earlier estimates, but integrates very different source inputs. $\delta^{13}\text{C}$ values are reported relative to V-PDB.

Lidar ratios of stratospheric volcanic ash and sulfate aerosols retrieved from CALIOP measurements

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Source: Atmos. Chem. Phys., 17, 8599-8618, 2017
<https://doi.org/10.5194/acp-17-8599-2017>

We apply a two-way transmittance constraint to nighttime CALIOP (Cloud-Aerosol Lidar with Orthogonal Polarization) observations of volcanic aerosol layers to retrieve estimates of the particulate lidar ratio (S_p) at 532 nm. This technique is applied to three volcanic eruption case studies that were found to have injected aerosols directly into the stratosphere. Numerous lidar observations permitted characterization of the optical and geometric properties of the volcanic aerosol layers over a time period of 1–2 weeks. For the volcanic ash-rich layers produced by the Puyehue-Cordón Caulle eruption (June 2011), we obtain mean and median particulate lidar ratios of $69 \pm 13 \text{ sr}$ and 67 sr , respectively. For the sulfate-rich aerosol layers produced by Kasatochi (August 2008) and Sarychev Peak (June 2009), the means of the retrieved lidar ratios were $66 \pm 19 \text{ sr}$ (median 60 sr) and $63 \pm 14 \text{ sr}$ (median 59 sr), respectively. The 532 nm layer-integrated particulate depolarization ratios (δ_p) observed for the Puyehue layers ($\delta_p = 0.33 \pm 0.03$) were much larger than those found for the volcanic aerosol layers produced by the Kasatochi ($\delta_p = 0.09 \pm 0.03$) and Sarychev ($\delta_p = 0.05 \pm 0.04$) eruptions. However, for the Sarychev layers we observe an exponential decay (e-folding time of 3.6 days) in δ_p with time from 0.27 to 0.03. Similar decreases in the layer-integrated attenuated colour ratios with time were observed for the Sarychev case. In general, the Puyehue layers exhibited larger colour ratios ($\chi' = 0.53 \pm 0.07$) than what was observed for the Kasatochi ($\chi' = 0.35 \pm 0.07$) and Sarychev ($\chi' = 0.32 \pm 0.07$) layers, indicating that the Puyehue layers were generally

composed of larger particles. These observations are particularly relevant to the new stratospheric aerosol subtyping classification scheme, which has been incorporated into version 4 of the level 2 CALIPSO (Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation) data products.

Arctic regional methane fluxes by ecotope as derived using eddy covariance from a low-flying aircraft

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Source: Atmos. Chem. Phys., 17, 8619-8633, 2017
<https://doi.org/10.5194/acp-17-8619-2017>

The Arctic terrestrial and sub-sea permafrost region contains approximately 30 % of the global carbon stock, and therefore understanding Arctic methane emissions and how they might change with a changing climate is important for quantifying the global methane budget and understanding its growth in the atmosphere. Here we present measurements from a new in situ flux observation system designed for use on a small, low-flying aircraft that was deployed over the North Slope of Alaska during August 2013. The system combines a small methane instrument based on integrated cavity output spectroscopy (ICOS) with an air turbulence probe to calculate methane fluxes based on eddy covariance. We group surface fluxes by land class using a map based on LandSat Thematic Mapper (TM) data with 30 m resolution. We find that wet sedge areas dominate the methane fluxes with a mean flux of $2.1 \mu\text{g m}^{-2} \text{s}^{-1}$ during the first part of August. Methane emissions from the Sagavanirktok River have the second highest at almost $1 \mu\text{g m}^{-2} \text{s}^{-1}$. During the second half of August, after soil temperatures had cooled by 7°C , methane emissions fell to between 0 and $0.5 \mu\text{g m}^{-2} \text{s}^{-1}$ for all areas measured. We compare the aircraft measurements with an eddy covariance flux tower located in a wet sedge area and show that the two measurements agree quantitatively when the footprints of both overlap. However, fluxes from sedge vary at times by a factor of 2 or more even within a few kilometers of the tower demonstrating the importance of making regional measurements to map out methane emissions spatial heterogeneity. Aircraft measurements of surface flux can play an important role in bridging the gap between ground-based measurements and regional measurements from remote sensing instruments and models.

Global anthropogenic emissions of particulate matter including black carbon

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Source: Atmos. Chem. Phys., 17, 8681-8723, 2017
<https://doi.org/10.5194/acp-17-8681-2017>

This paper presents a comprehensive assessment of historical (1990–2010) global anthropogenic particulate matter (PM) emissions including the consistent and harmonized calculation of mass-based size distribution (PM₁, PM_{2.5}, PM₁₀), as well as primary carbonaceous aerosols including black carbon (BC) and organic carbon (OC). The estimates were developed with the integrated assessment model GAINS, where source- and region-specific technology characteristics are explicitly included. This assessment includes a number of previously unaccounted or often misallocated emission sources, i.e. kerosene lamps, gas flaring, diesel generators, refuse burning; some of them were reported in the past for selected regions or in the context of a particular pollutant or sector but not included as part of a total estimate. Spatially, emissions were calculated for 172 source regions (as well as international shipping), presented for 25 global regions, and allocated to 0.5° × 0.5° longitude–latitude grids. No independent estimates of emissions from forest fires and savannah burning are provided and neither windblown dust nor unpaved roads emissions are included.

We estimate that global emissions of PM have not changed significantly between 1990 and 2010, showing a strong decoupling from the global increase in energy consumption and, consequently, CO₂ emissions, but there are significantly different regional trends, with a particularly strong increase in East Asia and Africa and a strong decline in Europe, North America, and the Pacific region. This in turn resulted in important changes in the spatial pattern of PM burden, e.g. European, North American, and Pacific contributions to global emissions dropped from nearly 30 % in 1990 to well below 15 % in 2010, while Asia's contribution grew from just over 50 % to nearly two-thirds of the global total in 2010. For all PM species considered, Asian sources represented over 60 % of the global anthropogenic total, and residential combustion was the most important sector, contributing about 60 % for BC and OC, 45 % for PM_{2.5}, and less than 40 % for PM₁₀, where large combustion sources and industrial processes are equally important. Global anthropogenic emissions of BC were estimated at about 6.6 and 7.2 Tg in 2000 and 2010, respectively, and represent about 15 % of PM_{2.5} but for some sources reach nearly 50 %, i.e. for the transport sector. Our global BC numbers are higher than previously published owing primarily to the inclusion of new sources.

This PM estimate fills the gap in emission data and emission source characterization required in air quality and climate modelling studies and health impact assessments at a regional and global level, as it includes both carbonaceous and non-carbonaceous constituents of primary particulate matter emissions. The developed emission dataset has been used in several regional and global atmospheric transport and climate model simulations within the ECLIPSE (Evaluating the Climate and Air Quality Impacts of Short-Lived Pollutants) project and beyond, serves better parameterization of the global integrated assessment models with respect to representation of black carbon and organic carbon emissions, and built a basis for recently published global particulate number estimates.

Comparison of primary and secondary particle formation from natural gas engine exhaust and of their volatility characteristics

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Source: Atmos. Chem. Phys., 17, 8739-8755, 2017
<https://doi.org/10.5194/acp-17-8739-2017>

Natural gas usage in the traffic and energy production sectors is a growing trend worldwide; thus, an assessment of its effects on air quality, human health and climate is required. Engine exhaust is a source of primary particulate emissions and secondary aerosol precursors, which both contribute to air quality and can cause adverse health effects. Technologies, such as cleaner engines or fuels, that produce less primary and secondary aerosols could potentially significantly decrease atmospheric particle concentrations and their adverse effects. In this study, we used a potential aerosol mass (PAM) chamber to investigate the secondary aerosol formation potential of natural gas engine exhaust. The PAM chamber was used with a constant UV-light voltage, which resulted in relatively long equivalent atmospheric ages of 11 days at most. The studied retro-fitted natural gas engine exhaust was observed to form secondary aerosol. The mass of the total aged particles, i.e., particle mass measured downstream of the PAM chamber, was 6–268 times as high as the mass of the emitted primary exhaust particles. The secondary organic aerosol (SOA) formation potential was measured to be 9–20 mg kg_{fuel}⁻¹. The total aged particles mainly consisted of organic matter, nitrate, sulfate and ammonium, with the fractions depending on exhaust after-treatment and the engine parameters used. Also, the volatility, composition and concentration of the total aged particles were found to depend on the engine operating mode, catalyst temperature and catalyst type. For example, a high catalyst temperature promoted the formation of sulfate particles, whereas a low catalyst temperature promoted nitrate formation. However, in particular, the concentration of nitrate needed a long time to stabilize – more than half an hour – which complicated the conclusions but also indicates the sensitivity of nitrate measurements on experimental parameters such as emission source and system temperatures. Sulfate was measured to have the highest evaporation temperature, and nitrate had the lowest. The evaporation temperature of ammonium depended on the fractions of nitrate and sulfate in the particles. The average volatility of the total aged particles was measured to be lower than that of primary particles, indicating better stability of the aged natural gas engine-emitted aerosol in the atmosphere. According to the results of this study, the exhaust of a natural gas engine equipped with a catalyst forms secondary aerosol when the atmospheric ages in a PAM chamber are several days long. The secondary aerosol matter has different physical characteristics from those of primary particulate emissions.

Response of the global surface ozone distribution to Northern Hemisphere sea surface temperature changes: implications for long-range transport

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Source: Atmos. Chem. Phys., 17, 8771-8788, 2017
<https://doi.org/10.5194/acp-17-8771-2017>

The response of surface ozone (O₃) concentrations to basin-scale warming and cooling of Northern Hemisphere oceans is investigated using the Community Earth System Model (CESM). Idealized, spatially uniform sea surface temperature (SST) anomalies of ± 1 °C are individually superimposed onto the North Pacific, North Atlantic, and North Indian oceans. Our simulations suggest large seasonal and regional

variability in surface O₃ in response to SST anomalies, especially in the boreal summer. The responses of surface O₃ associated with basin-scale SST warming and cooling have similar magnitude but are opposite in sign. Increasing the SST by 1 °C in one of the oceans generally decreases the surface O₃ concentrations from 1 to 5 ppbv. With fixed emissions, SST increases in a specific ocean basin in the Northern Hemisphere tend to increase the summertime surface O₃ concentrations over upwind regions, accompanied by a widespread reduction over downwind continents. We implement the integrated process rate (IPR) analysis in CESM and find that meteorological O₃ transport in response to SST changes is the key process causing surface O₃ perturbations in most cases. During the boreal summer, basin-scale SST warming facilitates the vertical transport of O₃ to the surface over upwind regions while significantly reducing the vertical transport over downwind continents. This process, as confirmed by tagged CO-like tracers, indicates a considerable suppression of intercontinental O₃ transport due to increased tropospheric stability at lower midlatitudes induced by SST changes. Conversely, the responses of chemical O₃ production to regional SST warming can exert positive effects on surface O₃ levels over highly polluted continents, except South Asia, where intensified cloud loading in response to North Indian SST warming depresses both the surface air temperature and solar radiation, and thus photochemical O₃ production. Our findings indicate a robust linkage between basin-scale SST variability and continental surface O₃ pollution, which should be considered in regional air quality management.

Atmospheric chemistry, sources and sinks of carbon suboxide, C₃O₂

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Source: *Atmos. Chem. Phys.*, 17, 8789-8804, 2017
<https://doi.org/10.5194/acp-17-8789-2017>

Carbon suboxide, O = C = C = C = O, has been detected in ambient air samples and has the potential to be a noxious pollutant and oxidant precursor; however, its lifetime and fate in the atmosphere are largely unknown. In this work, we collect an extensive set of studies on the atmospheric chemistry of C₃O₂. Rate coefficients for the reactions of C₃O₂ with OH radicals and ozone were determined as $k_{OH} = (2.6 \pm 0.5) \times 10^{-12} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$ at 295 K (independent of pressure between ~ 25 and 1000 mbar) and $k_{O_3} < 1.5 \times 10^{-21} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$ at 295 K. A theoretical study on the mechanisms of these reactions indicates that the sole products are CO and CO₂, as observed experimentally. The UV absorption spectrum and the interaction of C₃O₂ with water (Henry's law solubility and hydrolysis rate constant) were also investigated, enabling its photodissociation lifetime and hydrolysis rates, respectively, to be assessed.

The role of C₃O₂ in the atmosphere was examined using in situ measurements, an analysis of the atmospheric sources and sinks and simulation with the EMAC atmospheric chemistry–general circulation model. The results indicate sub-pptv levels at the Earth's surface, up to about 10 pptv in regions with relatively strong sources, e.g. influenced by biomass burning, and a mean lifetime of ~ 3.2 days. These predictions carry considerable uncertainty, as more measurement data are needed to determine ambient concentrations and constrain the source strengths.

Observations of particles at their formation sizes in Beijing, China

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Source: Atmos. Chem. Phys., 17, 8825-8835, 2017
<https://doi.org/10.5194/acp-17-8825-2017>

New particle formation (NPF) has been observed in many highly polluted environments of South East Asia, including Beijing, where the extent of its contribution to intense haze events is still an open question. Estimated characteristics of NPF events, such as their starting times and formation and growth rates of particles, are more accurate when the detection range of particles extends to smaller sizes. In order to understand the very first steps of particle formation, we used a neutral cluster and air ion spectrometer (NAIS) to investigate particle characteristics at sizes exactly at which atmospheric nucleation and cluster activity occurs. Observations over a continuous 3-month period in Beijing showed 26 NPF events. These events generally coincided with periods with relatively clean air when the wind direction was from the less industrialised north. No NPF events were observed when the daily mean PM_{2.5} concentration exceeded 43 $\mu\text{g m}^{-3}$, which was the upper threshold for particle formation in Beijing. The fraction of particles that are charged in the size range 2–42 nm was normally about 15 %. However, this fraction increased to 20–30 % during haze events and decreased to below 10 % during NPF events. With the NAIS, we very precisely determined the starting times of NPF to a greater accuracy than has been possible in Beijing before and provided a temporal distribution of NPF events with a maximum at about 08:30 LT. Particle formation rates varied between 12 and 38 $\text{cm}^{-3} \text{s}^{-1}$. Particle growth rates were estimated to be in the range of 0.5–9.0 nm h^{-1} . These results are more reliable than previous studies in Beijing as the measurements were conducted for the first time at the exact sizes at which clusters form into particles and provide useful insight into the formation of haze events.

Organic molecular tracers in the atmospheric aerosols from Lumbini, Nepal, in the northern Indo-Gangetic Plain: influence of biomass burning

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Source: Atmos. Chem. Phys., 17, 8867-8885, 2017
<https://doi.org/10.5194/acp-17-8867-2017>

To better understand the characteristics of biomass burning in the northern Indo-Gangetic Plain (IGP), total suspended particles were collected in a rural site, Lumbini, Nepal, during April 2013 to March 2014 and analyzed for the biomass burning tracers (i.e., levoglucosan, mannosan, vanillic acid). The annual average concentration of levoglucosan was $734 \pm 1043 \text{ ng m}^{-3}$ with the maximum seasonal mean concentration during post-monsoon season ($2206 \pm 1753 \text{ ng m}^{-3}$), followed by winter ($1161 \pm 1347 \text{ ng m}^{-3}$), pre-monsoon ($771 \pm 524 \text{ ng m}^{-3}$) and minimum concentration during monsoon

season ($212 \pm 279 \text{ ng m}^{-3}$). The other biomass burning tracers (mannosan, galactosan, p-hydroxybenzoic acid, vanillic acid, syringic acid and dehydroabietic acid) also showed the similar seasonal variations. There were good correlations among levoglucosan, organic carbon (OC) and elemental carbon (EC), indicating significant impact of biomass burning activities on carbonaceous aerosol loading throughout the year in Lumbini area. According to the characteristic ratios, levoglucosan/mannosan (lev/man) and syringic acid/vanillic acid (syr/van), we deduced that the high abundances of biomass burning products during non-monsoon seasons were mainly caused by the burning of crop residues and hardwood while the softwood had less contribution. Based on the diagnostic tracer ratio (i.e., lev/OC), the OC derived from biomass burning constituted large fraction of total OC, especially during post-monsoon season. By analyzing the MODIS fire spot product and 5-day air-mass back trajectories, we further demonstrated that organic aerosol composition was not only related to the local agricultural activities and residential biomass usage but also impacted by the regional emissions. During the post-monsoon season, the emissions from rice residue burning in western India and eastern Pakistan could impact particulate air pollution in Lumbini and surrounding regions in southern Nepal. Therefore, our finding is meaningful and has a great importance for adopting the appropriate mitigation measures, not only at the local level but also by involving different regions and nations, to reduce the biomass burning emissions in the broader IGP region nations.

Modeling the role of highly oxidized multifunctional organic molecules for the growth of new particles over the boreal forest region

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Source: Atmos. Chem. Phys., 17, 8887-8901, 2017
<https://doi.org/10.5194/acp-17-8887-2017>

In this study, the processes behind observed new particle formation (NPF) events and subsequent organic-dominated particle growth at the Pallas Atmosphere–Ecosystem Supersite in Northern Finland are explored with the one-dimensional column trajectory model ADCHEM. The modeled sub-micron particle mass is up to $\sim 75\%$ composed of SOA formed from highly oxidized multifunctional organic molecules (HOMs) with low or extremely low volatility. In the model the newly formed particles with an initial diameter of 1.5 nm reach a diameter of 7 nm about 2 h earlier than what is typically observed at the station. This is an indication that the model tends to overestimate the initial particle growth. In contrast, the modeled particle growth to CCN size ranges ($> 50 \text{ nm}$ in diameter) seems to be underestimated because the increase in the concentration of particles above 50 nm in diameter typically occurs several hours later compared to the observations. Due to the high fraction of HOMs in the modeled particles, the oxygen-to-carbon (O : C) atomic ratio of the SOA is nearly 1. This unusually high O : C and the discrepancy between the modeled and observed particle growth might be explained by the fact that the model does not consider any particle-phase reactions involving semi-volatile organic compounds with relatively low O : C. In the model simulations where condensation of low-volatility and extremely low-volatility HOMs explain most of the SOA formation, the phase state of the SOA (assumed either liquid or amorphous solid) has an insignificant impact on the evolution of the particle number size distributions. However, the modeled particle growth rates are sensitive to the method used to estimate

the vapor pressures of the HOMs. Future studies should evaluate how heterogeneous reactions involving semi-volatility HOMs and other less-oxidized organic compounds can influence the SOA composition- and size-dependent particle growth.

Global source attribution of sulfate concentration and direct and indirect radiative forcing

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Source: Atmos. Chem. Phys., 17, 8903-8922, 2017
<https://doi.org/10.5194/acp-17-8903-2017>

The global source–receptor relationships of sulfate concentrations, and direct and indirect radiative forcing (DRF and IRF) from 16 regions/sectors for years 2010–2014 are examined in this study through utilizing a sulfur source-tagging capability implemented in the Community Earth System Model (CESM) with winds nudged to reanalysis data. Sulfate concentrations are mostly contributed by local emissions in regions with high emissions, while over regions with relatively low SO₂ emissions, the near-surface sulfate concentrations are primarily attributed to non-local sources from long-range transport. Regional source efficiencies of sulfate concentrations are higher over regions with dry atmospheric conditions and less export, suggesting that lifetime of aerosols, together with regional export, is important in determining regional air quality. The simulated global total sulfate DRF is -0.42 W m^{-2} , with -0.31 W m^{-2} contributed by anthropogenic sulfate and -0.11 W m^{-2} contributed by natural sulfate, relative to a state with no sulfur emissions. In the Southern Hemisphere tropics, dimethyl sulfide (DMS) contributes 17–84 % to the total DRF. East Asia has the largest contribution of 20–30 % over the Northern Hemisphere mid- and high latitudes. A 20 % perturbation of sulfate and its precursor emissions gives a sulfate incremental IRF of -0.44 W m^{-2} . DMS has the largest contribution, explaining -0.23 W m^{-2} of the global sulfate incremental IRF. Incremental IRF over regions in the Southern Hemisphere with low background aerosols is more sensitive to emission perturbation than that over the polluted Northern Hemisphere.

Evaporation of sulfate aerosols at low relative humidity

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Source: Atmos. Chem. Phys., 17, 8923-8938, 2017
<https://doi.org/10.5194/acp-17-8923-2017>

Evaporation of sulfuric acid from particles can be important in the atmospheres of Earth and Venus. However, the equilibrium constant for the dissociation of H₂SO₄ to bisulfate ions, which is the one of

the fundamental parameters controlling the evaporation of sulfur particles, is not well constrained. In this study we explore the volatility of sulfate particles at very low relative humidity. We measured the evaporation of sulfur particles versus temperature and relative humidity in the CLOUD chamber at CERN. We modelled the observed sulfur particle shrinkage with the ADCHAM model. Based on our model results, we conclude that the sulfur particle shrinkage is mainly governed by H₂SO₄ and potentially to some extent by SO₃ evaporation. We found that the equilibrium constants for the dissociation of H₂SO₄ to HSO₄–(KH₂SO₄) and the dehydration of H₂SO₄ to SO₃ (xKSO₃) are $KH_2SO_4 = 2\text{--}4 \times 10^9 \text{ mol kg}^{-1}$ and $xKSO_3 \geq 1.4 \times 10^{10}$ at $288.8 \pm 5 \text{ K}$.

Speciation of organic aerosols in the Saharan Air Layer and in the free troposphere westerlies

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Source: Atmos. Chem. Phys., 17, 8939–8958, 2017
<https://doi.org/10.5194/acp-17-8939-2017>

We focused this research on the composition of the organic aerosols transported in the two main airflows of the subtropical North Atlantic free troposphere: (i) the Saharan Air Layer – the warm, dry and dusty airstream that expands from North Africa to the Americas at subtropical and tropical latitudes – and (ii) the westerlies, which flow from North America over the North Atlantic at mid- and subtropical latitudes. We determined the inorganic compounds (secondary inorganic species and elemental composition), elemental carbon and the organic fraction (bulk organic carbon and organic speciation) present in the aerosol collected at Izaña Observatory, $\sim 2400 \text{ m a.s.l.}$ on the island of Tenerife. The concentrations of all inorganic and almost all organic compounds were higher in the Saharan Air Layer than in the westerlies, with bulk organic matter concentrations within the range $0.02\text{--}4.0 \mu\text{g m}^{-3}$. In the Saharan Air Layer, the total aerosol population was by far dominated by dust (93 % of bulk mass), which was mixed with secondary inorganic pollutants ($< 5 \%$) and organic matter ($\sim 1.5 \%$). The chemical speciation of the organic aerosols (levoglucosan, dicarboxylic acids, saccharides, n-alkanes, hopanes, polycyclic aromatic hydrocarbons and those formed after oxidation of α -pinene and isoprene, determined by gas chromatography coupled with mass spectrometry) accounted for 15 % of the bulk organic matter (determined by the thermo-optical transmission technique); the most abundant organic compounds were saccharides (associated with surface soils), secondary organic aerosols linked to oxidation of biogenic isoprene (SOA ISO) and dicarboxylic acids (linked to several primary sources and SOA). When the Saharan Air Layer shifted southward, Izaña was within the westerlies stream and organic matter accounted for $\sim 28 \%$ of the bulk mass of aerosols. In the westerlies, the organic aerosol species determined accounted for 64 % of the bulk organic matter, with SOA ISO and dicarboxylic acids being the most abundant; the highest concentration of organic matter ($3.6 \mu\text{g m}^{-3}$) and of some organic species (e.g. levoglucosan and some dicarboxylic acids) were associated with biomass burning linked to a fire in North America. In the Saharan Air Layer, the correlation found between SOA ISO and nitrate suggests a large-scale impact of enhancement of the formation rate of secondary organic aerosols due to interaction with anthropogenic NO_x emissions.

Investigating diesel engines as an atmospheric source of isocyanic acid in urban areas

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Source: Atmos. Chem. Phys., 17, 8959-8970, 2017
<https://doi.org/10.5194/acp-17-8959-2017>

Isocyanic acid (HNCO), an acidic gas found in tobacco smoke, urban environments, and biomass-burning-affected regions, has been linked to adverse health outcomes. Gasoline- and diesel-powered engines and biomass burning are known to emit HNCO and hypothesized to emit precursors such as amides that can photochemically react to produce HNCO in the atmosphere. Increasingly, diesel engines in developed countries like the United States are required to use selective catalytic reduction (SCR) systems to reduce tailpipe emissions of oxides of nitrogen. SCR chemistry is known to produce HNCO as an intermediate product, and SCR systems have been implicated as an atmospheric source of HNCO. In this work, we measure HNCO emissions from an SCR system-equipped diesel engine and, in combination with earlier data, use a three-dimensional chemical transport model (CTM) to simulate the ambient concentrations and source/pathway contributions to HNCO in an urban environment. Engine tests were conducted at three different engine loads, using two different fuels and at multiple operating points. HNCO was measured using an acetate chemical ionization mass spectrometer. The diesel engine was found to emit primary HNCO (3–90 mg kg fuel⁻¹) but we did not find any evidence that the SCR system or other aftertreatment devices (i.e., oxidation catalyst and particle filter) produced or enhanced HNCO emissions. The CTM predictions compared well with the only available observational datasets for HNCO in urban areas but underpredicted the contribution from secondary processes. The comparison implied that diesel-powered engines were the largest source of HNCO in urban areas. The CTM also predicted that daily-averaged concentrations of HNCO reached a maximum of ~110 pptv but were an order of magnitude lower than the 1 ppbv level that could be associated with physiological effects in humans. Precursor contributions from other combustion sources (gasoline and biomass burning) and wintertime conditions could enhance HNCO concentrations but need to be explored in future work.

Power plant fuel switching and air quality in a tropical, forested environment

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Source: Atmos. Chem. Phys., 17, 8987-8998, 2017
<https://doi.org/10.5194/acp-17-8987-2017>

How a changing energy matrix for electricity production affects air quality is considered for an urban region in a tropical, forested environment. Manaus, the largest city in the central Amazon Basin of Brazil, is in the process of changing its energy matrix for electricity production from fuel oil and diesel to

natural gas over an approximately 10-year period, with a minor contribution by hydropower. Three scenarios of urban air quality, specifically afternoon ozone concentrations, were simulated using the Weather Research and Forecasting (WRF-Chem) model. The first scenario used fuel oil and diesel for electricity production, which was the reality in 2008. The second scenario was based on the fuel mix from 2014, the most current year for which data were available. The third scenario considered nearly complete use of natural gas for electricity production, which is the anticipated future, possibly for 2018. For each case, inventories of anthropogenic emissions were based on electricity generation, refinery operations, and transportation. Transportation and refinery operations were held constant across the three scenarios to focus on effects of power plant fuel switching in a tropical context. The simulated NO_x and CO emissions for the urban region decrease by 89 and 55 %, respectively, after the complete change in the energy matrix. The results of the simulations indicate that a change to natural gas significantly decreases maximum afternoon ozone concentrations over the population center, reducing ozone by > 70 % for the most polluted days. The sensitivity of ozone concentrations to the fuel switchover is consistent with a NO_x-limited regime, as expected for a tropical forest having high emissions of biogenic volatile organic compounds, high water vapor concentrations, and abundant solar radiation. There are key differences in a shifting energy matrix in a tropical, forested environment compared to other world environments. Policies favoring the burning of natural gas in place of fuel oil and diesel have great potential for ozone reduction and improved air quality for growing urban regions located in tropical, forested environments around the world.

Subtropical subsidence and surface deposition of oxidized mercury produced in the free troposphere

Viral Shah and Lyatt Jaeglé

Source: Atmos. Chem. Phys., 17, 8999-9017, 2017
<https://doi.org/10.5194/acp-17-8999-2017>

Oxidized mercury (Hg(II)) is chemically produced in the atmosphere by oxidation of elemental mercury and is directly emitted by anthropogenic activities. We use the GEOS-Chem global chemical transport model with gaseous oxidation driven by Br atoms to quantify how surface deposition of Hg(II) is influenced by Hg(II) production at different atmospheric heights. We tag Hg(II) chemically produced in the lower (surface–750 hPa), middle (750–400 hPa), and upper troposphere (400 hPa–tropopause), in the stratosphere, as well as directly emitted Hg(II). We evaluate our 2-year simulation (2013–2014) against observations of Hg(II) wet deposition as well as surface and free-tropospheric observations of Hg(II), finding reasonable agreement. We find that Hg(II) produced in the upper and middle troposphere constitutes 91 % of the tropospheric mass of Hg(II) and 91 % of the annual Hg(II) wet deposition flux. This large global influence from the upper and middle troposphere is the result of strong chemical production coupled with a long lifetime of Hg(II) in these regions. Annually, 77–84 % of surface-level Hg(II) over the western US, South America, South Africa, and Australia is produced in the upper and middle troposphere, whereas 26–66 % of surface Hg(II) over the eastern US, Europe, and East Asia, and South Asia is directly emitted. The influence of directly emitted Hg(II) near emission sources is likely higher but cannot be quantified by our coarse-resolution global model (2° latitude × 2.5° longitude). Over the oceans, 72 % of surface Hg(II) is produced in the lower troposphere because of higher Br concentrations in the marine boundary layer. The global contribution of the upper and middle

troposphere to the Hg(II) dry deposition flux is 52 %. It is lower compared to the contribution to wet deposition because dry deposition of Hg(II) produced aloft requires its entrainment into the boundary layer, while rain can scavenge Hg(II) from higher altitudes more readily. We find that 55 % of the spatial variation of Hg wet deposition flux observed at the Mercury Deposition Network sites is explained by the combined variation of precipitation and Hg(II) produced in the upper and middle troposphere. Our simulation points to a large role of the dry subtropical subsidence regions. Hg(II) present in these regions accounts for 74 % of Hg(II) at 500 hPa over the continental US and more than 60 % of the surface Hg(II) over high-altitude areas of the western US. Globally, it accounts for 78 % of the tropospheric Hg(II) mass and 61 % of the total Hg(II) deposition. During the Nitrogen, Oxidants, Mercury, and Aerosol Distributions, Sources, and Sinks (NOMADSS) aircraft campaign, the contribution of Hg(II) from the dry subtropical regions was found to be 75 % when measured Hg(II) exceeded 250 pg m⁻³. Hg(II) produced in the upper and middle troposphere subsides in the anticyclones, where the dry conditions inhibit the loss of Hg(II). Our results highlight the importance the subtropical anticyclones as the primary conduits for the production and export of Hg(II) to the global atmosphere

A wedge strategy for mitigation of urban warming in future climate scenarios

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Source: Atmos. Chem. Phys., 17, 9067-9080, 2017
<https://doi.org/10.5194/acp-17-9067-2017>

Heat stress is one of the most severe climate threats to human society in a future warmer world. The situation is further exacerbated in urban areas by urban heat islands (UHIs). Because the majority of world's population is projected to live in cities, there is a pressing need to find effective solutions for the heat stress problem. We use a climate model to investigate the effectiveness of various urban heat mitigation strategies: cool roofs, street vegetation, green roofs, and reflective pavement. Our results show that by adopting highly reflective roofs, almost all the cities in the United States and southern Canada are transformed into white oases – cold islands caused by cool roofs at midday, with an average oasis effect of –3.4 K in the summer for the period 2071–2100, which offsets approximately 80 % of the greenhouse gas (GHG) warming projected for the same period under the RCP4.5 scenario. A UHI mitigation wedge consisting of cool roofs, street vegetation, and reflective pavement has the potential to eliminate the daytime UHI plus the GHG warming.

Global atmospheric chemistry – which air matters

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Source: Atmos. Chem. Phys., 17, 9081-9102, 2017
<https://doi.org/10.5194/acp-17-9081-2017>

An approach for analysis and modeling of global atmospheric chemistry is developed for application to measurements that provide a tropospheric climatology of those heterogeneously distributed, reactive species that control the loss of methane and the production and loss of ozone. We identify key species (e.g., O₃, NO_x, HNO₃, HNO₄, C₂H₃NO₅, H₂O, HOOH, CH₃OOH, HCHO, CO, CH₄, C₂H₆, acetaldehyde, acetone) and presume that they can be measured simultaneously in air parcels on the scale of a few km horizontally and a few tenths of a km vertically. As a first step, six global models have prepared such climatologies sampled at the modeled resolution for August with emphasis on the vast central Pacific Ocean basin. Objectives of this paper are to identify and characterize differences in model-generated reactivities as well as species covariances that could readily be discriminated with an unbiased climatology. A primary tool is comparison of multidimensional probability densities of key species weighted by the mass of such parcels or frequency of occurrence as well as by the reactivity of the parcels with respect to methane and ozone. The reactivity-weighted probabilities tell us which parcels matter in this case, and this method shows skill in differentiating among the models' chemistry. Testing 100 km scale models with 2 km measurements using these tools also addresses a core question about model resolution and whether fine-scale atmospheric structures matter to the overall ozone and methane budget. A new method enabling these six global chemistry–climate models to ingest an externally sourced climatology and then compute air parcel reactivity is demonstrated. Such an objective climatology containing these key species is anticipated from the NASA Atmospheric Tomography (ATom) aircraft mission (2015–2020), executing profiles over the Pacific and Atlantic Ocean basins. This modeling study addresses a core part of the design of ATom.

Differentiating local and regional sources of Chinese urban air pollution based on the effect of the Spring Festival

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Source: Atmos. Chem. Phys., 17, 9103-9114, 2017
<https://doi.org/10.5194/acp-17-9103-2017>

The emission of pollutants is extremely reduced during the annual Chinese Spring Festival (SF) in Shenzhen, China. During the SF, traffic flow drops by ~50 % and the industrial plants are almost entirely shut down in Shenzhen. To characterize the variation in ambient air pollutants due to the Spring Festival effect, various gaseous and particulate pollutants were measured in real time in urban Shenzhen over three consecutive winters (2014–2016). The results indicate that the concentrations of NO_x, volatile organic compounds (VOCs), black carbon (BC), primary organic aerosols, chloride, and nitrate in submicron aerosols decrease by 50–80 % during SF periods relative to non-Spring Festival periods, regardless of meteorological conditions. This decrease suggests that these pollutants are mostly emitted or secondarily formed from urban local emissions. The concentration variation in species mostly from regional or natural sources, however, is found to be much less, such as for bulk fine particulate matter (PM_{2.5}). More detailed analysis of the Spring Festival effect reveals an urgent need to reduce emissions of SO₂ and VOCs on a regional scale rather than on an urban scale to reduce urban PM_{2.5} in Shenzhen, which can also be useful as a reference for other megacities in China.

OMI-measured increasing SO₂ emissions due to energy industry expansion and relocation in northwestern China

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Source: *Atmos. Chem. Phys.*, 17, 9115-9131, 2017

<https://doi.org/10.5194/acp-17-9115-2017>

The rapid growth of economy makes China the largest energy consumer and sulfur dioxide (SO₂) emitter in the world. In this study, we estimated the trends and step changes in the planetary boundary layer (PBL) vertical column density (VCD) of SO₂ from 2005 to 2015 over China measured by the Ozone Monitoring Instrument (OMI). We show that these trends and step change years coincide with the effective date and period of the national strategy for energy development and relocation in northwestern China and the regulations in the reduction of SO₂ emissions. Under the national regulations for the reduction of SO₂ emissions in eastern and southern China, SO₂ VCD in the Pearl River Delta (PRD) of southern China exhibited the largest decline during 2005–2015 at a rate of $-7\% \text{ yr}^{-1}$, followed by the North China Plain (NCP) ($-6.7\% \text{ yr}^{-1}$), Sichuan Basin ($-6.3\% \text{ yr}^{-1}$), and Yangtze River Delta (YRD) ($-6\% \text{ yr}^{-1}$). The Mann–Kendall (MK) test reveals the step change points of declining SO₂ VCD in 2009 for the PRD and 2012–2013 for eastern China responding to the implementation of SO₂ control regulation in these regions. In contrast, the MK test and regression analysis also revealed increasing trends of SO₂ VCD in northwestern China, particularly for several hot spots featured by growing SO₂ VCD in those large-scale energy industry bases in northwestern China. The enhanced SO₂ VCD is potentially attributable to increasing SO₂ emissions due to the development of large-scale energy industry bases in energy-abundant northwestern China under the national strategy for the energy safety of China in the 21st century. We show that these large-scale energy industry bases could overwhelm the trends and changes in provincial total SO₂ emissions in northwestern China and contribute increasingly to the national total SO₂ emissions in China. Given that northwestern China is more ecologically fragile and uniquely susceptible to atmospheric pollution than the rest of China, increasing SO₂ emissions in this part of China should not be overlooked and merit scientific research.

Observed trends in ground-level O₃ in Monterrey, Mexico, during 1993–2014: comparison with Mexico City and Guadalajara

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Source: *Atmos. Chem. Phys.*, 17, 9163-9185, 2017

<https://doi.org/10.5194/acp-17-9163-2017>

Here, we present an assessment of long-term trends in O₃ and odd oxygen (O₃ + NO₂) at the industrial Monterrey metropolitan area (MMA) in NE Mexico. Diurnal amplitudes in Ox (AV_d) are used as a proxy for net O₃ production, which is influenced by the NO₂ photolysis rate. No significant differences in the AV_d are observed between weekends and weekdays, although the largest AV_d values are observed at sites downwind of industrial areas. The highest O₃ mixing ratios are observed in spring, with minimum

values in winter. The largest annual variations in O₃ are typically observed downwind of the MMA, with the lowest variations generally recorded in highly populated areas and close to industrial areas. A wind sector analysis of mixing ratios of O₃ precursors revealed that the dominant sources of emissions are located in the industrial regions within the MMA and surrounding area. Significant increasing trends in O₃ in spring, summer, and autumn are observed depending on site location, with trends in annual averages ranging between 0.19 and 0.33 ppb yr⁻¹. Overall, from 1993 to 2014, within the MMA, O₃ has increased at an average rate of 0.22 ppb yr⁻¹ ($p < 0.01$), which is in marked contrast with the decline of 1.15 ppb yr⁻¹ ($p < 0.001$) observed in the Mexico City metropolitan area (MCMA) for the same period. No clear trend is observed from 1996 to 2014 within the Guadalajara metropolitan area (GMA).

Chemical composition of ambient PM_{2.5} over China and relationship to precursor emissions during 2005–2012

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Source: Atmos. Chem. Phys., 17, 9187–9203, 2017
<https://doi.org/10.5194/acp-17-9187-2017>

In this work, we presented the characteristics of PM_{2.5} chemical composition over China for the period of 2005–2012 by synthesis of in situ measurement data collected from literatures and satellite-based estimates using aerosol optical depth (AOD) data and the GEOS-Chem chemical transport model. We revealed the spatiotemporal variations in PM_{2.5} composition during 2005–2012 and investigated the driving forces behind the variations by examining the changes in precursor emissions using a bottom-up emission inventory. Both in situ observations and satellite-based estimates identified that secondary inorganic aerosols (i.e., sulfate, nitrate, and ammonium; SNA) ranked as the highest fraction of dust-free PM_{2.5} concentrations, followed by organic matter (OM) and black carbon (BC). For instance, satellite-based estimates found that SNA, OM, and BC contributed to 59, 33, and 8 %, respectively, of national population-weighted mean dust-free PM_{2.5} concentrations during 2005–2012. National population-weighted mean PM_{2.5} concentration increased from 63.9 µg m⁻³ in 2005 to 75.2 µg m⁻³ in 2007 and subsequently decreased to 66.9 µg m⁻³ from 2007 to 2012. Variations in PM_{2.5} concentrations are mainly driven by the decrease in sulfate and the increase in nitrate. Population-weighted mean sulfate concentration decreased by 2.4 % yr⁻¹ during 2005–2012 (from 14.4 to 12.9 µg m⁻³), while population-weighted mean nitrate concentration increased by 3.4 % yr⁻¹ during 2005–2012 (from 9.8 to 12.2 µg m⁻³), largely offsetting the decrease in sulfate concentrations. By examining the emission data from the Multi-resolution Emission Inventory for China (MEIC), we found that the changes in sulfate and nitrate concentrations were in line with the decrease in SO₂ emissions and the increase in NO_x emissions during the same period. The desulfurization regulation in power plants enforced around 2005 has been the primary contributor to the SO₂ emission reduction since 2006. In contrast, growth of energy consumption and lack of control measures for NO_x resulted in a persistent increase in NO_x emissions until the installation of denitrification devices on power plants late in 2011, which began to take effect in 2012. The results of this work indicate that the synchronized abatement of emissions for multipollutants is necessary for reducing ambient PM_{2.5} concentrations over China.

Uncertainty assessment and applicability of an inversion method for volcanic ash forecasting

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Source: Atmos. Chem. Phys., 17, 9205-9222, 2017
<https://doi.org/10.5194/acp-17-9205-2017>

Significant improvements in the way we can observe and model volcanic ash clouds have been obtained since the 2010 Eyjafjallajökull eruption. One major development has been the application of data assimilation techniques, which combine models and satellite observations such that an optimal understanding of ash clouds can be gained. Still, questions remain regarding the degree to which the forecasting capabilities are improved by inclusion of such techniques and how these improvements depend on the data input. This study explores how different satellite data and different uncertainty assumptions of the satellite and a priori emissions affect the calculated volcanic ash emission estimate, which is computed by an inversion method that couples the satellite retrievals and a priori emissions with dispersion model data. Two major ash episodes over 4 days in April and May of the 2010 Eyjafjallajökull eruption are studied. Specifically, inversion calculations are done for four different satellite data sets with different size distribution assumptions in the retrieval. A reference satellite data set is chosen, and the range between the minimum and maximum 4-day average load of hourly retrieved ash is 121 % in April and 148 % in May, compared to the reference. The corresponding a posteriori maximum and minimum emission sum found for these four satellite retrievals is 26 and 47 % of the a posteriori reference estimate for the same two periods, respectively. Varying the assumptions made in the satellite retrieval is seen to affect the a posteriori emissions and modelled ash column loads, and modelled column loads therefore have uncertainties connected to them depending on the uncertainty in the satellite retrieval. By further exploring our uncertainty estimates connected to a priori emissions and the mass load uncertainties in the satellite data, the uncertainty in the a priori estimate is found in this case to have an order-of-magnitude-greater impact on the a posteriori solution than the mass load uncertainties in the satellite. Part of this is explained by a too-high a priori estimate used in this study that is reduced by around half in the a posteriori reference estimate. Setting large uncertainties connected to both a priori and satellite mass load shows that they compensate each other, but the a priori uncertainty is found to be most sensitive. Because of this, an inversion-based emission estimate in a forecasting setting needs well-tested and well-considered assumptions on uncertainties for the a priori emission and satellite data. The quality of using the inversion in a forecasting environment is tested by adding gradually, with time, more observations to improve the estimated height versus time evolution of Eyjafjallajökull ash emissions. We show that the initially too-high a priori emissions are reduced effectively when using just 12 h of satellite observations. More satellite observations (> 12 h), in the Eyjafjallajökull case, place the volcanic injection at higher altitudes. Adding additional satellite observations (> 36 h) changes the a posteriori emissions to only a small extent for May and minimal for the April period, because the ash is dispersed and transported effectively out of the domain after 1–2 days. A best-guess emission estimate for the forecasting period was constructed by averaging the last 12 h of the a posteriori emission. Using this emission for a forecast simulation leads to better performance, especially compared to model simulations with no further emissions over the forecast period in the case of a continued volcanic eruption activity. Because of undetected ash in the satellite retrieval and diffusion in the model, the forecast simulations generally contain more ash than the observed fields, and the model ash is more spread out. Overall, using the a posteriori emissions in our model reduces the uncertainties in the ash plume forecast, because it corrects effectively for false-

positive satellite retrievals, temporary gaps in observations, and false a priori emissions in the window of observation.

Wildfire air pollution hazard during the 21st century

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Source: *Atmos. Chem. Phys.*, 17, 9223-9236, 2017
<https://doi.org/10.5194/acp-17-9223-2017>

Wildfires pose a significant risk to human livelihoods and are a substantial health hazard due to emissions of toxic smoke. Previous studies have shown that climate change, increasing atmospheric CO₂, and human demographic dynamics can lead to substantially altered wildfire risk in the future, with fire activity increasing in some regions and decreasing in others. The present study re-examines these results from the perspective of air pollution risk, focussing on emissions of airborne particulate matter (PM_{2.5}), combining an existing ensemble of simulations using a coupled fire–dynamic vegetation model with current observation-based estimates of wildfire emissions and simulations with a chemical transport model. Currently, wildfire PM_{2.5} emissions exceed those from anthropogenic sources in large parts of the world. We further analyse two extreme sets of future wildfire emissions in a socio-economic, demographic climate change context and compare them to anthropogenic emission scenarios reflecting current and ambitious air pollution legislation. In most regions of the world, ambitious reductions of anthropogenic air pollutant emissions have the potential to limit mean annual pollutant PM_{2.5} levels to comply with World Health Organization (WHO) air quality guidelines for PM_{2.5}. Worst-case future wildfire emissions are not likely to interfere with these annual goals, largely due to fire seasonality, as well as a tendency of wildfire sources to be situated in areas of intermediate population density, as opposed to anthropogenic sources that tend to be highest at the highest population densities. However, during the high-fire season, we find many regions where future PM_{2.5} pollution levels can reach dangerous levels even for a scenario of aggressive reduction of anthropogenic emissions.

NO_x emission trends over Chinese cities estimated from OMI observations during 2005 to 2015

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Source: *Atmos. Chem. Phys.*, 17, 9261-9275, 2017
<https://doi.org/10.5194/acp-17-9261-2017>

Satellite nitrogen dioxide (NO₂) observations have been widely used to evaluate emission changes. To determine trends in nitrogen oxides (NO_x) emission over China, we used a method independent of chemical transport models to quantify the NO_x emissions from 48 cities and seven power plants over China, on the basis of Ozone Monitoring Instrument (OMI) NO₂ observations from 2005 to 2015. We found that NO_x emissions over 48 Chinese cities increased by 52 % from 2005 to 2011 and decreased by

21 % from 2011 to 2015. The decrease since 2011 could be mainly attributed to emission control measures in power sector; while cities with different dominant emission sources (i.e., power, industrial, and transportation sectors) showed variable emission decline timelines that corresponded to the schedules for emission control in different sectors. The time series of the derived NO_x emissions was consistent with the bottom-up emission inventories for all power plants ($r = 0.8$ on average), but not for some cities ($r = 0.4$ on average). The lack of consistency observed for cities was most probably due to the high uncertainty of bottom-up urban emissions used in this study, which were derived from downscaling the regional-based emission data to city level by using spatial distribution proxies.

An assessment of ozone mini-hole representation in reanalyses over the Northern Hemisphere

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Source: Atmos. Chem. Phys., 17, 9277-9289, 2017
<https://doi.org/10.5194/acp-17-9277-2017>

An ozone mini-hole is a synoptic-scale region with strongly decreased total column ozone resulting from dynamical processes. Using total column measurements from the Ozone Monitoring Instrument and ozone profile measurements from the Microwave Limb Sounder, we evaluate the accuracy of mini-hole representation in five reanalyses. This study provides a metric of the reanalyses' ability to capture dynamically driven ozone variability. The reanalyses and the measurements show similar seasonal variability and geographical distributions of mini-holes; however, all of the reanalyses underestimate the number of mini-holes and their area, and in many reanalyses their location displays an eastward bias. The reanalyses' underestimation of mini-hole number ranges from about 34 to about 83 %. The mini-hole vertical representation in the reanalyses agrees well with that in the MLS measurements and, furthermore, is consistent with previously reported mechanisms for mini-hole formation. The skill of the reanalyses is not closely tied to the ozone fields assimilated, suggesting that the dynamics of the reanalysis models are more important than the assimilated ozone fields to reproducing ozone mini-holes.

On the multiday haze in the Asian continental outflow: the important role of synoptic conditions combined with regional and local sources

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Source: Atmos. Chem. Phys., 17, 9311-9332, 2017
<https://doi.org/10.5194/acp-17-9311-2017>

The air quality of the megacities in populated and industrialized regions like East Asia is affected by both local and regional emission sources. The combined effect of regional transport and local emissions on

multiday haze was investigated through a synthetic analysis of PM_{2.5} sampled at both an urban site in Seoul, South Korea and an upwind background site on Deokjeok Island over the Yellow Sea during a severe multiday haze episode in late February 2014. Inorganic components and carbonaceous species of daily PM_{2.5} samples were measured, and gaseous pollutants, local meteorological factors, and synoptic meteorological conditions were also determined. A dominance of fine-mode particles (PM_{2.5}/PM₁₀ ~ 0.8), a large secondary inorganic fraction (76 %), high OC/EC (> 7), and highly oxidized aerosols (oxygen-to-carbon ratio of ~ 0.6 and organic-mass-to-carbon ratio of ~ 1.9) under relatively warm, humid, and stagnant conditions characterize the multiday haze episode in Seoul; however, the early and late stages of the episode show different chemical compositions of PM_{2.5}. High concentrations of sulfate in both Seoul and the upwind background in the early stage suggest a significant regional influence on the onset of the multiday haze. At the same time, high concentrations of nitrate and organic compounds in Seoul, which are local and highly correlated with meteorological factors, suggest the contribution of local emissions and secondary formation under stagnant meteorological conditions to the haze. A slow eastward-moving high-pressure system from southern China to the East China Sea induces the regional transport of aerosols and potential gaseous precursors for secondary aerosols from the North China Plain in the early stage but provides stagnant conditions conducive to the accumulation and the local formation of aerosols in the late stage. A blocking ridge over Alaska that developed during the episode hinders the zonal propagation of synoptic-scale systems and extends the haze period to several days. This study provides chemical insights into haze development sequentially by regional transport and local sources, and shows that the synoptic condition plays an important role in the dynamical evolution of long-lasting haze in the Asian continental outflow region.

Net ecosystem exchange and energy fluxes measured with the eddy covariance technique in a western Siberian bog

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Source: Atmos. Chem. Phys., 17, 9333-9345, 2017
<https://doi.org/10.5194/acp-17-9333-2017>

Very few studies of ecosystem–atmosphere exchange involving eddy covariance data have been conducted in Siberia, with none in the western Siberian middle taiga. This work provides the first estimates of carbon dioxide (CO₂) and energy budgets in a typical bog of the western Siberian middle taiga based on May–August measurements in 2015. The footprint of measured fluxes consisted of a homogeneous mixture of tree-covered ridges and hollows with the vegetation represented by typical sedges and shrubs. Generally, the surface exchange rates resembled those of pine-covered bogs elsewhere. The surface energy balance closure approached 100 %. Net CO₂ uptake was comparatively high, summing up to 202 gC m^{−2} for the four measurement months, while the Bowen ratio was seasonally stable at 28 %. The ecosystem turned into a net CO₂ source during several front passage events in June and July. The periods of heavy rain helped keep the water table at a sustainably high level, preventing a usual drawdown in summer. However, because of the cloudy and rainy weather, the observed fluxes might rather represent the special weather conditions of 2015 than their typical magnitudes.

Trends and annual cycles in soundings of Arctic tropospheric ozone

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Source: Atmos. Chem. Phys., 17, 9347-9364, 2017
<https://doi.org/10.5194/acp-17-9347-2017>

Ozone soundings from nine Nordic stations have been homogenized and interpolated to standard pressure levels. The different stations have very different data coverage; the longest period with data is from the end of the 1980s to 2014.

At each pressure level the homogenized ozone time series have been analysed with a model that includes both low-frequency variability in the form of a polynomial, an annual cycle with harmonics, the possibility for low-frequency variability in the annual amplitude and phasing, and either white noise or noise given by a first-order autoregressive process. The fitting of the parameters is performed with a Bayesian approach not only giving the mean values but also confidence intervals.

The results show that all stations agree on a well-defined annual cycle in the free troposphere with a relatively confined maximum in the early summer. Regarding the low-frequency variability, it is found that Scoresbysund, Ny Ålesund, Sodankylä, Eureka, and Ørland show similar, significant signals with a maximum near 2005 followed by a decrease. This change is characteristic for all pressure levels in the free troposphere. A significant change in the annual cycle was found for Ny Ålesund, Scoresbysund, and Sodankylä. The changes at these stations are in agreement with the interpretation that the early summer maximum is appearing earlier in the year.

The results are shown to be robust to the different settings of the model parameters such as the order of the polynomial, number of harmonics in the annual cycle, and the type of noise.

Temporal characteristics of atmospheric ammonia and nitrogen dioxide over China based on emission data, satellite observations and atmospheric transport modeling since 1980

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Source: Atmos. Chem. Phys., 17, 9365-9378, 2017
<https://doi.org/10.5194/acp-17-9365-2017>

China is experiencing intense air pollution caused in large part by anthropogenic emissions of reactive nitrogen (Nr). Atmospheric ammonia (NH₃) and nitrogen dioxide (NO₂) are the most important precursors for Nr compounds (including N₂O₅, HNO₃, HONO and particulate NO₃⁻ and NH₄⁺) in the

atmosphere. Understanding the changes in NH₃ and NO₂ has important implications for the regulation of anthropogenic Nr emissions and is a requirement for assessing the consequence of environmental impacts. We conducted the temporal trend analysis of atmospheric NH₃ and NO₂ on a national scale since 1980 based on emission data (during 1980–2010), satellite observation (for NH₃ since 2008 and for NO₂ since 2005) and atmospheric chemistry transport modeling (during 2008–2015).

Based on the emission data, during 1980–2010, significant continuous increasing trends in both NH₃ and NO_x were observed in REAS (Regional Emission inventory in Asia, for NH₃ 0.17 and for NO_x 0.16 kg N ha⁻¹ yr⁻²) and EDGAR (Emissions Database for Global Atmospheric Research, for NH₃ 0.24 and for NO_x 0.17 kg N ha⁻¹ yr⁻²) over China. Based on the satellite data and atmospheric chemistry transport model (CTM) MOZART-4 (Model for Ozone and Related chemical Tracers, version 4), the NO₂ columns over China increased significantly from 2005 to 2011 and then decreased significantly from 2011 to 2015; the satellite-retrieved NH₃ columns from 2008 to 2014 increased at a rate of 2.37 % yr⁻¹. The decrease in NO₂ columns since 2011 may result from more stringent strategies taken to control NO_x emissions during the 12th Five Year Plan, while no control policy has focused on NH₃ emissions. Our findings provided an overall insight into the temporal trends of both NO₂ and NH₃ since 1980 based on emission data, satellite observations and atmospheric transport modeling. These findings can provide a scientific background for policy makers that are attempting to control atmospheric pollution in China. Moreover, the multiple datasets used in this study have implications for estimating long-term Nr deposition datasets to assess its impact on soil, forest, water and greenhouse balance.

Plume-exit modeling to determine cloud condensation nuclei activity of aerosols from residential biofuel combustion

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Source: Atmos. Chem. Phys., 17, 9399-9415, 2017
<https://doi.org/10.5194/acp-17-9399-2017>

Residential biofuel combustion is an important source of aerosols and gases in the atmosphere. The change in cloud characteristics due to biofuel burning aerosols is uncertain, in part, due to the uncertainty in the added number of cloud condensation nuclei (CCN) from biofuel burning. We provide estimates of the CCN activity of biofuel burning aerosols by explicitly modeling plume dynamics (coagulation, condensation, chemical reactions, and dilution) in a young biofuel burning plume from emission until plume exit, defined here as the condition when the plume reaches ambient temperature and specific humidity through entrainment. We found that aerosol-scale dynamics affect CCN activity only during the first few seconds of evolution, after which the CCN efficiency reaches a constant value. Homogenizing factors in a plume are co-emission of semi-volatile organic compounds (SVOCs) or emission at small particle sizes; SVOC co-emission can be the main factor determining plume-exit CCN for hydrophobic or small particles. Coagulation limits emission of CCN to about 10¹⁶ per kilogram of fuel. Depending on emission factor, particle size, and composition, some of these particles may not activate at low supersaturation (ssat). Hygroscopic Aitken-mode particles can contribute to CCN through self-coagulation but have a small effect on the CCN activity of accumulation-mode particles, regardless of composition differences. Simple models (monodisperse coagulation and average hygroscopicity) can

be used to estimate plume-exit CCN within about 20 % if particles are unimodal and have homogeneous composition, or when particles are emitted in the Aitken mode even if they are not homogeneous. On the other hand, if externally mixed particles are emitted in the accumulation mode without SVOCs, an average hygroscopicity overestimates emitted CCN by up to a factor of 2. This work has identified conditions under which particle populations become more homogeneous during plume processes. This homogenizing effect requires the components to be truly co-emitted, rather than sequentially emitted.

Sources of particulate matter components in the Athabasca oil sands region: investigation through a comparison of trace element measurement methodologies

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Source: Atmos. Chem. Phys., 17, 9435-9449, 2017
<https://doi.org/10.5194/acp-17-9435-2017>

The province of Alberta, Canada, is home to three oil sands regions which, combined, contain the third largest deposit of oil in the world. Of these, the Athabasca oil sands region is the largest. As part of Environment and Climate Change Canada's program in support of the Joint Canada-Alberta Implementation Plan for Oil Sands Monitoring program, concentrations of trace elements in PM_{2.5} (particulate matter smaller than 2.5 μm in diameter) were measured through two campaigns that involved different methodologies: a long-term filter campaign and a short-term intensive campaign. In the long-term campaign, 24 h filter samples were collected once every 6 days over a 2-year period (December 2010–November 2012) at three air monitoring stations in the regional municipality of Wood Buffalo. For the intensive campaign (August 2013), hourly measurements were made with an online instrument at one air monitoring station; daily filter samples were also collected. The hourly and 24 h filter data were analyzed individually using positive matrix factorization. Seven emission sources of PM_{2.5} trace elements were thereby identified: two types of upgrader emissions, soil, haul road dust, biomass burning, and two sources of mixed origin. The upgrader emissions, soil, and haul road dust sources were identified through both the methodologies and both methodologies identified a mixed source, but these exhibited more differences than similarities. The second upgrader emissions and biomass burning sources were only resolved by the hourly and filter methodologies, respectively. The similarity of the receptor modeling results from the two methodologies provided reassurance as to the identity of the sources. Overall, much of the PM_{2.5}-related trace elements were found to be anthropogenic, or at least to be aerosolized through anthropogenic activities. These emissions may in part explain the previously reported higher levels of trace elements in snow, water, and biota samples collected near the oil sands operations.

A review of current knowledge concerning PM_{2.5} chemical composition, aerosol optical properties and their relationships across China

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Source: Atmos. Chem. Phys., 17, 9485-9518, 2017
<https://doi.org/10.5194/acp-17-9485-2017>

To obtain a thorough knowledge of PM_{2.5} chemical composition and its impact on aerosol optical properties across China, existing field studies conducted after the year 2000 are reviewed and summarized in terms of geographical, interannual and seasonal distributions. Annual PM_{2.5} was up to 6 times the National Ambient Air Quality Standards (NAAQS) in some megacities in northern China. Annual PM_{2.5} was higher in northern than southern cities, and higher in inland than coastal cities. In a few cities with data longer than a decade, PM_{2.5} showed a slight decrease only in the second half of the past decade, while carbonaceous aerosols decreased, sulfate (SO₄²⁻) and ammonium (NH₄⁺) remained at high levels, and nitrate (NO₃⁻) increased. The highest seasonal averages of PM_{2.5} and its major chemical components were typically observed in the cold seasons. Annual average contributions of secondary inorganic aerosols to PM_{2.5} ranged from 25 to 48 %, and those of carbonaceous aerosols ranged from 23 to 47 %, both with higher contributions in southern regions due to the frequent dust events in northern China. Source apportionment analysis identified secondary inorganic aerosols, coal combustion and traffic emission as the top three source factors contributing to PM_{2.5} mass in most Chinese cities, and the sum of these three source factors explained 44 to 82 % of PM_{2.5} mass on annual average across China. Biomass emission in most cities, industrial emission in industrial cities, dust emission in northern cities and ship emission in coastal cities are other major source factors, each of which contributed 7–27 % to PM_{2.5} mass in applicable cities.

The geographical pattern of scattering coefficient (*bsp*) was similar to that of PM_{2.5}, and that of aerosol absorption coefficient (*bap*) was determined by elemental carbon (EC) mass concentration and its coating. *bsp* in ambient condition of relative humidity (RH) = 80 % can be amplified by about 1.8 times that under dry conditions. Secondary inorganic aerosols accounted for about 60 % of aerosol extinction coefficient (*bext*) at RH greater than 70 %. The mass scattering efficiency (MSE) of PM_{2.5} ranged from 3.0 to 5.0 m² g⁻¹ for aerosols produced from anthropogenic emissions and from 0.7 to 1.0 m² g⁻¹ for natural dust aerosols. The mass absorption efficiency (MAE) of EC ranged from 6.5 to 12.4 m² g⁻¹ in urban environments, but the MAE of water-soluble organic carbon was only 0.05 to 0.11 m² g⁻¹. Historical emission control policies in China and their effectiveness were discussed based on available chemically resolved PM_{2.5} data, which provides the much needed knowledge for guiding future studies and emissions policies.

Mixing state of oxalic acid containing particles in the rural area of Pearl River Delta, China: implications for the formation mechanism of oxalic acid

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Source: Atmos. Chem. Phys., 17, 9519-9533, 2017
<https://doi.org/10.5194/acp-17-9519-2017>

The formation of oxalic acid and its mixing state in atmospheric particulate matter (PM) were studied using a single-particle aerosol mass spectrometer (SPAMS) in the summer and winter of 2014 in Heshan, a supersite in the rural area of the Pearl River Delta (PRD) region in China. Oxalic-acid-containing particles accounted for 2.5 and 2.7 % in total detected ambient particles in summer and winter, respectively. Oxalic acid was measured in particles classified as elemental carbon (EC), organic carbon (OC), elemental and organic carbon (ECOC), biomass burning (BB), heavy metal (HM), secondary (Sec), sodium-potassium (NaK), and dust. Oxalic acid was found predominantly mixing with sulfate and nitrate during the whole sampling period, likely due to aqueous-phase reactions. In summer, oxalic-acid-containing particle number and ozone concentration followed a very similar trend, which may reflect the significant contribution of photochemical reactions to oxalic acid formation. The HM particles were the most abundant oxalic acid particles in summer and the diurnal variations in peak area of iron and oxalic acid show opposite trends, which suggests a possible loss of oxalic acid through the photolysis of iron oxalato-complexes during the strong photochemical activity period. In wintertime, carbonaceous particles contained a substantial amount of oxalic acid as well as abundant carbon clusters and BB markers. The general existence of nitric acid in oxalic-acid-containing particles indicates an acidic environment during the formation process of oxalic acid. The peak areas of nitrate, sulfate and oxalic acid had similar temporal change in the carbonaceous type oxalic acid particles, and the organosulfate-containing oxalic acid particles correlated well with total oxalic acid particles during the haze episode, which suggests that the formation of oxalic acid is closely associated with the oxidation of organic precursors in the aqueous phase.

Volatile organic compounds (VOCs) in photochemically aged air from the eastern and western Mediterranean

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Source: Atmos. Chem. Phys., 17, 9547-9566, 2017
<https://doi.org/10.5194/acp-17-9547-2017>

During the summertime CYPHEX campaign (Cyprus Photochemical Experiment 2014) in the eastern Mediterranean, multiple volatile organic compounds (VOCs) were measured from a 650 m hilltop site in western Cyprus (34° 57' N/32° 23' E). Periodic shifts in the northerly Etesian winds resulted in the site being alternately impacted by photochemically processed emissions from western (Spain, France, Italy) and eastern (Turkey, Greece) Europe. Furthermore, the site was situated within the residual layer/free troposphere during some nights which were characterized by high ozone and low relative humidity levels. In this study we examine the temporal variation of VOCs at the site. The sparse Mediterranean scrub vegetation generated diel cycles in the reactive biogenic hydrocarbon isoprene, from very low values at night to a diurnal median level of 80–100 pptv. In contrast, the oxygenated volatile organic compounds (OVOCs) methanol and acetone exhibited weak diel cycles and were approximately an order of magnitude higher in mixing ratio (ca. 2.5–3 ppbv median level by day, range: ca. 1–8 ppbv) than the locally emitted isoprene and aromatic compounds such as benzene and toluene. Acetic acid was present at mixing ratios between 0.05 and 4 ppbv with a median level of ca. 1.2 ppbv during the daytime. When data points directly affected by the residual layer/free troposphere were excluded, the acid followed a pronounced diel cycle, which was influenced by various local effects including photochemical production and loss, direct emission, dry deposition and scavenging from advecting air in fog banks. The Lagrangian model FLEXPART was used to determine transport patterns and photochemical processing times (between 12 h and several days) of air masses originating from eastern and western Europe. Ozone and many OVOC levels were ~ 20 and ~ 30 –60 % higher, respectively, in air arriving from the east. Using the FLEXPART calculated transport time, the contribution of photochemical processing, sea surface contact and dilution was estimated. Methanol and acetone decreased with residence time in the marine boundary layer (MBL) with loss rate constants of 0.74 and 0.53 day⁻¹ from eastern Europe and 0.70 and 0.34 day⁻¹ from western Europe, respectively. Simulations using the EMAC model underestimate these loss rates. The missing sink in the calculation is most probably an oceanic uptake enhanced by microbial consumption of methanol and acetone, although the temporal and spatial variability in the source strength on the continents might play a role as well. Correlations between acetone and methanol were weaker in western air masses ($r^2 = 0.68$), but were stronger in air masses measured after the shorter transport time from the east ($r^2 = 0.73$).

Spatial extent of new particle formation events over the Mediterranean Basin from multiple ground-based and airborne measurements

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Source: *Atmos. Chem. Phys.*, 17, 9567–9583, 2017
<https://doi.org/10.5194/acp-17-9567-2017>

Over the last two decades, new particle formation (NPF), i.e., the formation of new particle clusters from gas-phase compounds followed by their growth to the 10–50 nm size range, has been extensively observed in the atmosphere at a given location, but their spatial extent has rarely been assessed. In this work, we use aerosol size distribution measurements performed simultaneously at Erba (Corsica) and Finokalia (Crete) over a 1-year period to analyze the occurrence of NPF events in the Mediterranean

area. The geographical location of these two sites, as well as the extended sampling period, allows us to assess the spatial and temporal variability in atmospheric nucleation at a regional scale. Finokalia and Erso show similar seasonalities in the monthly average nucleation frequencies, growth rates, and nucleation rates, although the two stations are located more than 1000 km away from each other. Within this extended period, aerosol size distribution measurements were performed during an intensive campaign (3 July to 12 August 2013) from a ground-based station on the island of Mallorca, as well as onboard the ATR-42 research aircraft. This unique combination of stationary and mobile measurements provides us with detailed insights into the horizontal and vertical development of the NPF process on a daily scale. During the intensive campaign, nucleation events occurred simultaneously both at Erso and Mallorca over delimited time slots of several days, but different features were observed at Finokalia. The results show that the spatial extent of the NPF events over the Mediterranean Sea might be as large as several hundreds of kilometers, mainly determined by synoptic conditions. Airborne measurements gave additional information regarding the origin of the clusters detected above the sea. The selected cases depicted contrasting situations, with clusters formed in the marine boundary layer or initially nucleated above the continent or in the free troposphere (FT) and further transported above the sea.

Possible climatic implications of high-altitude black carbon emissions

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Source: Atmos. Chem. Phys., 17, 9623-9644, 2017
<https://doi.org/10.5194/acp-17-9623-2017>

On account of its strong absorption of solar and terrestrial radiation, black carbon (BC) aerosol is known to impact large-scale systems, such as the Asian monsoon and the Himalayan glaciers, in addition to affecting the thermal structure of the lower atmosphere. While most studies focus on the near-surface abundance and impacts of BC, our study examines the implications of sharp and confined layers of high BC concentration (called elevated BC layers) at altitudes more than 4 km over the Indian region using the online regional chemistry transport model (WRF-Chem) simulations. These elevated BC layers were revealed in the recent in situ measurements using high-altitude balloons carried out on 17 March 2010, 8 January 2011 and 25 April 2011. Our study demonstrates that high-flying aircraft (with emissions from the regionally fine-tuned MACCity inventory) are the most likely cause of these elevated BC layers. Furthermore, we show that such aircraft-emitted BC can be transported to upper tropospheric or lower stratospheric heights (~ 17 km) aided by the strong monsoonal convection occurring over the region, which is known to overshoot the tropical tropopause, leading to the injection of tropospheric air mass (along with its constituent aerosols) into the stratosphere. We show observational evidence for such an intrusion of tropospheric BC into the stratosphere over the Indian region using extinction coefficient and particle depolarisation ratio data from CALIOP Lidar on-board the CALIPSO satellite. We hypothesise that such intrusions of BC into the lower stratosphere and its consequent longer residence time in the stratosphere have significant implications for stratospheric ozone, especially considering the already reported ozone-depleting potential of BC.

Sources of springtime surface black carbon in the Arctic: an adjoint analysis for April 2008

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Source: *Atmos. Chem. Phys.*, 17, 9697-9716, 2017

<https://doi.org/10.5194/acp-17-9697-2017>

We quantify source contributions to springtime (April 2008) surface black carbon (BC) in the Arctic by interpreting surface observations of BC at five receptor sites (Denali, Barrow, Alert, Zeppelin, and Summit) using a global chemical transport model (GEOS-Chem) and its adjoint. Contributions to BC at Barrow, Alert, and Zeppelin are dominated by Asian anthropogenic sources (40–43 %) before 18 April and by Siberian open biomass burning emissions (29–41 %) afterward. In contrast, Summit, a mostly free tropospheric site, has predominantly an Asian anthropogenic source contribution (24–68 %, with an average of 45 %). We compute the adjoint sensitivity of BC concentrations at the five sites during a pollution episode (20–25 April) to global emissions from 1 March to 25 April. The associated contributions are the combined results of these sensitivities and BC emissions. Local and regional anthropogenic sources in Alaska are the largest anthropogenic sources of BC at Denali (63 % of total anthropogenic contributions), and natural gas flaring emissions in the western extreme north of Russia (WENR) are the largest anthropogenic sources of BC at Zeppelin (26 %) and Alert (13 %). We find that long-range transport of emissions from Beijing–Tianjin–Hebei (also known as Jing–Jin–Ji), the biggest urbanized region in northern China, contribute significantly (~10 %) to surface BC across the Arctic. On average, it takes ~12 days for Asian anthropogenic emissions and Siberian biomass burning emissions to reach the Arctic lower troposphere, supporting earlier studies. Natural gas flaring emissions from the WENR reach Zeppelin in about a week. We find that episodic transport events dominate BC at Denali (87 %), a site outside the Arctic front, which is a strong transport barrier. The relative contribution of these events to surface BC within the polar dome is much smaller (~50 % at Barrow and Zeppelin and ~10 % at Alert). The large contributions from Asian anthropogenic sources are predominately in the form of chronic pollution (~40 % at Barrow, 65 % at Alert, and 57 % at Zeppelin) on about a 1-month timescale. As such, it is likely that previous studies using 5- or 10-day trajectory analyses strongly underestimated the contribution from Asia to surface BC in the Arctic.

Aerosol And Air Quality Research

Analysing Temporal Trends in the Ratios of PM_{2.5}/PM₁₀ in the UK

Said Munir

Source: Volume 17, No. 1, January 2017, Pages 34-48 doi: 10.4209/aaqr.2016.02.0081

The size of atmospheric Particulate Matter (PM) is important as a determining factor for how long the particle stays in the atmosphere, and where it deposits in the human respiratory tract. Therefore, it is important to analyse PM_{2.5}/PM₁₀ ratios as an indicator of the fine particles and determine how the ratios vary both in space and time. In this paper mostly robust statistics, which are not sensitive to non-normal distributions and to extreme values in both tails of the distributions are applied to assess temporal trends in PM_{2.5}, PM₁₀ and their ratios. PM_{2.5}/PM₁₀ ratios demonstrated considerable temporal and spatial variability in the UK and 5 years median ranged from 0.4 to 0.8, resulting in overall median of 0.65. However, trend in PM_{2.5}/PM₁₀ ratios averaged over the 46 monitoring sites was insignificant. For further investigations: (a) Trends are adjusted for meteorological effect; (b) The emissions of PM₁₀ and PM_{2.5}(kilotonnes year⁻¹) and their ratios from 1990 to 2013 are analysed; (c) Temporal trends of the secondary particles (nitrate and sulphate) are analysed from 2000 to 2014; and (d) The diurnal, weekly and annual cycles in the ratios of PM_{2.5} and PM₁₀ are analysed.

Keywords: Fine Particulate Matter; PM₁₀; PM_{2.5}; PM_{2.5}/PM₁₀ ratio; Air Pollution.

Single-Particle Characterizations of Ambient Aerosols during a Wintertime Pollution Episode in Nanning: Local Emissions vs. Regional Transport

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Source: Volume 17, No. 1, January 2017, Pages 49-58 doi: 10.4209/aaqr.2016.01.0026

Ambient aerosol during a heavily polluted episode in wintertime was characterized using real-time single particle aerosol mass spectrometry (SPAMS) in urban Nanning, a capital city in the Southwestern China. More than two million individual particles analyzed by SPAMS were classified into 8 major clusters based on the mass spectral patterns. A group of vanadium-rich particles were identified as the emissions from mining and smelting of vanadium mineral and were taken as markers of regional transported industrial emissions when air masses traversed northeast inland regions from Nanning. Our observations suggest that biomass burning particles derived from industrial heat and electricity cogeneration processes could have a significant impact on the urban air quality without proper emission controls.

Keywords: Ambient aerosol; Bagasse combustion; Single particle mass spectrometry; Nanning.

Effects of Polluted Air-Masses Advection on Atmospheric Particles in a Semi-Rural Site in South Italy by SEM-EDX Analysis

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Source: Volume 17, No. 1, January 2017, Pages 69-83 doi: 10.4209/aaqr.2016.05.0216

The variation of aerosol properties under polluted air masses advection was studied in a semi-rural site in South Italy, by means of SEM-EDX analysis performed on particles collected on 13-stages impactor filters. Radiometric measurements, HYSPLIT back-trajectories and NAAPS maps helped to choose four measurement days of polluted air mass circulation, two of them collected in the warm season (31 July 2008, 16 September 2010) and two in the cold one (16 April 2009, 18 March 2010). Polluted aerosol characteristics were compared to those under background (BG) conditions (8 February 2011), highlighting differences in the particles chemical and morphological properties. One of the signatures of the air mass transport in the coarse fraction was the higher content of particles containing S, i.e., S-reacted, (27.5% on average) in comparison with BG conditions (1%). Two main sources of transported aerosols were identified: industrial processes and biomass burning, with fly ash, metal and S-rich particles in the first case, and K-salts and nitrate-coated Ca-bearing-particles in the second. Single particle analysis on the coarse fraction allowed large agglomerates of soot to be identified, with inclusions of silicate particles rich in Cu and Zn, Ca-S, fly ash and metals particles that are a clear indication of extensive modifications of aerosol size, chemical composition and, likely, radiative properties. In finer stages ($EAD \leq 0.94 \mu m$) concurrent collection of organic and inorganic particles originated an agglomerate state matter mainly characterized by K and S for polluted conditions and by K only for BG.

Keywords: Atmospheric aerosol; Long-range transport; Air pollution.

The Concentrations, Formations, Relationships and Modeling of Sulfate, Nitrate and Ammonium (SNA) Aerosols over China

Zhaoyu Cao, Xuehua Zhou, Yujie Ma, Linpeng Wang, Ruidong Wu, Bing Chen, Wenxing Wang

Source: Volume 17, No. 1, January 2017, Pages 84-97 doi: 10.4209/aaqr.2016.01.0020

Sulfate, nitrate and ammonium (SNA) are the dominant composition of secondary aerosols in the atmosphere and have a significant impact on public health, atmospheric chemistry process and climate. In this study, to evaluate SNA pollution in China, a first nationwide investigation derived from almost all published data in the field measurement before 2012 was carried out. The results show that SNA levels in China are about 3–5 times higher than those in USA and Europe. SNA account for $34.2 \pm 10.9\%$ in $PM_{2.5}$ and $28.2 \pm 8.5\%$ in PM_{10} . The highest SNA concentrations occur in urban areas of northern China. SNA all have peaks in winter, but the nadirs are in spring for sulfate and ammonium and in summer for nitrate. SOR (sulfur oxidation ratio) and NOR (nitrogen oxidation ratio) values show that the formations of sulfate and nitrate are distinct in different

regions and seasons. The low average $\text{NO}_3^-/\text{SO}_4^{2-}$ ratio (0.43 ± 0.26) indicates that the stationary emissions from coal combustion remain the main sources. There is a good relationship between ($2[\text{SO}_4^{2-}] + [\text{NO}_3^-]$) and $[\text{NH}_4^+]$ with near 1 slope, signifying that $(\text{NH}_4)_2\text{SO}_4$ and NH_4NO_3 are the predominant forms which SNA exist in particles in China. Based on the comprehensive observational data in China, the simulation for SNA aerosols by GISS in CMIP5 were evaluated.

Keywords: SNA; Field measurement; Model simulation; China.

A Typical Formation Mechanism of Heavy Haze-Fog Induced by Coal Combustion in an Inland City in North-Western China

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Source: Volume 17, No. 1, January 2017, Pages 98-107 doi: 10.4209/aaqr.2016.04.0143

$\text{PM}_{2.5}$, TSP, and the pollutant gases over Urumqi, an inland city near the Asian dust source region in north-western China, were measured for an entire year from March, 2008 to February, 2009. The chemical compositions of all these samples with daily visibilities were investigated for the sources and formation mechanisms of aerosol and the relationship of the optical property with various components in aerosols. The sources of the air pollutants that caused severe haze in Urumqi were clarified. It was found that the concentrations of SO_4^{2-} , Cl^- , NH_4^+ , and Na^+ in $\text{PM}_{2.5}$ in the heating period were more than 9 times of those in the non-heating period. The concentrations of total water soluble inorganic ions (TWSII) in $\text{PM}_{2.5}$ and TSP in the heating period were 7.5 and 5.5 times of those in the non-heating period. The enrichment factors of the pollution elements, e.g., S, Cd, As, Zn, Pb, Cu, Cr, and Na in $\text{PM}_{2.5}$ in the heating period were 3–13 times of those in the non-heating period. The formation mechanism of the severe haze and the micro-mechanism of the visibility reduction in Urumqi in the heating period, especially in winter, were quantitatively revealed. SNA (the three secondary inorganic ions, SO_4^{2-} , NO_3^- , and NH_4^+), organic matter (OM), and chloride (Cl^-) in $\text{PM}_{2.5}$ were the three main components to cause aerosol extinction in Urumqi. The contributions of SNA, OM, and Cl^- to aerosol extinction, i.e., to visibility reduction, were 57%, 20%, and 12% during the heating period from October 15 to April 15, and 67%, 17%, and 12% in winter, respectively. It was evident that the major source of the air pollutants that caused severe haze in Urumqi was from coal combustion, for most of these ions and pollution elements in aerosols were emitted from coal burning. The control of the quality and quantity of coal burning is the key to improve the air quality and the atmospheric visibility in Urumqi.

Keywords: Haze; Sources; Formation mechanism; Coal burning.

Characterization of Traffic-Related Particulate Matter Emissions in a Road Tunnel in Birmingham, UK: Trace Metals and Organic Molecular Markers

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Source: Volume 17, No. 1, January 2017, Pages 117-130 doi: 10.4209/aaqr.2016.01.0040

Road traffic is one of the key sources of particulate matter (PM) in urban areas, and an understanding of the chemical composition of traffic emissions is important for source apportionment analysis. In this study, PM samples were collected simultaneously in a road tunnel and at a background site in Birmingham (UK) and analysed for a suite of elemental and organic species (hopanes, alkanes and PAH) with an aim to characterize the vehicular emissions in a tunnel environment and to prepare a composite mixed fleet profile for PM_{2.5} traffic emissions. Large enrichment was observed for many organic and elemental species in the case of the tunnel samples with respect to the background site. The tunnel samples show a large enrichment of trace elements relative to the urban background with a mode at ca. 3 µm in the mass size distribution, indicative of emissions resulting from resuspension/abrasion sources. Cu, Ba and Sb were found to have the characteristic non-exhaust (brake wear) emission peaks in the coarse size range in the tunnel. A composite PM_{2.5} traffic profile was prepared using the data from the two sites, and was compared against previously reported profiles. The profile was also compared against other traffic profiles from Europe and USA, and was found to be very similar to the previously-reported PM_{2.5} composite traffic profile from the UK. However, the uncertainties associated with the species were found to be much lower in the case of the tunnel profile from this study, and we conclude that this profile would be very suitable for use in Chemical Mass Balance Model analyses for the UK and other countries with a similar road traffic fleet mix.

Keywords: Molecular markers; Source profile; Europe; Traffic; Road tunnel.

Number Concentrations and Modal Structure of Indoor/Outdoor Fine Particles in Four European Cities

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Source: Volume 17, No. 1, January 2017, Pages 131-146 doi: 10.4209/aaqr.2015.04.0267

Indoor/outdoor aerosol size distribution was measured in four European cities (Oslo–Norway, Prague–Czech Republic, Milan–Italy and Athens–Greece) during 2002 in order to examine the differences in the characteristics of the indoor/outdoor modal structure and to evaluate the effect of indoor sources to the aerosol size distributions. All the measurement sites were naturally ventilated and were occupied during the campaigns by permanent residents or for certain time periods by the technical staff responsible for the instrumentation. Outdoor particle number (PN) concentrations presented the higher values in Milan and Athens (median values $1.4 \times 10^4 \text{ \# cm}^{-3}$ and $2.9 \times 10^4 \text{ \# cm}^{-3}$ respectively) as a result of elevated outdoor emissions and led to correspondingly higher indoor values compared to Oslo and Prague. In absence of indoor activities, the indoor concentrations followed the fluctuations of the outdoor concentrations in all the measurement sites. Indoor activities (cooking, smoking, etc.) resulted in elevated indoor PN concentrations (maximum values ranging between $1.7 \times 10^5 \text{ \# cm}^{-3}$ and $3.2 \times 10^5 \text{ \# cm}^{-3}$) and to I/O ratios higher than one. The I/O ratios were size dependant and for periods without indoor activities, they presented the lowest values for particles < 50 nm (0.51 ± 0.15) and the ratios

increased with fine particle size (0.79 ± 0.12 for particles between 100–200 nm). The analysis of the modal structure showed that the indoor aerosol size distribution characteristics differ from the outdoors under the effect of indoor sources. The percentage of unimodal size distributions increased during indoor emissions, compared to periods without indoor sources, along with the number concentration of Aitken mode particles, indicating emissions in specific size ranges according to the type of the indoor source.

Keywords: Indoor/Outdoor aerosol; I/O ratio; Modal structure; Indoor sources.

Changes in Gas-Phase Air Pollutants across New York State, USA

Kevin L. Civerolo, Oliver V. Rattigan, H. Dirk Felton, James J. Schwab

Source: Volume 17, No. 1, January 2017, Pages 147-166 doi: 10.4209/aaqr.2016.04.0141

Over the past several decades, the New York State Department of Environmental Conservation (NYSDEC) has monitored air quality in urban, suburban, and rural locations across the state. Such data are used to determine compliance with national ambient air quality standards (NAAQS), as well as track the effectiveness of pollutant emission controls. This paper summarizes the changes in gas-phase criteria and related pollutants on an annual, seasonal and diurnal basis across New York, USA. Annual average and peak concentrations of carbon monoxide (CO), nitrogen dioxide (NO₂), and sulfur dioxide (SO₂) have decreased by 50% or more since 1980 across New York, generally consistent with anthropogenic emissions reductions across different source sectors over the past 35 years. At the same time, annual average concentrations of the secondary pollutant ozone (O₃) appear flat or are increasing even though peak daily 8-hour concentrations have declined. This pollutant will likely remain a challenging public health issue for the foreseeable future.

Keywords: Air pollution trends; Emissions trends; Criteria pollutants; Air monitoring; Air quality standards.

Investigating Criteria and Organic Air pollutant Emissions from Motorcycles by Using Various Ethanol-Gasoline Blends

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Source: Volume 17, No. 1, January 2017, Pages 167-175 doi: 10.4209/aaqr.2016.05.0174

Studies on the correlation between ethanol-gasoline blends and pollutant emissions of small engine motorcycles are scant. This study examined the effects of ethanol-gasoline blends, containing various ethanol contents, on air pollutant emissions from two four-stroke fuel-injection

motorcycles without engine adjustment. Three test blends, separately containing 15 (E15), 20 (E20), and 30 vol% (E30) ethanol in gasoline, were used to power the test motorcycles. Commercial unleaded gasoline was used as the reference fuel (as RF). The motorcycles were tested on a chassis dynamometer by using the Economic Commission for Europe test cycle. The target pollutants investigated in this study included criteria pollutants, volatile organic compounds (VOCs) and six species of organic air toxics. The results revealed that the emissions of CO, THC, total VOCs, alkanes, alkenes, and aromatic groups reduced when the ethanol-gasoline blends were used to fuel the motorcycles. E30 demonstrated approximately 1.2-fold increases in carbonyl group emissions compared with RF. Emissions of the target air toxics demonstrated a reduction potential on benzene, toluene, ethylbenzene, and xylene (BTEX), but increased the emissions of formaldehyde and acetaldehyde by 65% and 330%, respectively. Results also showed that the emission changes from fuel-injected motorcycle were generally smaller than the value of carburetor motorcycle. Fuel injection engine fueled with ethanol-gasoline blends may lead to emission reductions to CO, THC, and BTEX.

Keywords: Fuel-injection motorcycle; Small capacity engine; Renewable energy; Volatile organic compounds; Organic air toxics.

PAHs in Size Fractionate Mainstream Cigarette Smoke, Predictive Deposition and Associated Inhalation Risk

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Source: Volume 17, No. 1, January 2017, Pages 176-186 doi: 10.4209/aaqr.2015.03.0156

Size fractionated mainstream cigarette smoke (MCS) samples were collected with variable configuration cascade impactor (VCCI). Samples were extracted ultrasonically and analysis of sixteen priority polycyclic aromatic hydrocarbons (PAHs) was performed using high performance liquid chromatography (HPLC) coupled with UV-visible detector. Identification of PAHs were also carried out using gas chromatography coupled with mass spectrometry (GC-MS) technique. Data of size fractionate PAHs in MCS were used to calculate size dependent deposition in different compartment of human respiratory tract using multiple path particle dosimetry (MPPD) model. All brand cigarette smoke showed similar trends of particle mass and PAHs size distribution peaked at two sizes 0.3–0.1 μm and 0.75–1.13 μm aerodynamic diameter. All tested brands of MCS recorded around 48.75% of two and three-ring PAHs, 23 to 25% four-ring PAHs and 26–27% five and higher rings PAHs of total analyzed PAHs. Benzo[a]pyrene equivalent (B[a]P_{eq}) PAHs emission in MCS was found to be 110 ± 12 ($\mu \pm 1\sigma$) ng per cigarette for tested brands. Total deposition fraction in respiratory tract was found to be 0.69 ± 0.26 for tested size ranges. Average 5th and 95th percentile values of incremental lifetime cancer risk (ILCR) for smoker due to PAHs exposure were found to be 1.3×10^{-5} to 3.9×10^{-5} respectively for tested cigarette brands.

Keywords: Carcinogens; Liquid chromatography/Mass spectrometry; Respiratory health; ILCR; MPPD.

A Survey of VOC Emissions from Rendering Plants

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Source: *Volume 17, No. 1, January 2017, Pages 209-217 doi: 10.4209/aaqr.2016.09.0391*

Rendering is a global industry that recycles by-products resulting from butchering operations, which process billions of animals per year. About 50% of the weight of livestock is not consumed by humans and must be processed by rendering operations, which cook and separate the material into its protein and fat components. These products serve as a sustainable food source for livestock, feedstocks for oleochemicals, and raw material for biodiesel refineries. Due to the scale and nature of the raw materials and the cooking process, rendering operations emit a significant, but as yet poorly quantified, VOC load. Assessing this VOC load is important in order to calibrate the industry's contribution to global VOC emissions, and to help address nuisance odor problems. We conducted VOC air sampling of two facilities in California, USA during the winter and summer seasons. VOC and reduced sulfur analyses were conducted using 8 h ambient air samples. Analyses for amines, ammonia, aldehydes/ketones, and volatile fatty acids were conducted using sampling pumps. These analyses detected 43 compounds at the facilities, and the number and concentration of detectable compounds were seasonally dependent. The compounds present at the highest concentrations included: ammonia (1600–2800 ppb, i.e., winter–summer levels), acetic acid (80–320 ppb, along with twelve other fatty acids ranging from ~0.5–140 ppb), acetone (55–241 ppb, along with nine other aldehyde/ketone products ranging from 0.4–60 ppb), and ethanol (15–81 ppb). These constituents have low odor thresholds and thus contribute to nuisance odor problems. Further, the overall VOC contribution arising from rendering facilities on a global scale is as yet very poorly characterized. This analysis will be useful to guide the development of new odor abatement strategies and strategies for the reduction of VOC emissions associated with this critical industry.

Keywords: Rendering; VOCs; Sulfur; Amines; Volatile fatty acids; Carbonyls; Environmental sampling.

An Investigation into the Effects of Off-Shore Shipping Emissions on Coastal Black Carbon Concentrations

David M. Butterfield, Paul Quincey

Source: *Volume 17, No. 1, January 2017, Pages 218-229 doi: 10.4209/aaqr.2015.12.0688*

Shipping emissions are recognised as a significant but poorly understood contributor to air pollution in the UK. Away from port, lower grade fuels are permitted to be used, to the extent that sulphur emissions from shipping were forecast to exceed those from all land-based sources in the EU by 2020. To examine the impact of black carbon emissions from shipping, an Aethalometer was installed at Goonhilly Downs on the Lizard peninsula in Cornwall UK, which is typically downwind of busy shipping lanes, with very low background pollution levels, for a full year starting in November 2012. Black carbon and UV component concentrations were combined with local wind

speed and direction data to differentiate between shipping emissions, local sources and long-range transport. Black carbon concentrations were compared with PM_{2.5} mass concentrations from Plymouth Centre to allow for the influence of changes in regional background particulate concentrations. Black carbon concentrations showed no substantial increment above other rural UK locations, although some time periods show small elevated concentrations which could be attributed to emissions from the nearby shipping lanes. The long-term (annual) average contribution to black carbon concentrations from off-shore shipping is estimated to be less than 0.1 µg m⁻³, with the hourly peak values being in the range 1–2 µg m⁻³.

Keywords: Black carbon; Shipping emissions; Aethalometer; Goonhilly.

Association of Particulate Matter Impact on Prevalence of Chronic Obstructive Pulmonary Disease in Ahvaz, Southwest Iran during 2009–2013

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Source: Volume 17, No. 1, January 2017, Pages 230-237 doi: 10.4209/aaqr.2015.11.0628

Air pollutants produced in environments have many detrimental impacts on human health. Chronic obstructive pulmonary disease (COPD) is a common worldwide respiratory disease. The aim of this study was to estimate the association between the load of particulate matters and the prevalence of COPD in Ahvaz, southwest of Iran, during 2009–2013. This epidemiological and used-model study was performed in Ahvaz. Particulate matter equal or less than 10 micro meters (PM₁₀) was monitored by Ahvaz Environmental Protection Agency (AEPA). Sampling was performed hourly during the study period in 4 stations. In this study, 175200 (4 × 24 × 365 × 5) samples of air were taken and collected. Sampling and analysis were performed according to EPA guideline. We utilized the relative risk values and baseline incidence measures by the WHO (Middle East) drawn from Health Effects Association of Particulate Matter. Finally, prevalence of COPD attributed to particulate matter exposure was calculated by Air Q model. According to our findings, the prevalence of COPD attributed to particulate matters decreased during 2009–2013 and followed a decreasing trend. Accordingly, the yearly prevalence of COPD during the period 2009–2013 were 121, 111, 94, 102, and 98, and the yearly average PM₁₀ concentrations during the same period were 313.72, 281.98, 288.38, 278.12, and 242.29 µg m⁻³, respectively. Although the average of 5-year study was higher than WHO and NAAQS values, a descending trend for COPD indicates that the level of PM₁₀ was diminished from 2009 to 2013. Therefore mitigating air pollutant particularly PM₁₀ as one of the main hazards could be possibly led to remarkable decrease in the rate of mortality and morbidity particularly COPD attributed to PM₁₀.

Keywords: Chronic obstructive pulmonary disease; Prevalence; Particulate matter; Iran.

Daily Personal Exposure of Women Cooks to Respirable Particulate Matters during Cooking with Solid Bio-Fuels in a Rural Community of West Bengal, India

Sukanta Nayek, Pratap Kumar Padhy

Source: Volume 17, No. 1, January 2017, Pages 245-252 doi: 10.4209/aaqr.2016.01.0028

About 60% of the Indian population are still relying on the traditional bio-fuels comprising of firewood, cow dung cake, crop residue etc. More than 85% of the rural households use these unprocessed bio-fuels for cooking purposes. Biomass fuel burning in daily cooking with traditional inefficient earthen stoves, in an un-vented kitchen, usually emits very high levels of smoke containing a complex mixture of a wide array of potentially hazardous pollutants, especially, particulate matters of varying size range. Size smaller than the PM₁₀ particles, can penetrate further deep into the gas exchange region of the lungs and are termed as the respirable particulate matter with 50% cut point at 4 μm . Respirable particulate exposure assessment is particularly important in case of health hazard explanation because it can enter into the deepest of the lungs. Personal exposures to respirable particulate matters were assessed during cooking hours in the varied opened kitchens, considering the seasonal change and the meal preparation as covariates. Maximum variability for the particulate exposures was observed in the kitchens with openness range of 15–60%. Greater particulate exposures were found in the least opened kitchens. Two-way ANCOVA showed significant impact of seasonal change on the differential opened kitchens for personal exposure to respirable particulate matters. Tukey post hoc test reveals significant mean differences of respirable particulates in all pair-wise seasonal combinations and in all the pair-wise openness type combinations except for the < 15% and 15–30% opened kitchens. Winter season came out to be one of the significant predictor for the personal exposure prediction model. Across all the seasons and kitchen openness, average exposure concentration of the respirable particulate matters was 1445 $\mu\text{g m}^{-3}$.

Keywords: Bio-fuels; Personal exposure; Respirable particulate matters; Rural kitchen; Woman cook.

Evaluation of PM_{2.5} Surface Concentrations Simulated by Version 1 of NASA's MERRA Aerosol Reanalysis over Israel and Taiwan

Simon Provençal, Virginie Buchard, Arlindo M. da Silva, Richard Leduc, Nathalie Barrette, Emily Elhacham, Sheng-Hsiang Wang

Source: Volume 17, No. 1, January 2017, Pages 253-261 doi: 10.4209/aaqr.2016.04.0145

Version 1 of the NASA MERRA Aerosol Reanalysis (MERRAero) assimilates bias-corrected aerosol optical depth (AOD) data from MODIS-Terra and MODIS-Aqua, and simulates particulate matter (PM) concentration data to reproduce a consistent database of AOD and PM concentration around the world from 2002 to the end of 2015. The purpose of this paper is to evaluate MERRAero's

simulation of fine PM concentration against surface measurements in two regions of the world with relatively high levels of PM concentration but with profoundly different PM composition, those of Israel and Taiwan. Being surrounded by major deserts, Israel's PM load is characterized by a significant contribution of mineral dust, and secondary contributions of sea salt particles, given its proximity to the Mediterranean Sea, and sulfate particles originating from Israel's own urban activities and transported from Europe. Taiwan's PM load is composed primarily of anthropogenic particles (sulfate, nitrate and carbonaceous particles) locally produced or transported from China, with an additional contribution of springtime transport of mineral dust originating from Chinese and Mongolian deserts. The evaluation in Israel produced favorable results with MERRAero slightly overestimating measurements by 6% on average and reproducing an excellent year-to-year and seasonal fluctuation. The evaluation in Taiwan was less favorable with MERRAero underestimating measurements by 42% on average. Two likely reasons explain this discrepancy: emissions of anthropogenic PM and their precursors are largely uncertain in China, and MERRAero doesn't include nitrate particles in its simulation, a pollutant of predominately anthropogenic sources. MERRAero nevertheless simulates well the concentration of fine PM during the summer, when Taiwan is least affected by the advection of pollution from China.

Keywords: MERRAero; Evaluation; Fine particulate matter; Israel; Taiwan.

Performance Evaluation of the WRF-Chem Model with Different Physical Parameterization Schemes during an Extremely High PM_{2.5} Pollution Episode in Beijing

Dongsheng Chen, Xin Xie, Ying Zhou, Jianlei Lang, Tingting Xu, Nan Yang, Yuehua Zhao, Xiangxue Liu

Source: Volume 17, No. 1, January 2017, Pages 262-277 doi: 10.4209/aaqr.2015.10.0610

To understand the impacts of different combinations of planetary boundary layer (PBL), short-wave (SW) and long-wave (LW) radiation schemes on the simulation results of meteorological variables and PM_{2.5} concentrations under extremely heavy pollution conditions, the Weather Research and Forecasting model with Chemistry (WRF-Chem) model was applied in Beijing to investigate a high PM_{2.5} pollution episode that occurred in January, 2013. Four PBL schemes, two SW schemes and three LW schemes with a total of 12 ensemble experiments were conducted in this study. The simulated meteorological variables including the temperature at 2 m (T2), the wind speed at 10 m (WS10) and the relative humidity (RH) were compared with their actual observations and the PM_{2.5} concentrations. A correlation analysis between the PM_{2.5} and T2, WS10 and RH values was also explored. The results indicated that there were no ideal scheme combinations that were most suitable for all meteorological variable simulations during this heavy pollution episode in Beijing. With the same emissions input, the simulation results of the WRF-Chem model that were configured with different physical parameterization schemes may vary significantly. As for the PM_{2.5} simulation, the combination of the YSU PBL, Goddard SW and GFDL LW schemes showed the greatest consistency with the observed values. Although the PBL schemes have the dominant impacts on the simulations of meteorological variables, the selection of LW and SW schemes is of the same importance.

Keywords: Heavy haze pollution; Numerical simulation; Physical parameterization schemes.

Using Box Modeling to Determine Photodegradation Coefficients Describing the Removal of Gaseous Formaldehyde from Indoor Air

Ming-Wei Lin, Ching-Song Jwo, Hsin-Jr Ho, Liang-Yü Chen

Source: Volume 17, No. 1, January 2017, Pages 330-339 doi: 10.4209/aaqr.2016.09.0397

Human exposure to volatile organic compounds (VOCs) indoors is receiving increasing attention. Formaldehyde (HCHO) is the most common VOC emitted from household materials and is associated with many health risks, including sick building syndrome. In this study, a simple box model was developed and applied to help understand the fate and degradation mechanisms of HCHO in the indoor environment. The model was validated using observations from an air handling system under different conditions. A UV/TiO₂ filter reactor was installed in a closed box with the air conditioning unit. Three parameters, temperature, relative humidity, and circulation wind speed, were investigated for their effects on the performance of the air handling system. Our results show that the operation mode of the air handling system has a greater effect on the removal of HCHO than any of the air conditioning parameters. From a kinetic perspective, the removal of gaseous HCHO from a constant-volume box clearly represents a zero-order reaction. After UV irradiation with a TiO₂ filter for 2 hours, the removal efficiency of gaseous HCHO increases to approximately 90%. Contributions to the removal of gaseous HCHO from natural dissipation, photodegradation, and photocatalytic oxidation decomposition are 12%, 30%, and 58%, respectively. Our results have implications for reducing indoor air pollution and reducing stress on air conditioning systems. Meeting these goals is beneficial for human health and energy conservation in modern society.

Keywords: Heterogeneous catalysis; Titanium dioxide; Air cleaning; Reaction kinetics.

Development and Application of a New PM_{2.5} Source Apportionment Approach

Jianlei Lang, Shuiyuan Cheng, Wei Wen, Chao Liu, Gang Wang

Source: Volume 17, No. 1, January 2017, Pages 340-350 doi: 10.4209/aaqr.2015.10.0588

Due to the similarity of PM_{2.5} chemical species profiles of different sources, time synchronization of source contributions and the uncertainties of source-oriented models, it is difficult to get a well-separated and relatively accurate PM_{2.5} source apportionment result, especially for the secondary components, when only one method was applied. A new PM_{2.5} source apportionment approach, combining the receptor models, source-oriented models and emission inventory, was developed in this study. The proposed method had following strengths: (1) it could identify the source contributions to secondary components; (2) target (or expected) sources were optional; (3) mixed sources could be avoided. The new approach was then applied in two typical cities in North China –

Beijing and Tangshan, based on intensive PM_{2.5} observation results from 2011 to 2013. The source apportionment result indicated that the annual average contribution to PM_{2.5} in Tangshan was 7.4%, 21.5%, 7.6%, 18.0%, 14.5%, 10.9% and 20.0% for power, metallurgy, cement, coal combustion, vehicle, dust and other sources, respectively; the annual average contribution ratio for vehicle, industry and industrial coal combustion, residential coal combustion, dust and other sources in Beijing was 31.5%, 22.9%, 10.6%, 14.5% and 20.4%, respectively. Seasonal variation of the source contributions was also analyzed. The demonstration results showed that the combined method was feasible. In addition, the detailed source contribution results could also provide scientific support for making effective PM_{2.5} mitigation strategy.

Keywords: Source apportionment; Receptor model; WRF-CAMx-PSAT; Beijing; Tangshan.

Overview of the Special Issue "PM_{2.5} in Asia" for 2015 Asian Aerosol Conference

Rajasekhar Balasubramanian, Xiang Gao, Shiro Hatakeyama, Jungho Hwang, Chuen-Jinn Tsai

Source: Volume 17, No. 2, February 2017, Pages 351-355 doi: 10.4209/aaqr.2017.01.0042

The Asian Aerosol Conference (AAC), held every two years, is the most prestigious conference for aerosol researchers in Asian region. The 9th AAC (AAC 2015) was held by Japan Association of Aerosol Science and Technology (JAASST) on behalf of Asian Aerosol Research Assembly (AARA) from June 24 to 27, 2015 at Kanazawa, Japan. A total of 520 participants from 14 countries presented 430 papers, including 4 plenary lectures, 16 keynote speeches, 125 oral and 285 poster papers in the conference. There were 270 Japanese participants and 250 participants from abroad in which China (with 87 participants), Korea (with 77 participants) and Taiwan (with 57 participants) contributed to the most attendees. Among the papers presented in the conference, 'PM_{2.5} symposium', 'atmospheric aerosols' and 'urban air quality' sessions addressed many important issues related to PM_{2.5} and ambient aerosols in East Asian countries. The features of this conference is the inaugural presentation of six AARA fellows, bestowal of two Asian Young Aerosol Scientist Awards, and a half day symposium "Analysis of Aerosol by Young Asian Researchers" which was a great opportunity for the networking of young aerosol researchers in Asian region.

Keywords: 2015 Asian Aerosol Conference; AAC; Special issue.

Chemical Characteristics of PM_{2.5} during a 2016 Winter Haze Episode in Shijiazhuang, China

Fei Chen, Xiaohua Zhang, Xinsheng Zhu, Hui Zhang, Jixi Gao, Philip K. Hopke

Source: Volume 17, No. 2, February 2017, Pages 368-380 doi: 10.4209/aaqr.2016.06.0274

To better understand the chemical characteristics and the potential source regions of PM_{2.5} measured from 18 January until 22 January 2016 in Shijiazhuang, China, PM_{2.5} was measured continuously and integrated daily sampling using mid-volume samplers was conducted at the three sites. The mean concentration of PM_{2.5} at the three sites reached 113, 131 and 119 µg m⁻³ during the sampling period, the higher concentrations occurred at early morning and noon, similar variation trends were found in the three sites. The concentrations of OC were higher than EC at three sampling sites and the OC/EC ratios ranged from 9.09 to 12.4 with a daily mean value of 10.8 during a haze pollution episode (HPE), which suggested that carbonaceous compositions might be from same source. The total concentration of water soluble inorganic ions (WSII) at the sites ranged from 72.2 to 100.0 µg m⁻³ with a mean of 84.3 µg m⁻³. The dominant species were NO₃⁻, SO₄²⁻, NH₄⁺, Cl⁻, accounting for 88.4% of the mean PM_{2.5} WSII mass. The most abundant measured element was Na with average mass concentrations of 41.5, 37.0, and 38.1 µg m⁻³ during the HPE. Relative humidity during HPE was higher than during clean days with average values of 70.1% and 60.2%, suggesting that high relative humidity and low wind speed favored formation of secondary inorganic ions and accelerated hygroscopic growth. The Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) and a potential source contribution function (PSCF) analysis were used to assess the data. Back-trajectories for the three sites identified similar transport pathways. PSCF analysis showed a significant regional impact on PM_{2.5} at Shijiazhuang during the polluted period. The potential source areas for PM_{2.5} in Shijiazhuang were the Beijing-Tianjin region and Shandong Province. The results of the present study show the need for the development of PM_{2.5} control measurements on a regional scale.

Keywords: PM_{2.5}; Chemical composition; Meteorological factors; Backward trajectory; Potential source contribution function (PSCF).

Atmospheric Dispersion of PM_{2.5} Precursor Gases from Two Major Thermal Power Plants in Andhra Pradesh, India

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Source: Volume 17, No. 2, February 2017, Pages 381-393 doi: 10.4209/aaqr.2016.07.0294

Fine particulate matter (PM_{2.5}) predominantly comprises sulphates and nitrates, which results from sulphur dioxide (SO₂) and nitrogen oxide (NO_x) gases that are emanated from excessive industrial activities and transport systems. PM_{2.5} is known to affect respiratory health in humans. Coal-fired thermal power plants are a major source of SO₂ and NO_x gases. Evaluation of the dispersion characteristics of these precursor gases from the power plants would help understand the vulnerability. Meteorological conditions that prevail over the region would influence the dispersion characteristics. In this study, dispersion of SO₂ and NO from two major coal-fired thermal power plants in Andhra Pradesh, India have been studied using an integrated modeling approach of the Advanced Research Weather Research & Forecasting (ARW) model and Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model. Meteorological conditions are obtained at 3-km resolution using the ARW model and dispersions of SO₂ and NO is computed using the HYSPLIT model for the four seasons of winter, summer, monsoon and post-monsoon. Forward trajectories produced by the HYSPLIT model show diurnal variations and dispersion patterns show seasonal

variations indicating the influence of meteorological conditions. Dispersion characteristics show high dispersion in winter due to calm and stable atmospheric conditions to insignificant in summer season due to stronger winds and higher atmospheric instability. The study establishes the usefulness of integrated meteorological and dispersion models for the evaluation of pollutant dispersion.

Keywords: Particulate matter; Dispersion; Power plants; ARW model; HYSPLIT.

Extreme Events of Reactive Ambient Air Pollutants and their Distribution Pattern at Urban Hotspots

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Source: Volume 17, No. 2, February 2017, Pages 394-405 doi: 10.4209/aaqr.2016.06.0273

The occurrence of extreme events of air pollutant concentrations at urban hotspots is a routine phenomenon, particularly during the winter season. However, extreme events of reactive air pollutants are more frequent during the summer season. The assessment of air pollution extreme events will provide a platform to formulate an effective and efficient hotspot urban air quality management plan. The statistical distribution model (SDM) is widely used to describe the average as well as extreme air pollutant concentration in a more organized and efficient manner. In the present study, the best fit SDM has been evaluated for hourly average PM_{2.5} and NO₂ concentrations at one of the busiest traffic intersections in Delhi city (air pollution hotspot 1: APH-1) and for PM_{2.5} at one of the heavily trafficked road corridors in Chennai city (air pollution hotspot 2: APH - 2). The SDMs were developed for different seasons to evaluate the impacts of climatic conditions on the air pollution events. Results indicate that NO₂ concentrations were best fitted with lognormal and log logistic distribution models respectively, for winter and summer seasons at APH-1. However, lognormal distribution was best fitted to PM_{2.5} concentration of winter and summer seasons at both APHs.

Keywords: Extreme pollutant concentrations; Urban hotspot; Statistical distribution model; Goodness of fit test; Location and Scale parameters.

Chemical Characteristics of Particulate Matter Emission from a Heavy-Duty Diesel Engine Using ETC Cycle Dynamometer Test

Taosheng Jin, Kaibo Lu, Shuangxi Liu, Shuai Zhao, Liang Qu, Xiaohong Xu

Source: Volume 17, No. 2, February 2017, Pages 406-411 doi: 10.4209/aaqr.2016.06.0264

This paper presents chemical characteristics of diesel particulate matter (DPM). A heavy-duty diesel engine was tested in ETC cycle with an engine dynamometer. The DPM was sampled with quartz fiber membrane and organic membrane filter. Quartz fiber membrane was used for total carbon (TC) and particle-phase PAHs (p-PAHs) measurement, while the organic membrane was used for inorganic elements measurement. On the quartz fiber membrane, the total of 15 elements detected were 11% of DPM mass. Ca, Si, Na and Al were the major components, accounted for 79% of the 15 elements mass. On the organic membrane, total carbon (elemental carbon + organic carbon) was 90% of total DPM mass. Three-ring p-PAHs were the major components (66%) of total p-PAHs mass followed by four-ring (18%) and two-ring (16%). The FLT/(FLT + PYR) ratio was 0.62, indicative of diesel vehicle emissions.

Keywords: Diesel particulate matter (DPM); Inorganic elements; Total carbon (TC); Polycyclic aromatic hydrocarbons (PAHs).

Trends of PM_{2.5} and Chemical Composition in Beijing, 2000–2015

Jianlei Lang, Yanyun Zhang, Ying Zhou, Shuiyuan Cheng, Dongsheng Chen, Xiurui Guo, Sha Chen, Xiaoxin Li, Xiaofan Xing, Haiyan Wang

Source: Volume 17, No. 2, February 2017, Pages 412-425 doi: 10.4209/aaqr.2016.07.0307

PM_{2.5} is the major pollutant in most cities of China, especially contributed significantly to the poor air quality in Beijing. This study aimed to investigate the long-term trend (2000–2015) of PM_{2.5}, based on intensive observation and comprehensive literature investigation of PM_{2.5} and its chemical components. Results showed that the annual average concentration of PM_{2.5} generally decreased by 1.5 $\mu\text{g m}^{-3} \text{ year}^{-1}$ from 2000 to 2015 under the implementation of 16 phases' air pollution control measures. In the most polluted season (winter), four change stages were found related with emission control effect and meteorological conditions: PM_{2.5} decreased in 2000–2008 and 2010–2013, increased in 2008–2010, and was at a high level in 2013–2015. As for detailed chemical components, OC, soil dust and typical elements (Si, Ca, Fe, Mn, Cu, Pb and K) presented a downward trend generally. EC had almost no change before 2003, increased from 2003 to 2007, but decreased after 2007; this may be caused by the replacement of fossil fuel and control of biomass emission. The continuous rising of OC/EC and SOC/OC (secondary organic carbon, SOC) in recent years illustrated the secondary carbonaceous species pollution is becoming serious. SO_4^{2-} showed a slight increase from 2000–2013, but decreased obviously from 2013–2015. NO_3^- and NH_4^+ had an upward trend during the past decade. The proportion of secondary inorganic aerosol (SIA) increased at a rate of 0.7% yr^{-1} , and it has become the major composition of PM_{2.5} in Beijing instead of carbonaceous component since 2009. In general, PM_{2.5} change trend indicated the emission mitigation measures implemented in Beijing have reduced the primary PM_{2.5} effectively. However, the control of secondary components should be paid special attention in order to further improve the air quality in Beijing effectively.

Keywords: Fine particle; Chemical components; Long-term trends; Beijing; Emission control effect.

Development of an Automated System (PPWD/PILS) for Studying PM_{2.5} Water-Soluble Ions and Precursor Gases: Field Measurements in Two Cities, Taiwan

Ziyi Li, Yingshu Liu, Yujie Lin, Sneha Gautam, Hui-Chuan Kuo, Chuen-Jinn Tsai, Huajun Yeh, Wei Huang, Shih-Wei Li, Guo-Jei Wu

Source: Volume 17, No. 2, February 2017, Pages 426-443 doi: 10.4209/aaqr.2016.10.0440

An automated system consisting of a particle-into-liquid sampler (PILS) and a parallel plate wet denuder (PPWD) coupled with an ion chromatography was used for simultaneous measurement of ambient water-soluble ions in PM_{2.5} and precursor gases. The performance of the PPWD/PILS was validated by comparing it with the PDS (porous metal denuder sampler) for precursor gases (NH₃, HONO, HNO₃ and SO₂) and PM_{2.5} ionic species (NH₄⁺, NO₃⁻, SO₄²⁻, Na⁺, Cl⁻ and K⁺) measured in Taipei and Hsinchu Cities of Taiwan. Good correlations were demonstrated with linear regression slopes ranging from 0.92 to 1.04 and 0.84 to 0.97 as well as R² ranging from 0.76 to 0.83 and 0.89 to 0.94, for precursor gases and PM_{2.5} ions, respectively. The accuracy of the current system for precursor gases outperforms the other commercial systems. Field continuous data showed that NH₃ was the most abundant precursor gas with the diurnal pattern peaking at low nocturnal boundary heights and during rush hours with local traffic emissions in Taipei, and with the pattern peaking only at mid-day associated with regional sources in Hsinchu. A reverse diurnal pattern for HONO in Taipei reflected the daytime photolysis and its nocturnal heterogeneous reaction, while its concentration was relatively constant at very low level in Hsinchu. SO₄²⁻, NH₄⁺ and NO₃⁻ exhibited very similar diurnal patterns with the mean concentrations of 4.56 ± 3.14, 1.55 ± 1.16 and 0.52 ± 0.5 µg m⁻³ in Taipei, and 7.95 ± 5.52, 2.41 ± 1.95 and 0.96 ± 1.10 µg m⁻³ in Hsinchu, respectively. Correspondingly high concentrations of major ions to precursor gases were associated with the photochemical secondary aerosol formations and heavy traffic in Taipei. Based on an ammonia-rich atmosphere and high SOR values, (NH₄)₂SO₄ and NH₄NO₃ were inferred to be the dominant inorganic salts in PM_{2.5} at both sites, which were also verified by the ion balance analysis.

Keywords: PM_{2.5}; Automated measurement system PPWD/PILS; Inorganic ions; Precursor gases; Seasonal variation; Diurnal variation.

Continuous Observation of the Mass and Chemical Composition of PM_{2.5} using an Automatic Analyzer in Kumamoto, Japan

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Source: Volume 17, No. 2, February 2017, Pages 444-452 doi: 10.4209/aaqr.2016.07.0290

Due to economic growth in China, emissions of gaseous components from factories and automobiles have been increasing, which has resulted in severe air pollution. During the winter and spring seasons, Japan, which is on the leeward side of the Asian continent, is on the receiving end of

this increasingly problematic transboundary air pollution. In this study, the mass concentration and chemical components of the particulate PM_{2.5} were continuously observed using an automatic analyzer at Kumamoto on the west coast of Japan from October 2014 to March 2015. A greater number of high PM_{2.5} days were observed in winter than in autumn. This seasonal change in concentrations was believed to be due to transboundary air pollution traveling from the Asian continent due to seasonal monsoons. The analysis of the chemical composition of PM_{2.5} supported this idea. The factors leading to high PM_{2.5} concentrations were investigated and categorized into transboundary air pollution, local air pollution, and volcanic activity based on the analysis of sulfate (SO₄²⁻) and sulfur dioxide (SO₂) concentrations and model simulations. The average concentration of chemical components showed that local air pollution also influenced air quality in Kumamoto.

Keywords: ACSA; Source apportionment; Transboundary air pollution; Volcanic activity; Local air pollution.

Analysing PM_{2.5} and its Association with PM₁₀ and Meteorology in the Arid Climate of Makkah, Saudi Arabia

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Source: Volume 17, No. 2, February 2017, Pages 453-464 doi: 10.4209/aaqr.2016.03.0117

Atmospheric Particulate Matter (PM) is considered one of the most critical air pollutants in terms of its detrimental health impacts, environmental degradations and visibility. Particles size, their chemical composition and atmospheric levels are important factors for determining their adverse health impacts. In this paper various aspects of PM_{2.5} are analysed including PM_{2.5}/PM₁₀ ratios and association with meteorological parameters using data collected from January 2014 to September 2015 in Makkah Saudi Arabia. During the study period, mean PM_{2.5}/PM₁₀ ratio was found to be 0.64, whereas median and maximum ratios were 0.69 and 0.99, respectively. Diurnal, weekly and annual cycles of PM₁₀, PM_{2.5} and their ratios were analysed, which demonstrated considerable variations during various hours of the day, days of the week and months of the year. PM_{2.5}/PM₁₀ ratios were lower in summer (June and July) and higher in winter (November and December), likewise the ratios were lower during afternoon and higher in the morning and evening. As expected, there was a positive correlation between PM₁₀ and PM_{2.5} ($r = 0.51$) and both PM₁₀ and PM_{2.5} showed negative association with relative humidity and positive with wind speed and temperature. Furthermore, PM_{2.5}/PM₁₀ ratios were lower (< 0.45) at lower relative humidity ($< 16\%$) and higher (> 0.70) at higher relative humidity (35–90%), indicating a shift towards high PM_{2.5} concentrations at higher relative humidity. Polar plots showed lowest ratios at high wind speed ($> 3 \text{ m s}^{-1}$) blowing from west and southwest direction in summer, and highest ratios at low wind speed ($< 2 \text{ m s}^{-1}$) in winter. Polar plots were successfully applied to show the interaction between various meteorological parameters and PM_{2.5}/PM₁₀ ratios. Further work on source apportionment and receptor modelling of PM is required to help develop air quality index and prepare an effective air quality plan for Makkah.

Keywords: PM₁₀; PM_{2.5}; Particulate matter; Air quality; Makkah Saudi Arabia.

Trajectory-Based Models and Remote Sensing for Biomass Burning Assessment in Bangladesh

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Source: Volume 17, No. 2, February 2017, Pages 465-475 doi: 10.4209/aaqr.2016.07.0304

Biomass burning is a major global source of fine primary carbonaceous particles including strongly light absorbing compounds and marker compounds. In a prior study, particulate matter (PM) sampling was conducted during 2010–2012 period at sites in Rajshahi, Dhaka, Khulna, and Chittagong. PM samples were collected using dichotomous samplers in the PM_{2.5} and PM_{2.5–10} size fractions. The samples were analyzed for mass, black carbon at 370 nm (UVBC) and 880 nm (BC), Delta-C (UVBC-BC), and elemental compositions with X-ray fluorescence. Source apportionment using PMF was performed to identify and quantify the PM sources. Results showed that biomass burning contributions during winters in Rajshahi were substantially higher than in Dhaka, Khulna, or Chittagong. Agricultural burning areas of the Indo-Gangetic Plain were hypothesized as the primary source region. The present study explores the relationships between the source regions using trajectory ensemble models and determines if transported biomass PM has disproportionately affected air quality in Rajshahi. The probable source locations that were identified included Pakistan, northern India, Nepal, Bangladesh, Northeastern India, and Myanmar. To assess the model results, satellite measurements of fire radiative power (FRP) were calculated based on fire data acquired by the MODerate-resolution Imaging Spectro-radiometer (MODIS) sensor in six defined areas. High fire occurrences from MODIS coincident with the source regions identified in Nepal, Northeastern India and Myanmar in winter. The instantaneous FRP values ranged between 4.4 MW and 2449 MW. The mean winter FRP values for Nepal and Northeastern India were higher than for the other regions with Nepal having the overall highest value. Fire locations with their mean power, NASA Satellite pictures and particles speed along trajectories have been analyzed. In summary, the integrated outcome of the different techniques has identified Northern India and Nepal as the main source area responsible for the increased biomass burning concentration difference at Rajshahi.

Keywords: Biomass burning; PM_{2.5}; Trajectory ensemble models; Fire radiative power (FRP); MODIS.

Identification of Sources of Fine Particulate Matter in Kandy, Sri Lanka

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Source: Volume 17, No. 2, February 2017, Pages 476-484) doi: 10.4209/aaqr.2016.03.0123

Kandy is the second largest city in Sri Lanka and a major tourist destination. It is a fast growing city with continuous construction of buildings, roads and historical places. More than 100 samples of fine particulate matter (PM) were collected using a GENT stacked filter sampler from a fixed site at

the regional sampling station of Department of Meteorology situated in Katugastota, Kandy over the period of 2012 to 2014. Black carbon (BC) in these filters were determined by reflectance measurements while their elemental compositions were determined using the X-ray fluorescence spectrometry. Analysis of the elemental data suggests that the PM in Kandy originates largely from re-suspended soil and anthropogenic sources. The fine particulate matter data including BC and major elements (Na, Mg, Al, Si, Cl, Fe, Zn, Ni, Cu, V, S, Br, Pb, Cr, K, Ca and Ti) was analyzed using EPA-PMF version 5.0 (Positive Matrix Factorization) to explore the possible sources of the PM at the study site. Five factors were found and identified as soil, aged sea salt, vehicular emissions, biomass burning, and industrial sources.

Keywords: Particulate matter; Source apportionment; Positive Matrix Factorization; Sri Lanka.

Low Molecular Weight Monocarboxylic Acids in PM_{2.5} and PM₁₀: Quantification, Seasonal Variation and Source Apportionment

Nidhi Verma, Aparna Satsangi, Anita Lakhani, K. Maharaj Kumari

Source: Volume 17, No. 2, February 2017, Pages 485-498 doi: 10.4209/aaqr.2016.05.0183

PM_{2.5} and PM₁₀ aerosols from a semi-urban site of Agra (27°10'N, 78°05'E) in North-Central India were analyzed for carbonaceous aerosols (Organic and Elemental carbon), low molecular weight monocarboxylic acids (Acetic and Formic acid) along with inorganic ions (Cl⁻, NO₃⁻, SO₄²⁻, K⁺ and Ca²⁺) during April 2014 to August 2015. The average PM_{2.5} and PM₁₀ mass concentrations were 86.3 ± 71.3 and 169.7 ± 100.5 µg m⁻³, respectively; about 45% of PM_{2.5} and 67% of PM₁₀ samples were above NAAQ (National Ambient Air Quality) standards. The average organic carbon (OC) and elemental carbon (EC) concentrations were 18.2 ± 12.3 and 6.7 ± 4.5 µg m⁻³, respectively in PM_{2.5} and 25.2 ± 14.1 and 8.1 ± 5.9 µg m⁻³ respectively in PM₁₀. The average concentration of acetic acid (AA) in PM_{2.5} and PM₁₀ were 330 ± 211 and 392 ± 224 ng m⁻³ respectively. The average concentration of formic acid (FA) in PM_{2.5} and PM₁₀ were 348 ± 193 and 336 ± 175 ng m⁻³ respectively. Formic acid concentration was higher in PM_{2.5} than PM₁₀ but the difference is not statistically significant. Both AA and FA showed similar seasonal variation: winter > post-monsoon > summer > monsoon. Low temperature and high relative humidity in winter season favours gas to particle conversion resulting in high concentrations. The average FA to AA (F/A) ratio was 0.69 indicating dominance of primary sources at the study site. Correlation analysis of AA and FA with major ions (Cl⁻, NO₃⁻, SO₄²⁻, K⁺ and Ca²⁺), EC, secondary organic carbon and trace gases (O₃ and CO) was performed to identify their primary or secondary origin. The results of correlation analysis suggest that AA is mainly contributed by primary sources while FA originates from secondary sources.

Keywords: Carboxylic acid; PM_{2.5}; PM₁₀; Primary sources; Secondary sources.

Characteristics of PM_{2.5} and Assessing Effects of Emission-Reduction Measures in the Heavy Polluted City of Shijiazhuang, before, during, and after the Ceremonial Parade 2015

Gang Wang, Shuiyuan Cheng, Jianlei Lang, Xiaowen Yang, Xiaoqi Wang, Guolei Chen, Xiaoyu Liu, Hanyu Zhang

Source: Volume 17, No. 2, February 2017, Pages 499-512 doi: 10.4209/aaqr.2016.05.0181

The measurement of PM_{2.5} was conducted from 15th August to 17th September, 2015 in Shijiazhuang, China, covering the period of a ceremonial parade. The PM_{2.5} concentrations and the major chemical components were analyzed. The concentrations of PM_{2.5} was 26.5 µg m⁻³ during control, which were 57.0% and 51.1% lower compared to before and after control, respectively. The lowest concentrations of elements and water-soluble ions were also found during control with a decreasing trend of 31.1%–44.2%, and 57.1%–64.2%, respectively. Two typical pollution episodes characterized by significantly elevated PM_{2.5} concentration were found during no control due to the combination of no emission-reduction measures and unfavorable weather conditions. The mass percentage of secondary inorganic ions was larger during no control (38.1%–40.3%), pointing to the strong contribution of atmospheric chemical processes. The NO₃⁻/SO₄²⁻ ratios were 0.85, 0.94, and 0.85 before, during, and after control, respectively, and the elevated ratios during control indicate a greater proportion of the PM_{2.5} originated from vehicle exhaust. The WRF-CMAQ modeling system was also used to assess the effectiveness of emission reduction measures and weather conditions. The results indicated that the PM_{2.5} concentration increased by 21.6% and 32.1% if no emission-reduction measures were taken and weather conditions in 2014 were used.

Keywords: PM_{2.5} pollution; Element; Water-soluble ions; Emission control assessment; Simulation modeling.

A Study of Characteristics and Origins of Haze Pollution in Zhengzhou, China, Based on Observations and Hybrid Receptor Models

Si Wang, Shaocai Yu, Pengfei Li, Liqiang Wang, Khalid Mehmood, Weiping Liu, Renchang Yan, Xianjue Zheng

Source: Volume 17, No. 2, February 2017, Pages 513-528 doi: 10.4209/aaqr.2016.06.0238

To obtain a comprehensive picture of characteristics and sources of haze pollution in Zhengzhou, we analyzed annual air pollutant (fine particulate matter (PM_{2.5}), inhalable particulate matter (PM₁₀), carbon monoxide (CO), nitrogen dioxide (NO₂), sulfur dioxide (SO₂) and ozone (O₃)) observations at nine monitoring stations from March 1, 2013 to February 28, 2014. A case study on haze pollution was carried out using observations, meteorological data, aerosol optical depth (AOD) values and Hybrid receptor models. Results of annual variations of air pollutants indicated that

PM_{2.5} pollution in Zhengzhou was the most severe. Monthly variations revealed that all air pollutants except O₃ showed peak values in December because of the increased local emissions during heating, while the lowest value found in August was probably because of the favorable dispersion conditions. The monthly variation patterns of O₃ concentrations show the peak values in August due to higher temperature and stronger solar radiation. The diurnal variations showed that PM_{2.5} concentration variations were consistent with the traffic flow. The high values of PM_{2.5}/PM₁₀ and PM_{2.5}/CO occurred in the afternoon probably due to the strong photochemical reactions. Results of the case study showed that relative humidity and wind speed were the main meteorological factors influencing PM_{2.5} concentrations. Back trajectories show that regional transport from the northeast and southeast of Zhengzhou (such as Puyang, Kaifeng, Zhoukou, and Xuchang in Henan province) also made a big contribution to the PM_{2.5} pollution in Zhengzhou. Our results demonstrated that the spatial-temporal distributions of PM_{2.5} in Zhengzhou were determined by complex factors such as primary emissions, secondary production, meteorological conditions and local/regional-transport.

Keywords: Haze; Primary emissions; Meteorological factors; Potential sources.

Chemical Fingerprint and Source Identification of Atmospheric Fine Particles Sampled at Three Environments at the Tip of Southern Taiwan

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James J. Lee

Source: Volume 17, No. 2, February 2017, Pages 529-542 doi: 10.4209/aaqr.2016.03.0102

The spatiotemporal variation, chemical fingerprints, transportation routes, and source apportionment of atmospheric fine particles (PM_{2.5}) along the coastal region of southern Taiwan were investigated at three environments in the tip of southern Taiwan. Three representative sampling sites at Chien-Chin (urban site), Siao-Gang (industrial site) and Che-Cheng (background site) were selected for simultaneous PM_{2.5} sampling from December 2014 to May 2015. Regular sampling of 24-h PM_{2.5} was conducted for continuous 6–9 days in each month. After sampling, the chemical composition, including water-soluble ions, metallic elements and the carbonaceous content of PM_{2.5}, was further analyzed within two weeks. The levoglucosan concentration was further compared to OC and K⁺ in PM_{2.5} originating from biomass burning. Moreover, the potential sources of PM_{2.5} and their respective contribution were further resolved by backward trajectory simulation, combined with chemical mass balance (CMB) receptor modeling. The field sampling results indicated that the PM_{2.5} concentrations at the urban and industrial sites were always higher than those at the background site. The most abundant water-soluble ionic species of PM_{2.5} are SO₄²⁻, NO₃⁻ and NH₄⁺, implying that PM_{2.5} is mainly composed of secondary ammonium sulfate and ammonium nitrate. The most abundant metallic elements of PM_{2.5} included crustal elements (Al, Fe and Ca) and anthropogenic (generated by humans) elements (V, Ni, As, Cd, Zn and Pb). Moreover, the concentrations of OC and EC at the Chien-Chin and Siao-Gang sites were generally higher than those at the Che-Cheng site, mainly due to the emissions from urban and industrial anthropogenic sources. Vehicular exhausts and industrial emissions were the main sources of PM_{2.5} at the Chien-Chin and Siao-Gang sites, respectively, while biomass burning and soil dusts were the dominant sources of PM_{2.5} at the Che-Cheng site.

Keywords: Atmospheric fine particles (PM_{2.5}); Chemical fingerprint; Spatiotemporal variation; Source identification.

Impact of Air Humidity Fluctuation on the Rise of PM Mass Concentration Based on the High-Resolution Monitoring Data

Liyuan Zhang, Yan Cheng, Yue Zhang, Yuanping He, Zhaolin Gu¹, Chuck Yu

Source: Volume 17, No. 2, February 2017, Pages 543-552 doi: 10.4209/aaqr.2016.07.0296

Hourly particulate matter mass concentrations and meteorological parameters, recorded by the China National Environmental Monitoring Center (CNEMC) and China Observatory, between June 2013 and March 2016 in Beijing, Xi'an, Shanghai and Guangzhou were examined to explore correlations. Characteristics of a rapid (abrupt) rise in PM_{2.5} mass concentration during early stage of a serious urban PM pollution event were examined and compared with pollution events with a gradual (accumulative) rise in PM_{2.5} mass concentration. The accumulative rise in air pollution is characterized by a prolonged slow PM_{2.5} growth rate (3–5 $\mu\text{g m}^{-3} \text{h}^{-1}$), and could eventually lead to middle level pollution (ambient PM_{2.5} mass concentration of about 150 $\mu\text{g m}^{-3}$), accompanied with an uncertain temporal variation in SO₂, NO₂, O₃ and CO concentrations. The abrupt rise process is characterized by a short-term high aerosol growth rate (> 10 $\mu\text{g m}^{-3} \text{h}^{-1}$), and could eventually form severe air pollution (PM_{2.5} mass concentration exceeds 250 $\mu\text{g m}^{-3}$), with a constant increase in gaseous pollutants concentrations. The average relative humidity (RH) was observed to have a less impact on the rise of PM_{2.5} concentration, but the fluctuation in RH was found to have a strong correlation with the rise in PM_{2.5} concentration. Further analysis has indicated that both abrupt and accumulative rise in PM_{2.5} mass concentration could be due to the RH fluctuation in atmosphere. The RH variation is important for the study of the fine-particle growth and for prediction of PM pollution episodes.

Keywords: Relative humidity (RH); Fluctuation of RH; Aerosol concentration; PM abrupt rise; PM accumulative rise.

Size Specific Distribution Analysis of Perfluoroalkyl Substances in Atmospheric Particulate Matter – Development of a Sampling Method and their Concentration in Meeting Room/Ambient Atmosphere

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Source: Volume 17, No. 2, February 2017, Pages 553-562 doi: 10.4209/aaqr.2016.07.0292

The international regulation of persistent organic pollutants (POPs) according to the Stockholm convention started in May 2001, and is intended to regulate the production and use of hazardous chemicals on a global scale. PFOS is one of the newly listed emerging POPs and only one of a diverse

huge group of perfluoroalkyl substances (PFASs), which are known as a “super set” of chemical tracers that include more than ninety related chemicals. The comprehensive monitoring of PFASs is necessary to develop a reliable understanding of environmental kinetics related to these pollutants. However, the extent of atmospheric pollution by PFASs is still unclear because their distribution and sources are not fully understood. Hence, a reliable analytical method for precisely measuring the levels of PFASs in particulate matter is needed. In this study, in order to investigate the levels of PFASs in atmospheric particles including PM_{2.5}, the use of new sampling equipment was evaluated by obtaining multiple samples of air from a stable meeting room environment. Meanwhile, by simultaneously obtaining samples from a roadside environment, the characteristics of PFASs from two different types of air samples were compared.

Keywords: POPs; PFASs; Particulate aerosol; PM_{2.5}.

Characteristics, Sources, and Health Risks of Atmospheric PM_{2.5}-Bound Polycyclic Aromatic Hydrocarbons in Hsinchu, Taiwan

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Source: Volume 17, No. 2, February 2017, Pages 563-573 doi: 10.4209/aaqr.2016.06.0283

This study investigated PM_{2.5}-bound polycyclic aromatic hydrocarbons (PAHs) in order to determine the seasonal changes in total benzo[a]pyrene equivalent (BaPeq) concentrations and to identify contamination sources by using a positive matrix factorization model, a conditional probability function, and characteristic ratios of PAHs in Hsinchu. The sampling period was from September 2014 to August 2015. PM_{2.5} samplers equipped with 47-mm quartz membrane filters were operated at a flow rate of 16.7 L min⁻¹ for 48 h. The concentrations of 20 PAHs were determined through gas chromatography–mass spectrometry. The results revealed the PM_{2.5}, total PAHs, and BaPeq mass concentrations in the four seasons ranged from 4.91 to 58.5 µg m⁻³, 0.21 to 8.08 ng m⁻³, and 0.03 to 0.78 ng m⁻³, respectively. The PM_{2.5}, total PAHs, and BaPeq mass concentrations were in the order winter > autumn > spring > summer and exhibited significant seasonal variations. The carcinogenic potency of PAHs in winter was approximately 6.21 times higher than that in summer. The major BaPeq contributors were BaP, BbF, INP, and DBA. BaP accounted for 49.0% of BaPeq concentrations in PM_{2.5} in all four seasons. The annual average lifetime excess cancer risk of PM_{2.5}-bound PAHs (1.60×10^{-5}) was higher than that specified in the United States Environmental Protection Agency guidelines (10^{-6}). The two major sources were stationary emission sources and unburned petroleum and traffic emissions, which together accounted for 90.3% of PM_{2.5}-bound PAHs.

Keywords: PM_{2.5}; Polycyclic aromatic hydrocarbon; Total benzo[a]pyrene equivalent; Hsinchu.

Comparison of Oxidative Abilities of PM_{2.5} Collected at Traffic and Residential Sites in Japan. Contribution of Transition Metals and Primary and Secondary Aerosols

Yuji Fujitani, Akiko Furuyama, Kiyoshi Tanabe, Seishiro Hirano

Source: Volume 17, No. 2, February 2017, Pages 574-587 doi: 10.4209/aaqr.2016.07.0291

Filter environmental samples of PM_{2.5} were collected at a traffic intersection in Kawasaki, Japan, and at a residential site (Tsukuba, Japan) in summer and winter, and the chemical compositions of the samples and their oxidative abilities in the dithiothreitol (DTT) assay were determined. Laboratory-generated aerosols (diesel exhaust particles [DEPs], gasoline direct injection spark ignition particles, and secondary organic aerosols [SOAs] generated from various precursors) were also investigated. To assess the effects of transition metals in the filter samples, we also conducted DTT assays on solutions of metal compounds similarly to the filter samples. In addition, the samples were pretreated with chelating reagents to mask the effects of transition metals. The DTT consumption average values for the filter samples collected at the traffic site were 53 and 50 pmol min⁻¹ µg⁻¹ in summer and winter, respectively, and these values were 1.3 and 1.1 times the corresponding values at the residential site and were also higher than the values for the laboratory-generated aerosols. Transition metals (Cu, Fe, Ni, and Mn) in the environmental samples were considered to be major contributors to DTT consumption (more than 80%). After removal of the effect of these transition metals with the chelating reagents, the oxidative abilities of the environmental samples were correlated with the amounts of organic carbon, water-soluble organic carbon, and organic acids and were weakly correlated with the amounts of elemental carbon and inorganic ions. We also found that the oxidative abilities based on the amount of organic carbon after removal of the effects of transition metals for DEPs, photochemically generated SOAs, and environmental samples except in the case of the traffic site in summer were compatible.

Keywords: Dithiothreitol assay; Oxidative activity, Traffic intersection; Primary aerosol; Secondary organic aerosol.

Significance of PM_{2.5} Air Quality at the Indian Capital

Shovan Kumar Sahu, Sri Harsha Kota.

Source: Volume 17, No. 2, February 2017, Pages 588-597 doi: 10.4209/aaqr.2016.06.0262

In New Delhi, the capital city of India, concentrations of regulated air pollutants often exceed the Indian national ambient air quality standards (INAAQS). As the sources of these pollutants differ, it is of utmost priority to understand the most dangerous air pollutant to formulate better control strategies in the city. In this study, regulated air pollutant concentrations in New Delhi during 2011 to 2014 were collected. Compared to other pollutants, PM_{2.5} concentrations exceeded the INAAQS quite often. While PM_{2.5} exceeded INAAQS during 85% of the days, NO₂, O₃, CO and SO₂ exceeded only on 37, 14, 11 and 0% of the days, respectively. Using air quality index approach, the most dominant pollutant was identified as PM_{2.5}, for 75 to 90% of the days. However, a seasonal variation

in the percentage dominance of PM_{2.5} was observed. For example, PM_{2.5} was dominant during 95% of the winter and 68% of monsoon days. In addition to absolute concentrations, pollutants can also be ranked by studying their associated short term mortality impacts. However, such studies are rare in India. For the first time, the short term impact of PM_{2.5} concentrations on non-disease specific mortality in New Delhi was assessed using Poisson regression models. Results indicated that the excessive risk associated with PM_{2.5} estimated was 0.57, which was higher than the other regulated pollutants. This indicates a projected 6.2 and 6.5% decrease in mortality by meeting the PM_{2.5} Indian standards and WHO set limits, respectively.

Keywords: Air quality index; New Delhi; PM_{2.5}; Health impact assessment.

Infiltration of Ambient PM_{2.5} through Building Envelope in Apartment Housing Units in Korea

Dong Hee Choi, Dong Hwa Kang

Source: Volume 17, No. 2, February 2017, Pages 598-607 doi: 10.4209/aaqr.2016.06.0287

Air pollution due to PM_{2.5} is of public concern in Korea. Ambient PM_{2.5} can penetrate indoors through the building envelope, affecting the indoor PM_{2.5} concentrations. Most people stay indoors for approximately 80% of every day, implying that their primary exposure to PM_{2.5} could be determined by the indoor air. This study aims to investigate the infiltration of ambient PM_{2.5} through the building envelope in apartment housing units in Korea. The on-site infiltration test method, by using a blower-door depressurization procedure, was suggested in order to maintain an identical indoor-outdoor pressure difference among the tests. On-site experiments were conducted in 11 apartment housing units to estimate the PM_{2.5} infiltration factors. The results showed that the average infiltration factor of all the test housing units was 0.65 ± 0.13 (average \pm standard deviation), with a minimum of 0.38 and a maximum of 0.88. Furthermore, the results from the relation of the building airtightness data to the infiltration factors suggests that a leaky housing unit with high ACH50, or a high specific effective leakage area (ELA), would be more significantly influenced by the ambient PM_{2.5}. The study demonstrated that the suggested infiltration test procedure was useful to assess the infiltration factors in conditions of controlled indoor-outdoor pressure differences in real housing units.

Keywords: PM_{2.5}; Infiltration; Building envelope; Apartment housing unit; Indoor air quality; Blower-door testing.

Characteristics of Exposure to Particles due to Incense Burning inside Temples in Kanpur, India

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Source: Volume 17, No. 2, February 2017, Pages 608-615 doi: 10.4209/aaqr.2016.04.0146

Incense burning in temples is a common and popular ritual in India and other Asian countries. This study aims at assessing the quantity and size segregated distribution of particulate matter in temples of Kanpur city, India. Active air sampling was performed in three temples using the Micro Orifice Uniform Deposit Impactor (MOUDI). PM₁₀ mass concentrations as high as 2184 µg m⁻³ were recorded inside the temples. Mass concentration values for all samples exceeded the Central Pollution Control Board (CPCB) National Ambient Air Quality Standard (NAAQS) of 100 µg m⁻³. Particle counts were high, and more than 99% of the numbers of particles generated were PM_{2.5}. Particle coagulation is an active mechanism leading to formation of polydispersed particles. Most abundant particles occur in the accumulation mode (dp < 1 µm). Ventilation conditions and amount of incense burned are major factors affecting particle size distribution. Exposure through inhalation, to critically high concentrations of fine particulate matter generated via incense smoke especially for worshippers who frequent the temple and temple workers, raises health concerns. The results clearly indicate that incense smoke is a major source of particulate matter in the temple microenvironment and their chemical characteristics need further examination.

Keywords: Indoor air quality; Incense burning; MMAD; Most abundant PM size.

Source Apportionment of PM_{2.5} Particles: Influence of Outdoor Particles on Indoor Environment of Schools Using Chemical Mass Balance

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Source: Volume 17, No. 2, February 2017, Pages 616-625 doi: 10.4209/aaqr.2016.07.0297

Children have higher lung function than adults and they spend most of their day time in schools. Also, children studying at schools located in the vicinity of busy roadways are vulnerable to childhood asthma and respiratory disorders. The present study is focused on estimating the sources of PM_{2.5} particles present in the indoor air quality in schools which are located adjacent to urban and suburban roadways. The indoor PM_{2.5} samples from all the four schools were collected using fine dust sampler from 8 a.m. to 4 p.m. The sampling was carried out for one complete week during various seasons including both working and non-working days. The chemical compositions of the PM_{2.5} samples were analyzed for certain elements like Ba, Cd, Cr, Cu, Fe, Mn, Ni, Pb, Sr, Ti, V and Zn using Inductively Coupled Plasma Optical Emission Spectrometry (ICP OES) and ions like F⁻, Cl⁻, NO₃⁻, PO₄³⁻, SO₄²⁻, K⁺, Ca²⁺, Mg²⁺, NH₄⁺, Na⁺ using Ion Chromatography (IC). Source apportionment study using Chemical Mass Balance was carried out using the species concentration of the collected samples. The major sources were found to be Paved Road Dust, Soil Dust, Gasoline Vehicle Emissions, Diesel Vehicle Emissions and Marine Source Emissions. Among these, vehicular emissions contribution was found to be higher for the schools located close to roadways rather than the school located at a considerable distance from highway. The difference in source type contribution at each school clearly depicts the difference in nature of location and type of activities in the vicinity of the sampling sites.

Keywords: Chemical Mass Balance; Particle concentration; Receptor modeling; PM_{2.5}; Vehicular emissions.

PM_{2.5} Emission Reduction by Technical Improvement in a Typical Coal-Fired Power Plant in China

Zizhen Ma, Zhen Li, Jingkun Jiang, Jianguo Deng, Yu Zhao, Shuxiao Wang, Lei Duan

Source: Volume 17, No. 2, February 2017, Pages 636-643 doi: 10.4209/aaqr.2016.05.0200

To investigate PM_{2.5} reduction by technical improvement in typical Chinese coal-fired power plants, two units built in different time with different particulate matter (PM) control technologies but with the same coal-fired boiler type were selected to characterize the concentrations of PM_{2.5} generated and emitted from coal-fired power plants. We found that significant benefit of PM_{2.5} emission reduction was achieved by technological improvement. Due to the increase in the installed capacity and the application of low NO_x burner alone, PM_{2.5} emission factor without adopting other air pollution control devices decreased from 0.153 kg t⁻¹ (the 100 MW unit) to 0.123 kg t⁻¹ (the 300 MW unit). With the help of an improved electrostatic precipitator (ESP) of which removal efficiency increased from 76.4% to 97.5%, PM_{2.5} emission factor further decreased from 0.014 kg t⁻¹ (the unit with a normal ESP) to 0.003 kg t⁻¹ (the unit with a cold-side ESP and a wet flue gas desulphurization (WFGD)). However, the application of flue gas denitrification and desulfurization devices may alter PM_{2.5} compositions and their emissions. For instance, the installation of a WFGD was found to largely increase the emissions of water-soluble ions in PM_{2.5} (e.g., SO₄²⁻, Ca²⁺, and NH₄⁺).

Keywords: PM_{2.5}; Emission factor; Coal-fired power plant; Water-soluble inorganic ion; Wet flue gas desulphurization (WFGD).

Transformations of Aerosol Particles from an Outdoor to Indoor Environment

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Source: Volume 17, No. 3, March 2017, Pages 653-665 doi: 10.4209/aaqr.2016.08.0355

2017/02/09 Aerosol particle size and chemical composition during summer and winter were investigated in this study. An automated switching valve allowed for indoor and outdoor environments to be sampled near-simultaneously with the same high temporal-resolution instrumentation. During the study, no known indoor sources were present and the sampled room was unoccupied throughout. Accumulation mode indoor/outdoor (I/O) ratios were substantially lower in winter than in summer. This reduction was attributed to particles of outdoor origin shrinking as they entered the warmer and drier indoor environment. An essential factor in this process appeared to be the difference (gradient) between the temperature and relative humidity of the indoor and outdoor environments during the winter. Online aerosol mass spectrometer measurements recorded a 34–38% decrease in I/O ratios for all nonrefractory species during the winter relative to the summer. A similar change in I/O ratios for all species indicated that physical, rather than chemical, processes were responsible. To assess the relative influence of various

physical factors on I/O relationships, Spearman rank statistical tests were carried out. These identified wind speed to be negatively correlated to the indoor concentrations for all species. Wind roses incorporating I/O ratios were applied and showed that the wind speed and direction influenced the changes in the indoor composition. The relative outdoor concentration of different aerosol species, steepness of the I/O temperature gradient, and wind speed variability are concluded to be essential factors in I/O aerosol transformations.

Keywords: I/O ratio; Shrinkage; Dissociation; Aerosols; Ammonium nitrate

Relationships between Outdoor and Personal Exposure of Carbonaceous Species and Polycyclic Aromatic Hydrocarbons (PAHs) in Fine Particulate Matter (PM_{2.5}) at Hong Kong

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Source: Volume 17, No. 3, March 2017, Pages 666-679 doi: 10.4209/aaqr.2016.08.0349

Personal and ambient fine particulate matter (PM_{2.5}) samples were simultaneously collected at Hong Kong during winter in 2014. Mass concentration, organic carbon (OC), elemental carbon (EC), and polycyclic aromatic hydrocarbons (PAHs) relationships were analyzed. The correlations of personal and ambient concentrations of PM_{2.5}, OC, and EC indicated the ambient concentrations were the factors showing influences on the personal exposures. Personal to ambient (P/A) ratios in PM_{2.5}, OC, and EC were all > 1, suggesting influences between indoor sources and/or personal activities. Significant higher ambient ΣPAHs concentrations with P/A ratios were nevertheless < 1. The Σ15 U.S. EPA priority PAHs accounted for 50.6% and 70.8% of ΣPAHs in personal and ambient samples, respectively. The ratios of indicator compounds confirmed the origin of PAHs in personal PM_{2.5}, which were found to be associated predominantly with traffic emissions and the influence by the indoor sources.

Keywords: Personal exposure; Fine particulate matter; Carbonaceous aerosol; Polycyclic aromatic hydrocarbons.

Evaluating the Effects of Springtime Dust Storms over Beijing and the Associated Characteristics of Sub-Micron Aerosol

Peng Xu, Junke Zhang, Dongsheng Ji, Zirui Liu, Guiqian Tang, Bo Hu, Changsheng Jiang, Yuesi Wang

Source: Volume 17, No. 3, March 2017, Pages 680-692 doi: 10.4209/aaqr.2016.05.0195

In order to understand the characteristics, sources and processes of non-refractory submicron particles (NR-PM₁), an Aerodyne high-resolution time-of-flight aerosol mass spectrometer (HR-

ToF-AMS) was deployed to acquire observational data during the spring (April 1 to 30) in Beijing, China, in 2012. Based on PM₁₀, PM_{2.5} and NR-PM₁ mass concentrations observation, satellite images and the back trajectory analysis, one haze and dust storm episodes were recorded during the campaign, in addition, one clean episodes was also added to the comparison as a reference. The NR-PM₁ mass concentration was 97 $\mu\text{g m}^{-3}$ during the haze episodes, while it was approximately 12 times and 1.7 times that on the clean and dust episodes, respectively. In addition, the secondary inorganic aerosol (sulfate, nitrate and ammonium) contributed the largest fraction of NR-PM₁ (69%) during the haze episodes. The dust storms originated from the northwestern caused the PM₁₀ peaking at 826 $\mu\text{g m}^{-3}$, with an average of $364 \pm 186 \mu\text{g m}^{-3}$ and higher than the haze episodes (241 $\mu\text{g m}^{-3}$). In addition, compared to the clean episodes (the NR-PM₁ mass was 8 $\mu\text{g m}^{-3}$), the dust storms caused the average NR-PM₁ mass reaching 56 $\mu\text{g m}^{-3}$, corresponding to the secondary components significantly increased, including sulfate (9.5 $\mu\text{g m}^{-3}$), nitrate (8 $\mu\text{g m}^{-3}$), ammonium (6 $\mu\text{g m}^{-3}$) and OOA (6 $\mu\text{g m}^{-3}$). The backward trajectory clustering analysis indicated the air mass from the southeast (at a frequency more than 30%) contained the higher NR-PM₁ concentration (more than 80 $\mu\text{g m}^{-3}$) corresponding to the higher sulfate, nitrate and ammonium contributions.

Keywords: NR-PM₁; Organic aerosols; Dust storm; Springtime; Beijing.

The Impacts of Emission Control and Regional Transport on PM_{2.5} Ions and Carbon Components in Nanjing during the 2014 Nanjing Youth Olympic Games

Derong Zhou, Bing Li, Xin Huang, Aki Virkkula, Haisuo Wu, Qiuyue Zhao, Jie Zhang, Qiang Liu, Li Li, Chunyan Li, Feng Chen, Siyu Yuan, Yuezheng Qiao, Guofeng Shen, Aijun Ding

Source: Volume 17, No. 3, March 2017, Pages 730-740 doi: 10.4209/aaqr.2016.03.0131

Highly time-resolved measurements of water soluble ions, organic and elemental carbon concentrations in the particle diameter size range $D_p < 2.5 \mu\text{m}$ (PM_{2.5}) were performed at a downwind urban site in Nanjing in the western part of the Yangtze River Delta (YRD) in eastern China during the 2014 Youth Olympic Games (YOG). In this study, we discuss the impacts of emission control in Nanjing and the surrounding areas during the YOG and regional/long-range transport on PM_{2.5} pollution in Nanjing. The average concentrations of NO₃⁻, SO₄²⁻, NH₄⁺ were 12.1 ± 9.9 , 16.5 ± 9.2 , $9.0 \pm 5.4 \mu\text{g m}^{-3}$ during the YOG, and increased 34.3%, 53.7%, 43.9% after the YOG, respectively. The control of construction or on-road soil dust and control of industry led to the decrease of Ca²⁺ concentration by 55% and SO₂ concentration by 46%. However, SO₄²⁻ concentrations remained at relatively high levels, suggesting a significant impact of regional pollution to secondary fine particles in Nanjing. Strong correlations between OC and EC were observed during and after the YOG. A higher percentage (41%) of secondary organic carbon in Nanjing during the YOG periods was consistent with high potential photochemistry and low contributions from coal combustion. Lagrangian dispersion modelling results proved that the city clusters along the Nanjing and Shanghai axis were the major source region for high PM_{2.5} pollution in upwind Nanjing. This work shows that short-term strict control measures could improve the air quality, especially that affected by the primary pollutants; however, regional collaborative control strategy across administrative borders in the YRD is needed for a substantial improvement of air quality.

Keywords: Air quality; Fine particulate matter; Emission control; Regional transport; The 2014 Nanjing Youth Olympic Games.

Spatial and Temporal Variability of the PM_{2.5}/PM₁₀ Ratio in Wuhan, Central China

Gang Xu, Limin Jiao, Boen Zhang, Suli Zhao, Man Yuan, Yanyan Gu, Jiafeng Liu, Xin Tang

Source: Volume 17, No. 3, March 2017, Pages 741-751 doi: 10.4209/aaqr.2016.09.0406

Fine particles (PM_{2.5}) and coarse particles (PM_{2.5-10}) are generally produced by different sources, so the PM_{2.5}/PM₁₀ ratio reveals characteristics of particle pollution. The ratio can be used to characterize the underlying atmospheric processes and evaluate historical PM_{2.5} pollution in absence of direct measurements. However, application of the ratio needs its varying pattern because PM concentrations change significantly at time and space. Hourly PM_{2.5} and PM₁₀ observations at nine monitoring sites in urban area (Urban-sites) and one remote Background-site in Wuhan in 2013–2015 were collected to investigate both long-term, short-term temporal variation and spatial distribution, spatial disparity of the ratio at a city scale. The results show that annual average PM_{2.5}/PM₁₀ ratio is 0.62 at Urban-sites and 0.68 at Background-site with apparent seasonal, monthly and daily variations. The ratio reaches the maximum in winter because of stable atmospheric conditions. There are apparent night-day differences of daily variation of the ratio, which increases at night in all seasons in consequence of temperature inversion and declines in the daytime with a moderate rise in the afternoon. We find obvious spatial gradients of the ratio that gradually increases from urban core to urban fringe and to suburban. This study provides further insights to the spatio-temporal variability of PM_{2.5}/PM₁₀ ratio. The evidence indicates that the variability of PM_{2.5}/PM₁₀ should be noticed in its applications.

Keywords: Air pollution; Particulate matter; Ratio; Spatio-temporal variability; China.

Propensity to Migrate and Willingness to Pay Related to Air Pollution among Different Populations in Wuhan, China

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Source: Volume 17, No. 3, March 2017, Pages 752-760 doi: 10.4209/aaqr.2016.05.0178

Objective: To understand parents' propensity to migrate and willingness to pay with respect to outdoor air pollution, and to explore related affecting factors. Methods: This study used a convenience sample and subjects were collected from a community in

Wuchang District and Children's Hospital of Wuhan, respectively. A designed questionnaire was used for this study. Univariable and multivariable logistic regression models were applied to analyze the relationship between parents' individual and familial characteristics and related behavioral intentions to air quality improvement. Statistical analysis was done with SAS 9.1. Results: The questionnaire was completed by 865 subjects (response rate = 86.5%). The number of people with migrant intent was 150(36.4%) from hospital group, and 139(30.7%) from community group. In the hospital group, subjects with higher knowledge of air quality ($OR = 6.268, p < 0.05$) and higher average annual household income (AAHI), which was equal or more than 50,000 Yuan ($OR = 2.045, p < 0.01$), were found to be more intent to migrate. AAHI ($OR = 1.939, p < 0.05$) was also the affecting factor in the community group correspondingly. Those willing to pay for air quality improvement included 297 people (72.1%) from the hospital group and 333 people (73.5%) from the community group, and affecting factors was the public responsibility for air quality improvement (hospital group: $OR = 3.380, p < 0.01$; community group: $OR = 4.436, p < 0.01$).

Conclusions: This study indicated high tendency of propensity to migrate for avoiding poor air condition and willingness to pay to improve air quality in Wuhan. Local governments should pay more attention to parents' knowledge of air pollution and attitudes towards government management of air quality, especially those willing to migrate.

Keywords: Air pollution; Propensity to migrate; Willingness to pay; Income; Responsibility.

Cross-Sectional View of Atmospheric Aerosols over an Urban Location in Central India

Subin Jose, Biswadip Gharai, Pamaraju Venkata Narasimha Rao

Source: Volume 17, No. 3, March 2017, Pages 761-775 doi: 10.4209/aaqr.2016.04.0154

Surface, column and vertically resolved variations of physical and optical properties of atmospheric aerosol over Hyderabad, a tropical urban location in central India are explored on the basis of ground based and satellite retrieved data. Annual mean aerosol optical depth (τ) observed with Microtops sun-photometer is 0.61 ± 0.07 and seasonally it varied from 0.71 ± 0.06 in pre-monsoon to 0.55 ± 0.05 in winter. Aerosol types are categorized based on Ångström exponent (α) and τ relations; revealed that the study region is dominated by mixed type (MT) aerosol followed by urban/industrial aerosols under high τ (HUI) category. A consistent diurnal variation of black carbon (BC) is observed irrespective of seasonal variation with annual BC mass concentration is found to be $9.7 \pm 1.9 \mu\text{g m}^{-3}$. During Telangana Survey day, which was the least pollutant day showed a reduction of 75% BC concentration during day time in comparison to five years average values, indicating the influence of anthropogenic effect over Hyderabad. Vertical information's on aerosol are analyzed using Cloud Aerosol Lidar Pathfinder Satellite Observations (CALIPSO) and ground based Lidar (LAMP) data. LAMP data analysis shows a significant elevated aerosol layer up to 4 km during pre-monsoon while aerosols are confined below 3 km during post-monsoon and winter. Long term CALIPSO observations revealed that during post-monsoon to winter, the study area is dominated (~60%) by 'urban' aerosol; while during pre-monsoon period ~75% of the aerosol type belongs to 'dusty mix' category. A decline in short wave flux at the top of the atmosphere ($0.66 \text{ Wm}^{-2} \text{ yr}^{-1}$) is observed, as revealed by long term Clouds and Earths Radiant

Energy System (CERES) data analysis with higher decline rate observed in winter ($1 \text{ Wm}^{-2} \text{ yr}^{-1}$) followed by pre-monsoon ($0.8 \text{ Wm}^{-2} \text{ yr}^{-1}$).

Keywords: Aerosol optical depth; Black carbon; Aerosol backscatter coefficient; SW flux at TOA.

Airborne Bacterial Communities of PM_{2.5} in Beijing-Tianjin-Hebei Megalopolis, China as Revealed By Illumina MiSeq Sequencing: A Case Study

Jing-Feng Gao, Xiao-Yan Fan, Hong-Yu Li, Kai-Ling Pan

Source: Volume 17, No. 3, March 2017, Pages 788-798 doi: 10.4209/aaqr.2016.02.0087

Bacteria are ubiquitous and abundant in the atmosphere and some of them are potential pathogens known to cause diseases or allergies in humans. However, the quantities and compositions of total airborne bacterial community and their relationships with environmental factors remain poorly investigated. Here, a case study of the total airborne bacteria of PM_{2.5} collected at six cities in Beijing-Tianjin-Hebei (BTH) megalopolis, China were profiled using quantitative polymerase chain reaction (qPCR) and Illumina MiSeq (PE300) sequencing. qPCR results showed the high abundance of total airborne bacteria of PM_{2.5} in BTH, ranging from $4.82 \times 10^4 \pm 1.58 \times 10^3$ to $2.64 \times 10^5 \pm 9.63 \times 10^4$ cell m⁻³ air, and averaged 1.19×10^5 cell m⁻³ air. The six PM_{2.5} samples were classified into three groups. *Proteobacteria*, *Cyanobacteria*, *Actinobacteria* and *Firmicutes* were the four dominant phyla of PM_{2.5}. 18 common potential pathogens with extremely low percentage (3.61%) were observed, which were dominated by *Enterococcus faecium* and *Escherichia coli*. Plants and soil are probably the main sources of bacteria in PM_{2.5}, as suggested by the high percentages of Chloroplast, plant-associated bacteria (e.g., *Rhizobiales* and *Sphingomonadales*) and soil-inhabiting bacteria (e.g., *Burkholderiales* and *Pseudomonadales*). Variation partitioning analysis (VPA) indicated that the atmospheric pollutants explained the most of the variation (31.90%) in community structure of PM_{2.5}, followed by meteorological conditions (15.73%) and the chemical compositions of PM_{2.5} (11.32%). The case study furthers our understanding of the diversity and composition of airborne bacterial communities of PM_{2.5} in BTH, and also identified the main factors shaping the bacterial communities.

Keywords: PM_{2.5}; Illumina MiSeq sequencing; Airborne bacterial communities; Potential pathogens; Beijing-Tianjin-Hebei megalopolis.

Time-Dependent Size-Resolved Bacterial and Fungal Aerosols in Beijing Subway

Hanqing Fan, Xinyue Li, Jiahao Deng, Guillaume Da, Evelyne Gehin, Maosheng Yao

Source: Volume 17, No. 3, March 2017, Pages 799-809 doi: 10.4209/aaqr.2016.03.0114

Despite of an important concern, human bioaerosol emission into subway is not well and directly characterized. Here, we used bioaerosol detector and next generation sequencing methods to investigate time-dependent bioaerosol size distributions in Beijing subway system between March and April, 2015. In contrast to weekends, weekday microbial aerosol results exhibited strong time dependence with higher bacterial and fungal aerosol levels up to 2083 CFU m⁻³ and 483 CFU m⁻³ observed, respectively, for the peak hours. During the peak hour (17:30–18:30), bioaerosol emissions at 0.8–3 µm was detected, while about three times higher concentration levels were observed compared to those during the off-peak hour (22:00–23:00). Similar bioaerosol size distributions were observed between ventilation outlets and subway platform air. During off-peak hours, subway bioaerosols had similar size distributions with the outside air. Sequence results revealed a vast array of airborne microbial species which varied from one station to another, but with *Aspergillus* spp. as dominant fungal species, and *Staphylococcus*, *Pseudomonas* as primary bacterial genera including human opportunistic ones. Our results provide direct online observations of human contributions to subway size-resolved bioaerosols, and enhancing ventilation system might help for controlling the exposure especially during the peak-hours.

Keywords: Bacteria; Fungi; Beijing subway; Fluorescent bioaerosol particle; Ventilation.

Influence of Surgical Smoke on Indoor Air Quality in Hospital Operating Rooms

Dong Hee Choi, Seock Hwan Choi, Dong Hwa Kang

Source: Volume 17, No. 3, March 2017, Pages 821-830 doi: 10.4209/aaqr.2016.05.0191

The objective of this study is to analyze the volatile organic compounds (VOCs) in the surgical smoke generated during laparoscopic surgery, and determine their influence on the indoor air quality in hospital operating rooms (ORs). Field measurements were carried out during eight surgeries in conventional and robotic ORs in which an electrosurgery system was being used, thus continuously generating surgical smoke. The VOCs were measured at three different locations, in the patients' abdominal cavities, beside the surgical table, and at the exhaust vent. Other indoor pollutants including carbon monoxide (CO), carbon dioxide (CO₂), and total airborne bacteria (TAB) in the indoor air were measured at the exhaust to assess the general indoor air quality in the ORs. The results from the patients' abdominal cavities confirmed that the surgical smoke contained abundant VOCs, with the levels of benzene and toluene exceeding the health guidelines. Compared to the results obtained in the abdominal cavity, the measurements obtained at the surgical table and exhaust vent exhibited low levels of VOCs, indicating that the actual exposure to these compounds was minimized in a highly-ventilated operating room. However, the benzene concentration in the operating room approached a level that threatens the health of the occupants. Therefore, the results of this study suggest that there is a potential health risk to the surgeon who is closest to the point of origin of the surgical gas, as well as a need for further attention to identify the local pollutant dispersion near the surgical table while the ventilation system is operating.

Keywords: Surgical smoke; Hospital operating room; IAQ; Volatile organic compounds (VOCs)

Assessment of City Level Human Health Impact and Corresponding Monetary Cost Burden Due to Air Pollution in India taking Agra as a Model City

Kamal Jyoti Maji, Anil Kumar Dikshit, Ashok Deshpande

Source: Volume 17, No. 3, March 2017, Pages 831-842 doi: 10.4209/aaqr.2016.02.0067

Objectives of the present study are to provide quantitative estimations of air pollution health impacts and monetary burden on people living in Agra city, the fourth most populated city in Uttar Pradesh, India. To estimate the direct health impacts of air pollution in Agra city during year 2002 to 2014, 'Risk of Mortality/Morbidity due to Air Pollution' model was used which is adopted from air quality health impact assessment software, developed by world health organization (WHO). Concentrations of NO₂, SO₂ and PM₁₀ have been used to assess human health impacts in terms of attributable proportion of the health outcome as- annual number of excess cases of total mortality, cardiovascular mortality, respiratory mortality, hospital admission chronic obstructive pulmonary disease (COPD), hospital admission respiratory disease and hospital admission cardiovascular disease and it was observed that attributable number of cases were 1325, 908, 155, 138, 1230 and 348 respectively in year 2002. However, after thirteen years these figures increased to 1607, 1095, 189, 167, 1568 and 394 respectively. From these results, it was observed that from 2002 to 2014, the attributable number of cases increased almost by 13.43 to 27.52%. As a result, the monetary cost burden due to air pollution related health effects also increased very highly; it was 67.99 million US\$ in 2002, which transformed into 254.52 million US\$ in 2014. In future, if air quality continues to follow current pollutant concentration trend, the monetary cost burden will reach a level of US\$ 570.12 million in year 2020, which is not only a thoughtful matter but also a threatful matter and it signifies the importance of rectification measures for air quality in Agra city.

Keywords: Air pollution; Relative risk; AirQ; Health endpoint; Monetary cost.

Measured Indoor Aerosol Concentration Arising from Commonly-Used Food and Medicinal Powders: A Pilot Study

Roy Thomas, Miriam Byrne

Source: Volume 17, No. 3, March 2017, Pages 843-848 doi: 10.4209/aaqr.2016.08.0345

Inhalation exposures in the ambient environment may trigger allergic or other adverse responses in susceptible individuals, and this study aims to elucidate the contribution, if any, of airborne particles resulting from commonly-used medicinal and food powders to this response. In a laboratory room, six powders (two types of paracetamol-containing sachet, dried skimmed milk, instant coffee powder containing milk, a non-dairy coffee whitener containing milk protein, and a powdered peanut butter) were individually utilised under representative "real life" conditions, with ten replicates in each case. Particle concentrations were measured at the emission location and at a distance of 1 m. For each powder, a large variation in evolved particle concentrations is seen between individual events. Of the powders tested, only flavoured paracetamol demonstrated any

potential for dispersion to a distance of 1 m from the source. Short term exposures were estimated and from these, it was concluded that the particle concentrations evolving from powdered peanut butter and paracetamol powders were of little concern, although further investigation of specific scenarios is merited. The range of short-term exposures calculated for milk powder products was 0.019–0.087 μg , which was comparable to estimated levels that have elicited adverse health responses in other studies.

Keywords: Powder; Human Exposure; Indoor air pollution.

Quantification of Non-Linear Dynamics and Chaos of Ambient Particulate Matter Concentrations in Muzaffarabad City

Sharjil Saeed, Wajid Aziz, Muhammad Rafique, Imtiaz Ahmad, Kimberlee J. Kearfott, Salma Batool

Source: Volume 17, No. 3, March 2017, Pages 849-856 doi: 10.4209/aaqr.2016.04.0137

The present study was carried out for quantification of non-linear dynamics and chaos of ambient particulate matter (PM) concentrations in Muzaffarabad city. $\text{PM}_{1.0}$ and $\text{PM}_{2.5}$ concentrations were monitored at six different locations for a continuous 6 h period. The linear behavior of the acquired time series data was analyzed using descriptive statistics. Specifically, the chaotic temporal behavior of the PM concentration was analyzed using phase space reconstruction, the Hurst exponent, and the largest Lyapunov exponent. The average mutual function was used to calculate proper time delay, while the false nearest neighbor method was used to calculate the proper embedding dimension for the phase space reconstruction. No health-protective quantitative standard exists for $\text{PM}_{1.0}$ concentrations. However, the mean concentration of $\text{PM}_{2.5}$ was considerably higher than the standards, developed by WHO, US-EPA and European Union directives, at all six locations. For $\text{PM}_{1.0}$ minimum, $293 \pm 149 \mu\text{g m}^{-3}$, and maximum, $544 \pm 490 \mu\text{g m}^{-3}$, values were recorded at CMH Chowk and Chehla Bridge locations respectively. For $\text{PM}_{2.5}$ minimum, $394 \pm 262 \mu\text{g m}^{-3}$ and maximum, $633 \pm 426 \mu\text{g m}^{-3}$, values were recorded at CMH Chowk and at old secretariat respectively. The results indicates $\text{PM}_{1.0}$ and $\text{PM}_{2.5}$ concentration time series can be modeled using phase space reconstruction by properly selecting the embedding parameters. The positive largest Lyapunov exponent reveals a strong chaotic signature in the system dynamics for both particulate sizes. Furthermore, Hurst exponent for both particulates was close to 1, showing highly persistent time series pattern.

Keywords: Fine particulate matter; Chaotic behavior; Hurst exponent; Time series analysis.

Particulate Air Pollution at Schools: Indoor-Outdoor Relationship and Determinants of Indoor Concentrations

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Source: Volume 17, No. 3, March 2017, Pages 857-864 doi: 10.4209/aaqr.2016.03.0128

This study aimed to assess the relationship between indoor and outdoor particulate air pollution at primary schools, and identify the determinants of indoor pollution concentrations. The study was conducted in six classrooms within six primary schools in Sari, Northern Iran. Indoor concentrations of particulate matter (PM) with an aerodynamic diameter of less than 10 μm (PM_{10}), 2.5 μm ($\text{PM}_{2.5}$), and 1 μm ($\text{PM}_{1.0}$) were assessed in classrooms, and outdoor concentrations of $\text{PM}_{2.5}$ on the school playgrounds were monitored simultaneously by using two real-time and portable dust monitors during autumn, winter, and spring, yielding 26 sampling days for each school in total. The highest outdoor and indoor $\text{PM}_{2.5}$ concentrations were found in winter and spring, respectively. The mean indoor $\text{PM}_{2.5}$ concentration ($46.9 \pm 32.9 \mu\text{g m}^{-3}$) was higher than that measured outdoors ($36.8 \pm 33.2 \mu\text{g m}^{-3}$). Indoor $\text{PM}_{2.5}$ and $\text{PM}_{1.0}$ were moderately correlated with outdoor $\text{PM}_{2.5}$ concentrations, which was the main determinant for all indoor particulate concentrations, however, a distinct pattern was observed for PM_{10} and $\text{PM}_{2.5}$ compared to $\text{PM}_{1.0}$. While meteorological variables (i.e., ambient temperature, relative humidity) could predict indoor PM_{10} and $\text{PM}_{2.5}$ concentrations, the total area of the windows and the number of students in a classroom were predictors for $\text{PM}_{1.0}$ levels. The findings of this study could inform policymakers in implementing evidence-based targeted interventions aimed at reducing air pollution in school settings.

Keywords: Classrooms; Particulate matter; $\text{PM}_{2.5}$; PM_{10} ; Iran.

Characterization of Inorganic Elements within $\text{PM}_{2.5}$ and PM_{10} Fractions of Fly Ashes from Coal-Fired Power Plants

Zhiyong , Yaqin , Huiqiao , Peng Zhao, Xiangchao Zeng, Songtao , Yunjun , Lei Wang, Aiqin, Huiying Gao, Fude , John Kennedy Mwangi

Source: Volume: 17, Issue: 4, Pages: 1105-1116 DOI: 10.4209/aaqr.2017.02.0071

In this study 15 fly ash samples were collected from 15 large-scale coal-fired power plants (CFPPs) in China. The samples were then re-suspended through $\text{PM}_{2.5}$ and PM_{10} inlets and analyzed for the contents of 39 inorganic elements (IEs) using inductively coupled plasma-mass spectroscopy (ICP-MS) and inductively coupled plasma-optical emission spectrometry (ICP-OES). The results show that the particle size distributions for the 15 FAs samples exhibited bimodal patterns. The $\Sigma 39\text{IEs}$ (g g^{-1}) for the $\text{PM}_{2.5}$ (0.292–0.564) in all the 15 CFPPs were higher than that of PM_{10} (0.269–0.403). Except for Cu, all the other 38 IEs were more enriched in the $\text{PM}_{2.5}$ with the $\text{PM}_{2.5}/\text{PM}_{10}$ ratios being 1.06–1.73. Considering 13 heavy metals, the same orders occurred between $\text{PM}_{2.5}$ and PM_{10} with $\text{Al} \gg \text{Cr} > \text{Zn} > \text{Mn} > \text{Cu} > \text{V} > \text{Pb} > \text{Sn} > \text{Co} > \text{As} > \text{Sb} > \text{Tl} > \text{Cd}$. More attention should be

paid to the high contents of Cr in both PM_{2.5} (1310 mg g⁻¹) and for PM₁₀ (1240 mg g⁻¹) from all 15 CFPPs. 23 IEs for PM₁₀ and 26 IEs for PM_{2.5} had the geo-accumulation index (I_{geo}) values higher than 0, indicating different pollution levels for them. On the other hand, there was moderate to extreme levels of pollution for Cr, Zn, Cu, Pb, Sn, Sb, Tl, Cd and Al based on I_{geo} values. The element profiles for PM_{2.5} or PM₁₀ from 15 CFPPs were similar based on low coefficients of divergence for PM_{2.5} (0.254 ± 0.038) and PM₁₀ (0.244 ± 0.054) according to the comparison between any two CFPPs. Most elements with low relative enrichment factors (REF) as less than 0.70 or 0.70–1.30 indicated no or weak condensation occurred for them during coal combustion, while Cr, Cu, Zn, Sn, W and Pb had REF values higher than 1.30 indicated that significant condensation occurred for these elements.

Keywords: Coal-fired power plant Inorganic elements Fly ash PM_{2.5} PM₁₀

Impact of Magnetic Tube on Pollutant Emissions from the Diesel Engine

Chia-Yang Chen, Wen-Jhy Lee, John Kennedy Mwangi , Lin-Chi Wang , Jau-Huai Lu

Source: *Volume: 17, Issue: 4, Pages: 1097-1104 DOI: 10.4209/aaqr.2016.11.0478*

Magnetic field applied to fuel can alter its characteristics in terms of forces that hold the hydrocarbons together. This principle was used to investigate the impact of incorporating a magnetic tube in the fuel intake in a diesel generator on specifically the energy performance and pollutant emissions. A diesel generator was fitted with a magnetic tube in the fuel intake, which had valves to switch from without magnetic tube case to with magnetic tube case. The diesel generator was operated at constant speed of 1800 rpm at idle condition, 25% and 50% loads, respectively. Additionally, two real diesel cars were deployed with magnetic tube and their fuel consumption compared with that of a car without magnetic tube. With application of magnetic tube, the brake specific fuel consumption and fuel consumption were decreased by an average of 3.5% and 15%, respectively, while the brake thermal efficiency was improved by approximately 3.5%. The particulate matter, carbon monoxide, hydrocarbons and carbon dioxide emissions reduced in the range of 21.9–33.3%, 5.4–11.3%, 29.4–64.7% and 2.68–4.18%, respectively. Both the total PAH concentrations and total BaP_{eq} concentrations can be reduced by about 63%, 45% and 51%, respectively for idle condition, 25% and 50% loads, respectively. These results show that application of magnetic tubes in the diesel engine is a promising technology in pollutant reduction and energy saving.

Keywords: Diesel engine Magnetic tube Regular pollutant Energy performance PAHs

Decomposition Analysis of CO₂ Emissions from Coal - Sourced Electricity Production in South Africa

Mohamed Beidari, Sue-Jane , Charles Lewis

Source: Volume: 17 , Issue: 4 , Pages: 1043-1051 DOI: 10.4209/aaqr.2016.11.0477

In 2013, the electricity sector was the largest source of South Africa's CO₂ emissions, accounting for about 60% of its total. South Africa (SA) is one of the highest CO₂ emitters on a per capita basis when compared to many developed and developing countries. For a better understanding of the driving forces leading the electricity-related CO₂ emission per person, this paper applies the Log Mean Divisia Index (LMDI) to analyze the influence of the factors which ruled electricity generation-related CO₂ emission in SA over the period 1990–2013. We focused on coal which is the dominant fuel used in SA for electricity generation. The results show that the electricity generation intensity effect plays the dominant role in decreasing CO₂ emissions. However, the effect of economic activity is the major determinant that contributes to increasing CO₂ emissions. In order to reduce its greenhouse gas emissions levels, meet the agreement of the COP21 agreement, and fight against CO₂ emissions per-capita associated with electricity generation, it is recommended that SA's government should improve the efficiency of its existing electricity power generation plants and expand more of its renewable energy sources (nuclear included).

Keywords: Carbon dioxide Coal consumption Electricity generation LMDI method

Effects of Injection Pressure on Geological CO₂ Storage in the Northwest Taiwan Basin

Chun-Ping Jen , Cai Li , Keni Zhang

Source: Volume: 17 , Issue: 4 , Pages: 1033-1042 DOI: 10.4209/aaqr.2016.12.0526

Geological storage of CO₂ has been viewed as an effective means of reducing CO₂ emissions and mitigating the greenhouse effect. In the Taiwan area, the Western Taiwan Basin is suitable for million-ton-scale geological CO₂ storage. Numerical methods were used in this study to investigate reservoir performance under various injection pressures. Three formations in the basin, the Chingshui Formation, Kueichulin Formation and Nanchunag Formation, were modeled. Three different injection pressures (1.3, 1.5 and 1.7 times the initial pressure) were considered. The simulation results show that the cumulative injected CO₂ mass is proportional to the applied injection pressure and that the storage security increases over time. An annual injection rate of 5 Mt year⁻¹ could be achieved by applying an injection pressure of 1.5 times the initial pressure at the injection well. The pressure accumulation in the system featured three stages. The overpressurization effects associated with the injection in the system decrease, and the pressure in the system almost returns to the original pressure conditions after 50 years following cessation of injection. The CO₂ gas plumes simulated in this study also suggest that the modeled injection scenarios are safe in terms of CO₂ leakage from the vertical fault in this area.

Keywords: CO₂ Geological storage Simulation Injection pressure Fault

Modeling the Airflow and Particle Dispersion in Street Canyons under Unsteady Thermal Environment with Sinusoidal Variation

Dan Mei, Yan Wang, Qihong Deng

Source: Volume: 17, Issue: 4, Pages: 1021-1032 DOI: 10.4209/aaqr.2016.12.0559

Unstable temperature stratification conditions have a considerable influence on pollutant diffusion inside street canyons. In this study, we tried to model the air flow and particle dispersion in street canyons under unsteady thermal environment. The sinusoidal variation was used to model the atmosphere temperature on the basis of the solar radiation cycle that occurs over a day. The two-dimensional model of step-up building layouts was applied as the research object and an RNG turbulence model was applied to study the dynamic characteristics of instantaneous airflow, the dimensionless air exchange rate (ACH) and turbulent kinetic energy (TKE) in the different canyons. A series of numerical simulations were performed for different Richardson numbers (Ri). The results demonstrated that the stream function within the street canyons exhibited a periodic shift over a day, and the flow morphology gradually evolved from paralleled bilateral vortexes into a row of vortexes, particularly when the ground temperature increased. Moreover, the local PM concentrations at different times were obtained, and they were affected by the flow field patterns. The total PM mass in the street canyon decreased as the air velocity increased. Furthermore, the dimensionless ACH and TKE exhibit a noticeable fluctuation with time at higher Ri and stronger buoyancy. As the Ri decreases, the enhanced forced convection causes the ACH and TKE to remain constant over time. From these results, inhabitants should be advised to adopt preventative measures aimed at the PM pollution according to the time and location where they live.

Keywords: Unsteady thermal environment Sinusoidal variation Step-up street canyon Air flow Particle dispersion

Arsenic Exposure during Preventive Maintenance of an Ion Implanter in a Semiconductor Manufacturing Factory

Seunghon Ham, Chungsik Yoon, Sunju Kim, Jihoon Park, Ohun Kwon, Jungjin Heo, Donguk Park, Sangjun Choi, Seungwon Kim, Kwonchul Ha, Won Kim

Source: Volume: 17, Issue: 4, Pages: 990-999 DOI: 10.4209/aaqr.2016.07.0310

Workers in the semiconductor fabrication process may be exposed to higher levels of hazardous materials, such as arsenic, during preventive maintenance (PM) tasks than during the regular operation of the fabrication process. This study investigates the exposure to arsenic and other metals during PM tasks in the ion implantation process. Airborne arsenic samples and bulk samples were obtained during various ion implanter PM tasks in a semiconductor fabrication factory. The arithmetic mean (AM) and standard deviation (SD) of airborne arsenic in personal samples were $0.64 \mu\text{g m}^{-3} \pm 0.92 \mu\text{g m}^{-3}$ ($n = 9$), and the highest level was $2.39 \mu\text{g m}^{-3}$ during medium-current ion implanter PM tasks. For area samples, the AM and SD were $0.42 \mu\text{g m}^{-3} \pm 0.69 \mu\text{g m}^{-3}$ ($n = 5$) and the highest level was $1.79 \mu\text{g m}^{-3}$ during medium-current ion implanter PM tasks. Arsenic was

also found in the bulk samples of debris produced during PM tasks. Other metals (Ag, Al, Cu, Pb, Cr, Sn, Mn, Ti, Fe, and W) were found, but at low levels, prompting few health concerns compared with those of arsenic. This study found that PM workers were exposed to airborne arsenic levels that differed significantly according to the type of ion implanter used.

Keywords : Semiconductor Semiconductor Arsenic Arsenic Preventive maintenance Preventive maintenance Ion implantation Ion implantation Industrial hygiene Industrial hygiene

A Case Study of Long-Range Transport of Smoke Aerosols from Eastern Siberia to Northeast China in July 2014

Xiaojing Li, Xiangao Xia , Jingjing Song, Yufei Wu, Xiaoling Zhang, Renjian Zhang

Source: Volume: 17 , Issue: 4 , Pages: 965-974 DOI: 10.4209/aaqr.2016.04.0166

Long-range transport of biomass burning aerosols from Eastern Siberia to Northeast China in July 2014 was studied by using ground-based ambient measurements and satellite products. Intensive active fires were revealed in Eastern Siberia during the late of July by the Moderate Resolution Imaging Spectroradiometer (MODIS) active fire products. Under the favorable synoptic pattern, the smoke layer was transported to Northeast China, which led to significant enhancement of surface PM_{2.5} concentration. The peak PM_{2.5} concentration exceeded 100 $\mu\text{g m}^{-3}$ that was 3–6 times larger than the background level. High aerosol optical depth at 550 nm with daily value exceeding 1.0 was observed at a background site in Northeast China. Smoke aerosols were characterized by fine-mode dominated particles with very weak absorption. Air quality in Northeast China was revealed to be potentially impacted by the long-range transport of smoke aerosols from Eastern Siberia during the biomass burning season, which probably impacted human health, weather and climate. Therefore, further study on this issue is urgently required for quantitatively evaluating potential contribution of long-range transport to regional air pollution in Northeast China.

Keywords: Smoke Long-range transport Eastern Siberia Northeast China Air pollution

Spatial Variation of Ground Level Ozone Concentrations and its Health Impacts in an Urban Area in India

Amit Kumar Gorai , Paul B. Tchounwou, Gargi Mitra

Source: Volume: 17 , Issue: 4 , Pages: 951-964 DOI: 10.4209/aaqr.2016.08.0374

The present study was designed to analyze the spatial distributions of ground-level ozone (GLO) concentrations in Ranchi (Jharkhand, India) using geostatistical approaches. From September 2014 to August 2015, monthly GLO concentrations were monitored in 40-identified locations distributed

in the region of study. In every month, the monitoring was done at three different time periods of the day; 5.30 AM to 7.30 AM, 11.30 AM to 1.30 PM, and 5.30 PM to 8 PM). The time duration was assigned based on the temporal variations of GLO concentrations. The descriptive statistics indicate that the spatial mean ozone concentrations ranged from 23.45 $\mu\text{g m}^{-3}$ to 53.91 $\mu\text{g m}^{-3}$ in morning hours, from 82.50 $\mu\text{g m}^{-3}$ to 126.66 $\mu\text{g m}^{-3}$ in the day time and from 40.04 $\mu\text{g m}^{-3}$ to 71.25 $\mu\text{g m}^{-3}$ in the evening hours. The higher level of spatial variance observed in the months of December (standard deviation: 24.21), July (standard deviation: 29.59) and November (standard deviation: 19.60) for the morning, noon, and evening time, respectively. The effects of meteorological factors (wind speed and wind direction) on the ozone concentrations were also analysed. The study confirmed that wind speed is not the dominant factor for influencing the GLO concentrations. The study also analysed the ozone air quality index (OZAQI) for assessing the health impacts in the study area. The result suggests that most of the area had the moderate category of OZAQI (101-200) and that leads to breathing discomfort for people with lung and heart disease.

Keywords: Ground level ozone (GLO) Geostatistics Ordinary kriging AQI mapping Health effect

Characterization of Ambient Volatile Organic Compounds (VOCs) in the Area Adjacent to a Petroleum Refinery in Jinan, China

Guiqin Zhang, Ning Wang , Xiaojing Jiang, Ying Zhao

Source: Volume: 17 , Issue: 4 , Pages: 944-950 DOI: 10.4209/aaqr.2016.07.0303

56 VOCs were monitored for a whole year in a refinery vicinity area in suburban of Jinan, as well as PM_{2.5}, PM₁₀, and ozone. The results of VOCs and particulate matters showed that January was the most polluted month during the sampling period, possibly resulting from coal burning in the district heating period of Jinan. According to the concentration profiles of monthly variety of the ambient pollutants, the change of VOCs showed a certain extent of positive correlation with PM_{2.5} and PM₁₀, while negative correlation with ozone. It was found that the average concentration of total VOCs was 50.58 $\mu\text{g m}^{-3}$ while ethane and ethene were the most abundant VOC species with the concentrations of 24.58 and 3.94 $\mu\text{g m}^{-3}$, respectively. The high relative contribution of ethane was unusual, compared with the previous related VOC research in urban area of same city or other cities in China. Based on the analysis of the monitoring data, this area was not majorly affected by major VOCs pollutants from refinery emission. m & p-Xylene of 1.34 $\mu\text{g m}^{-3}$ was the most abundant pollutant of BTEX. The BTEX ratio analysis presented that traffic exhaust was not the major VOCs source.

Keywords: VOCs PM Ozone Refinery Ethane

Inorganic Chemical Composition of Fine Particulates in Medium-Sized Urban Areas: A Case Study of Brazilian Cities

Alexandra Beal, Camila A. Bufato, Daniela S. de Almeida, Rafaela Squizzato, Adriana Zemiani, Newmar Vernilo, Carla Estefani Batista, Graziela Salvador, Daniel L.G. Borges, Maria C. Solci, Alessandra F. da Silva, Jorge A. Martins, Leila D. Martins

Source: Volume: 17, Issue: 4, Pages: 920-932 DOI: 10.4209/aaqr.2016.07.0317

The aim of this study was to characterize the inorganic chemical composition of fine inhalable atmospheric particulate matter (PM_{2.5}) in medium-sized cities in Brazil. These cities account for a significant proportion of the population and are growing at rates above the national average, thereby demonstrating the importance of carefully analysing the possible impact of such growth on air quality over the coming decades. In 2013 and 2014, this study collected PM_{2.5} samples from sites in the cities of Londrina and Maringa in two seasons: winter and summer. The mean concentration of PM_{2.5} ranged from 4.4 $\mu\text{g m}^{-3}$ and 3.7 $\mu\text{g m}^{-3}$ during the summer to 10.3 $\mu\text{g m}^{-3}$ and 8.0 $\mu\text{g m}^{-3}$ in winter in Londrina and Maringa, respectively. The analysis of the major water-soluble ions, nitrate, sulphate and chloride, showed that they accounted for between 16.5% and 35.1% from the mass of PM_{2.5}, with sulphate providing the greatest contribution in all the campaigns. The nitrate/sulphate ratios ranged from 0.2 and 0.6, which are similar to the figures cited in the literature for other regions of the world. Although the PM_{2.5} concentrations are much lower than those observed in Asia and in Sao Paulo the participation of ions (%) is very close to that observed in Asian cities and significantly higher than the values recorded in other areas of Brazil, possibly as a result of the increased influence of burning of biomass and waste. The metals Zn, Pb, Cu and Mn found in the samples from all campaigns indicate that, in general, mobile sources are the main contributor to PM_{2.5}. The winter campaigns showed the highest concentrations of black carbon equivalent (BC_e). Absolute principal component analysis and enrichment factor analysis indicate the contribution of vehicular emission sources and biomass and waste burning to the inorganic chemical composition of PM_{2.5}.

Keywords: Emission sources Atmospheric particulate matter Metals Ions Black carbon

Chemical Characteristics of Aerosols in Coastal and Urban Ambient Atmospheres

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Source: Volume: 17, Issue: 4, Pages: 908-919 DOI: 10.4209/aaqr.2016.08.0342

Chemical characteristics of aerosols (PM₁ (on-line measurement) and PM_{2.5} (off-line measurement)) were investigated in coastal and urban atmospheres. Organics were the most dominant species in PM₁ at both sites, exhibiting little difference in the relative fractions of chemical constituents in PM₁ (organics, sulfate, black carbon (BC), nitrate, ammonium and

chloride) between two sites. However, a clear difference in the types of organics was found between the two sites. The fraction of oxidized (aged) organics was much higher at coastal site than at urban site. The nitrate fraction significantly increased at both sites in PM_{2.5} compared with that in PM₁, suggesting that a significant amount of nitrate exists at particle sizes of 1 μm –2.5 μm . Additionally, the aerosols observed at coastal site were acidic. At both sites, photochemical activity played an important role in enhancing sulfate and oxidized organics in the afternoon, thereby overcoming the dilution effect. More distinct diurnal patterns were observed for nitrate, BC and organics at the urban site compared to the coastal site. Chemical characteristics also varied with different air masses. The highest PM concentration was associated with the northwest continental air mass (the air mass passed over heavy industrial areas before arriving at the site and moved slowly compared to other air masses). Three PM events (sulfate-dominant versus organic-dominant) were observed during the sampling periods and were considered long-range transport (LTP) events. The water-soluble organic carbon (WSOC) and oxidized organic contents significantly increased during LTP events, suggesting that organics were highly aged during transport.

Keywords: Urban Coastal PM_{2.5} Chemical characteristics Aerosol mass spectrometer

Characterization of Chemical Composition in PM_{2.5} in Beijing before, during, and after a Large-Scale International Event

Xiaowen Yang, Shuiyuan Cheng, Jianbing Li, Jianlei Lang, Gang Wang

Source: Volume: 17, Issue: 4, Pages: 896-907 DOI: 10.4209/aaqr.2016.07.0321

To commemorate the 70th anniversary of the victory of the Chinese people's Anti-Japanese War and the World Anti-Fascist War, an international parade was held in Beijing in September 2015. In order to ensure satisfactory air quality during this event, a phased emission control measures were taken in Beijing and its surrounding provinces. The 24-h PM_{2.5} samples were collected in Beijing from August 1 to September 15, 2015 covering the period before, during and after this large-scale event. The observed PM_{2.5} data, meteorological data, emission reduction measures, and air mass trajectory simulation results were systematically analyzed to understand the pollution characteristics and chemical compositions of PM_{2.5} in Beijing. The results indicated that PM_{2.5} concentration during the two emission control phases was reduced by 61.7% comparing to the non-control period, but the regional transport of pollutants and meteorological conditions had a more prominent impact on PM_{2.5} than emission reduction during phase 2. The secondary water-soluble ions including SO₄²⁻, NO₃⁻, and NH₄⁺ were found as the main ions present in PM_{2.5}. During the entire emission control period, organic carbon (OC) and elemental carbon (EC) mass concentrations were decreased by 53.1% and 57.9%. A PM_{2.5} mass balance was analyzed, and it was found that the organic matter accounted for 29.3, 37.6 and 28.5% of the PM_{2.5} mass before, during and after the emission control, while the contribution of mobile sources to PM_{2.5} was relatively outstanding after a series of emission control measures.

Keywords: Air mass trajectory Chemical composition Emission control Water-soluble ion PM_{2.5}

Pollution Characteristics of PM_{2.5} Aerosol during Haze Periods in Changchun, China

Chunsheng Fang, Zhida Zhang, Meiying Jin, Pengchao Zou, Ju Wang

Source: Volume: 17, Issue: 4, Pages: 888-895 DOI: 10.4209/aaqr.2016.09.0407

To study the pollution characteristics of PM_{2.5} during the haze period in October 2014, the hourly automatic monitoring data and 22-h atmospheric PM_{2.5} samples were collected in Changchun city. According to the statistical results of the hourly average PM_{2.5} concentration, heavy pollution occurred from October 13 to November 1 in 2014 in Changchun city. The daily concentration of PM_{2.5} during this period was 53.18 $\mu\text{g m}^{-3}$ to 450.69 $\mu\text{g m}^{-3}$. In addition, the PM_{2.5} concentration was higher at night and lower during the daytime on haze days, which is affected by pollutant emission and meteorological conditions. The Pearson correlation coefficient between PM_{2.5} and CO was high ($r = 0.745$), which implies that the concentration suddenly increased, probably because of the combustion of fossil fuels and other organic matter. The meteorological condition investigation shows that the wind speed, temperature and pressure are low and that the RH value is relatively high on haze days. In addition, stable weather during haze days makes the pollution heavier. The analysis of water-soluble inorganic ions (WSIIs) suggests that WSIIs constituted up to 21–56% of PM_{2.5}, and the total ions, anions and cations showed a significant correlation. During the haze days, the NO₃⁻/SO₄²⁻ ratio was substantial (3.7–4.6), except at the CP and JZP stations, which suggests that the pollutants in the atmosphere are from a mixture of stationary emission and mobile emission. The NOR and SOR values were more than 0.1 at most sampling stations, so serious secondary pollution occurs in the atmosphere on haze days, and secondary ion formation is enhanced by the atmospheric conditions and emissions of gaseous SO₂ and NO₂.

Two-Step Flotation Treatment for Removal of Toxic Matter from Hospital Solid Waste Incinerator Fly Ash

Han-Qiao Liu, Fang Liu, Guo-Xia Wei, Rui Zhang, Dan-Dan Zang

Source: Volume: 17, Issue: 5, Pages: 1329-1340 DOI: 10.4209/aaqr.2017.02.0090

Application of two-step flotation for removing dioxins and heavy metals from hospital solid waste incinerator (HSWI) fly ash was studied. In the first step, decarburization flotation was executed to remove dioxins from raw fly ash. The second step of precipitation flotation was carried out to recycle the toxic metals in the residual slurry. Finally, an acid leaching-sulphide precipitation-flotation method was proposed. The results showed that carbon removal rate was 90.7%, and the dioxin removal percentage was 91.0% during decarburization flotation. And Zn, Pb and Cu in residual slurry were acid-leached with the removal efficiencies being 91.3%, 79.2% and 85.6%, respectively. The optimum sulphidation condition of heavy metals in the residual slurry were Na₂S molar ratio of 1.4 and sulphidation time of 30 min. Subsequently, precipitation flotation could recover 49.9% of Zn, 42.0% of Pb and 48.7% of Cu from the raw fly ash. After the two-step flotation, the toxicity of the tailings reduced considerably, and the tailings could be disposed in landfill.

Therefore, this two-step flotation treatment of HSWI fly ash may result in both detoxification and potential resource recovery.

Keywords: Incinerator fly ash Two-step flotation Sulphidation Dioxins Heavy metals

Radiative Forcing Estimation of Aerosols at an Urban Site near the Thar Desert Using Ground-Based Remote Sensing Measurements

Sunita Verma, Divya Prakash, Atul Kumar Srivastava, Swagata Payra

Source: Volume: 17, Issue: 5, Pages: 1294-1304 DOI: 10.4209/aaqr.2016.09.0424

The focus of present study is to quantify the radiation budget of aerosols over Jaipur (Northwestern, India) from 2011 to 2015. The Aerosol radiative forcing (ARF) has been determined for shortwave spectrum (0.3–3.0 μm) individually for the top of the atmosphere (TOA), bottom of the atmosphere (BOA) and within the atmosphere (ATM) over study region. Santa Barbara DISORT Atmospheric Radiative Transfer model (SBDART) is used to simulate the aerosols radiative effect.

The inter-annual monthly average of ARF at TOA during 2011–2015 is found between -11.40 to -5.60 W m^{-2} , while the ARF at BOA is found to be between -32.2 to -22.49 W m^{-2} . Likewise, the ARF within the atmosphere (ATM) comes between 14.04 to 22.47 W m^{-2} over Jaipur.

The SBDART model is run discretely for Dust period (DSP) and non-Dust Period (NDP) during the year 2012 to inspect the change in ARF during extreme events over the Jaipur site. During DSP, the net TOA and BOA forcing are found in the range -20.71 to -16.81 W m^{-2} and -45.15 to -39.6 W m^{-2} , respectively, and net ATM forcing varies in the range 22.7 to 24.4 W m^{-2} . For the NDP, the corresponding value varies in the range -10.1 to -6.6 W m^{-2} and -23.6 to -22.3 W m^{-2} . The net ATM forcing during NDP is between 12.2 to 17.05 W m^{-2} . The value of BOA increases more than $\sim 67\%$ during DSP than NDP. The more increase ($-ve$) in surface forcing represents the cooling of the surface during DSP. The results depict that dust over Jaipur in the vicinity of the Thar Desert is scattering in nature with high value (> 0.95) of SSA. The scattering is mostly high during summer and low in winter.

Keywords: Aerosols Dust Radiative forcing AOT SSA

The Association between Intermodal (PM_{1-2.5}) and PM₁, PM_{2.5}, Coarse Fraction and Meteorological Parameters in Various Environments in Central Europe

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Source: Volume: 17, Issue: 5, Pages: 1234-1243 DOI: 10.4209/aaqr.2016.06.0242

Fine and coarse fractions of atmospheric aerosol overlap in the particle size range of about $1\text{--}2.5 \mu\text{m}$ (aerodynamic diameter). Sources of both fractions contribute to PM_{1-2.5} to different extents due to meteorological and spatial conditions. Therefore, there is ongoing discussion as to whether

PM_{2.5} or PM₁ should be included for monitoring as a fine particulate pollutant by the national ambient air quality standard (NAAQS). The aim of the presented study is to examine the association between the intermodal and PM₁, PM_{2.5}, coarse fraction, and meteorological parameters in various environments. Outdoor 24-h mass concentrations of size-resolved PM and meteorological conditions were measured at 12 sites within 42 campaigns between 11/2005 and 3/2015. The data set was divided into 10 environments reflecting season, locality, total measured PM, and placement of the impactor. We used two types of statistic methods: nonparametric correlation analysis and multiple linear regression (MLR). Median PM_{1-2.5} in PM₁₀ or TSP percentages were 7% and 6% in summer and 7% and 9% in winter. On the other hand, PM_{1-2.5} accounted for a higher mass portion of PM_{2.5} during summer. Stronger positive correlation and relationship were identified between PM_{1-2.5} and the coarse fraction than between PM_{1-2.5} and PM₁ in all environments. MLR confirmed the dependence of PM_{1-2.5} on PM₁ in only 3 environments. This study found that PM_{1-2.5} in Central Europe represents mostly the “tail” of the coarse mode and probably has the same sources. Therefore, PM₁ should be considered by the NAAQS as a fine particulate pollutant in Central Europe.

Keywords: Intermodal fraction Personal cascade impactor sampler PM_x Rural aerosol Urban aerosol

Criteria Pollutants and Volatile Organic Compounds Emitted from Motorcycle Exhaust under Various Regulation Phases

Jiun-Horng Tsai, Yung-Chen Yao, Pei-Hsiu Huang, Hung-Lung Chiang

Source: Volume: 17, Issue: 5, Pages: 1214-1223 DOI: 10.4209/aaqr.2016.04.0155

Establishment of emission standards is an important measure for controlling vehicle exhaust. This study examined the emission factors of air pollutants from 40 four-stroke motorcycles of various emission standard phases, ages, and mileage. Based on the emission standards, the motorcycles were divided into three groups (Phases III, IV and V). Regulated air pollutants (CO, HC, and NO_x), CO₂, and 52 volatile organic compounds were evaluated on a chassis dynamometer using the Economic Commission for Europe (ECE) test cycle. The sequence of CO and HC emission factors was Phase III > Phase IV > Phase V, and their ratios of emission factor of Phase IV to Phase III and Phase V to Phase III were 0.66 and 0.42 for CO and 0.61 and 0.57 for HC, respectively. Exhaust from motorcycles deteriorates with age and mileage. For NO_x emission, the sequence of emission factor was Phase V > Phase IV > Phase III. However, the relationship was insignificant between CO₂ emission factor and motorcycle age. The total VOC emissions of Phase V motorcycles were the lowest (0.59 g km⁻¹) among all test motorcycles; however, the fraction of VOC groups was similar among all test motorcycles regardless of different regulation phases. For organic air toxics, the emissions of benzene, toluene, ethylbenzene, and xylene (BTEX) decreased by 37–58% and 44–62%, respectively, for Phases IV and V motorcycles compared to those of Phase III motorcycles. Results also indicated that the ozone formation potential (OFP) was high in older motorcycles with high mileage. In summary, emissions of CO, HC, total VOCs, BTEX, and OFP may decrease with the decrease of motorcycle age and mileage as well as the phase of emission standards. The results implied that tightening emission standards indeed encourages motorcycle manufacturers to

improve engine technology and combustion efficiency, resulting in reduced emission of air pollutants, except NO_x emission in this study.

Keywords: Volatile organic compounds (VOCs) Organic air toxics ozone formation potential (OFP) Motorcycle deterioration

Assessment of Air Pollution around the Panzhihua V-Ti Magnetite Mine Region, Southwest China

Xin Cheng, Yi Huang , Chao Liu, Shi-Jun Ni, Rui Wang, Zhi-Jie Long

Source: Volume: 17 , Issue: 5 , Pages: 1204-1213 DOI: 10.4209/aaqr.2016.10.0452

This study investigated variations in air quality by evaluating trace gases, inhalable particulate matter (PM₁₀), and associated trace elements at three sites in Panzhihua (a mining city located in Panxi Rift Valley, Southwest China) between January and December 2014. The concentrations of 19 trace elements in PM₁₀ were determined through inductively coupled plasma mass spectrometry. Single particle morphology and chemical composition were determined through scanning electron microscopy with energy-dispersive X-ray analysis to identify their possible sources. Mean sulfur dioxide, nitrogen dioxide, and carbon monoxide concentrations were highest near the steel smelting district, whereas ozone concentrations were highest in the residential region. Annual mean concentrations of PM₁₀ at three sites were 129.4, 165.5, and 187.2 $\mu\text{g m}^{-3}$; all these exceed the annual mean (70.0 $\mu\text{g m}^{-3}$) of the National Ambient Air Quality Standard. In addition, the concentrations of trace elements in PM₁₀ exhibited significant spatial and seasonal variations at the three sites. The mean concentrations of trace elements in PM₁₀ were in the order of Fe > Ti > Zn > Pb > Cu > Mn > Ba > V > Cr > Ni > Sr > Bi > Cd > As > Co > Sb > Sc > Tl > U. The enrichment factor values of the trace elements suggested that anthropogenic activities were the dominant sources of As, Cd, Sb, Ti, Tl, Zn, Cu, Pb, and Bi. Particle morphology and chemical composition analysis revealed five major particle types, namely aluminosilicate, Fe-containing, mineral, soot, and Ca-containing particles.

Keywords: Gaseous pollutants PM₁₀ Trace elements Enrichment factor (EF) Scanning electron microscopy with energy-dispersive X-ray analysis (SEM-EDX)

Study of Local and Regional Influence on PM_{2.5} Concentration during Odd-Even Rule in Delhi Using Causal Analysis

Asha B. Chelani

Source: Volume: 17 , Issue: 5 , Pages: 1190-1203 DOI: 10.4209/aaqr.2016.06.0267

PM_{2.5} concentration observed during odd-even rule in Delhi is analysed for assessing its effectiveness in curbing the levels. The local and regional influence is analysed by using similarity

and causality analysis. Causality analysis is usually carried out by using nonlinear dynamical technique which predicts one variable using another. In this study a simple approach is presented based on nearest neighbour method. It is observed that PM_{2.5} in Delhi has regional influence in addition to local sources. Although the effectiveness of odd-even rule is not observed in curbing the PM_{2.5} levels, it is suggested that extended implementation of the rule may provide more insight to the impact. Similarity analysis suggested that PM_{2.5} concentrations in Delhi have somewhat similar temporal behaviour with neighbouring locations in the southeast (SE) and west (W)-southwest (SW) sector. The control policies in Delhi need to be adopted keeping in mind the local and regional influences on PM_{2.5} levels in the area.

Keywords: Causal analysis Similarity analysis Nearest neighbour Local and regional influences Odd-even rule

Investigation of Particulate Matter Regional Transport in Beijing Based on Numerical Simulation

Jianjun He , Hongjun Mao , Sunling Gong, Ye Yu, Lin Wu, Hongli Liu, Ying Chen, Boyu Jing, Peipei Ren, Chao Zou

Source: Volume: 17, Issue: 5, Pages: 1181-1189 DOI: 10.4209/aaqr.2016.03.0110

The frequent occurrence of regional air pollution makes it challenging to control. Based on source sensitivity research performed with the Chinese Unified Atmospheric Chemistry Environment (CUACE) model and dispersion simulation performed with the Flexible Particle dispersion model (FLEXPART), the regional transport of particulate matter (PM), potential source regions, and transport pathways were investigated for Beijing in summer (July) and winter (December) 2013. The mean near-surface trans-boundary contribution ratio (TBCR) of PM_{2.5} in Beijing was 53.4% and 36.1% in summer and winter 2013, respectively, and 51.8% and 35.1% for PM₁₀. Regional transport in summer was more significant than that in winter. Seasonal difference of meteorological condition combined with the distribution of emission is responsible for seasonal difference of TBCR. The secondary aerosol is mostly contributed by regional transport. The transport of PM is mostly from Hebei province and Tianjin municipality. Based on backward trajectories analysis, the air mass source occurred from different directions in summer, while occurred from northwest in winter. The pollution level and the TBCR were closely related to the transport pathways and distance, especially in summer.

Keywords: Regional transport Particulate matter Backward trajectory CUACE FLEXPART

Chemical Characteristics of PM_{2.5} during 2015 Spring Festival in Beijing, China

Yangyang Zhang, Jiaming Wei, Aohan Tang , Aihua Zheng, Zexi Shao, Xuejun Liu

Source: Volume: 17, Issue: 5, Pages: 1169-1180 DOI: 10.4209/aaqr.2016.08.0338

Air pollution especially of PM_{2.5} pollution is a serious problem in Beijing. In order to quantify the effect of a festival in which pollution was expected to be reduced, we collected and analyzed PM_{2.5} samples in urban Beijing during the 2015 Spring Festival (from February 9th to March 6th 2015). We divided the sampling period into three phases: non-haze, haze and firework days. The average concentration of PM_{2.5} was highest on firework days at 248.9 $\mu\text{g m}^{-3}$, followed by haze days (199.9 $\mu\text{g m}^{-3}$) and non-haze days (90.8 $\mu\text{g m}^{-3}$). The air quality of non-haze days during the holiday was better than that during non-holiday periods. Secondary inorganic ions (SO_4^{2-} , NO_3^- and NH_4^+) were enriched on the haze days, while those on firework days contained large amounts of Cl^- and K^+ , but small amounts of NO_3^- and NH_4^+ . Ratios of $\text{NO}_3^-/\text{SO}_4^{2-}$, $\text{SO}_4^{2-}/\text{K}^+$ and Cl^-/K^+ effectively distinguished the characteristics of PM_{2.5} between firework events and haze days. Ion balance calculations indicated that the acidity of PM_{2.5} from firework days was higher than that from haze and non-haze days. A method using enrichment factors (EF) found that crustal elements (EF < 10 on all three types of days) included Ca, Al, Fe, Na, Co, Ni, P, Ti, and V; firework elements (EF > 10 on firework days, significantly higher than haze days) were made up of Ba, Cr, Cu, Mg, Pb, S, Si, and Zn; common anthropogenic pollution elements (EF > 10 in all three types of days) were As, Cd, Cu, Pb, S, Sb, Zn. Differences in chemical characteristics indicated that holidays such as the Spring Festival can affect air pollution patterns in two ways: a decrease in the population and vehicles but an increase in activities such as firework displays.

Keywords: PM_{2.5} Chemical characteristics Fireworks Haze Spring Festival

Mass Concentration and Size-Distribution of Atmospheric Particulate Matter in an Urban Environment

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Source: Volume: 17 , Issue: 5 , Pages: 1142-1155 DOI: 10.4209/aaqr.2016.08.0344

To investigate the ambient mass concentration, size-distribution and temporal variability of atmospheric particulate matter (PM), a long-term monitoring campaign was undertaken at an urban background site in Como, Northern Italy, from May 2015 to March 2016. A 13-stage Low Pressure Impactor (DLPI) was used for the collection of size-segregated particulates in the 0.028–10 μm size range. The results revealed a good level of agreement between DLPI and a co-located Harvard-type PM_{2.5} Impactor, allowing them to be classified as comparable and characterized by a reciprocal predictability. The PM concentration levels varied greatly between the different 5-days monitoring sessions, with higher mean mass concentrations during the heating season. Appreciable seasonal differences were found for particles between 0.15 and 1.60 μm that, on average, registered concentration levels 3.5 times higher during the heating season (mean: 28.2 $\mu\text{g m}^{-3}$; median: 24.4 $\mu\text{g m}^{-3}$) compared to the non-heating season (mean: 8.3 $\mu\text{g m}^{-3}$; median: 7.6 $\mu\text{g m}^{-3}$). No relevant and significant differences were detected for the coarser ranges (> 1.60 μm). Temporal variabilities were influenced by typical PM urban sources (e.g., household heating, traffic), that significantly affected fine and submicrometer particles, and were related to meteorological factors. Ambient air particles exhibited a trimodal distribution: a first and sharp peak more pronounced during the heating season was identified between 0.3 and 0.5 μm and two other slight peaks in the coarse mode were centered on approximately 3 and 8 μm . No relevant differences were found in the shape

of the size-distribution between the two investigated seasons. The mean PM_{2.5} (22.4 $\mu\text{g m}^{-3}$) and PM₁₀ (27.7 $\mu\text{g m}^{-3}$) concentrations monitored in the study area exceeded the annual Air Quality Guideline Values (respectively equal to 10 $\mu\text{g m}^{-3}$ and 20 $\mu\text{g m}^{-3}$) established by the World Health Organization.

Keywords: Size-segregated particles DLPI performance Temporal variability Mass size-distribution Heating and non-heating season

Emissions of Polycyclic Aromatic Hydrocarbons and Particle-Bound Metals from a Diesel Engine Generator Fueled with Waste Cooking Oil-Based Biodiesel Blends

Sheng-Lun Lin, Jen-Hsiung Tsai, Shui-Jen Chen, Kuo-Lin Huang, Chih-Chung Lin, Ho-Tsang Huang, Yi-Chin Hsieh, Chuen-Huey Chiu

Source: Volume: 17, Issue: 6, Pages: 1679-1689 DOI: 10.4209/aaqr.2017.04.0151

This study investigates the emission of a heavy-duty diesel engine generator fueled with waste cooking oil (WCO)-based biodiesel blends (W) and operated at 1.5 and 3.0 kW loads. A brand of pure fossil diesel was adopted as the base fuel, with 20% and 40% WCO-based biodiesel added into the based fuel to form W20 and W40 blends, respectively. The emission characteristics of PM, metals and PAHs were analyzed. Experimental results indicate that alternative WCO-based fuels had slightly higher fuel consumption rates (FCR) and brake specific fuel consumptions (BSFC) than conventional diesel (0.6–4.1% for FCR and 1.0–7.6% for BSFC), and similar engine thermal efficiency. The PM emissions reductions when using W20 and W40 were 19% and 6.5%, respectively, at 1.5 kW, and 27% and 19%, respectively, at 3.0 kW. The emissions of particle-bound metals were 13.6–13.8% lower when using W20 than using conventional diesel, but 12.0–12.3% higher when using W40. The metal contents of PM rose with the addition of WCO-based biodiesel. The metal elements of PM were dominated (> 90% mass) by Na, Mg, Al, K, Ca, Fe and Zn, while the major trace metals were Mn, Cu, Sr and Pb. The use of WCO-based biodiesel blends reduced the emissions of total-PAHs (44.0% in average) and total-BaP_{eq} (80.2% in average). The mass reductions of MMW- and HMW-PAHs using W20 and W40 were more significant at 3.0 kW than at 1.5 kW, while the reduction of LMW-PAHs was greater at 1.5 kW than at 3.0 kW. Thus, the reduction in total-BaP_{eq} was greater at the higher engine load. Accordingly, we conclude that the WCO-based biodiesel is a potential candidate of cleaner alternative energy sources.

Keywords: Polycyclic aromatic hydrocarbons Particle bound metals Waste cooking oil Biodiesel Diesel engine generator

Multiplier Effects of Energy Consumption and CO2 Emissions by Input-Output Analysis in South Africa

Mohamed Beidari, Sue-Jane Lin , Charles Lewis

Source: Volume: 17 , Issue: 6 , Pages: 1666-1678 DOI: 10.4209/aaqr.2017.04.0150

This paper analyzed the energy consumption and CO2 emission from 18 industrial sectors, and also evaluated the direct and indirect energy consumption and CO2 emission of changes in the final demand of South Africa's (SA) economy. To accomplish this goal, the input-output linkage and multiplier methods have been applied to investigate the interconnectedness of the 18 sectors' input-output tables for the years 1995, 2000, 2005, 2010 and 2012, and to measure their total impact of energy commodity input coefficients and CO2 emissions output coefficients for the year 2012. Results revealed that the electricity sector has a weak linkage with others sectors, which means it is mostly independent of other sectors. In another words, it does not induce and enable economic growth. Moreover, two sectors, such as Chemical and Petrochemical Industries and Basic Metals, were found as key sectors in SA's economy in 1995, 2000 and 2012. In 2005 and 2010, only Chemical and Petrochemical Industries was the most important sector in SA. Additionally, Commercial and Public Services was the strongest forward linkage sector in SA. Our findings also showed that the electricity sector was the main direct monetary energy consumer and CO2 emitter, and therefore the most dominant source in terms of energy and CO2 intensities among all the 18 sectors in SA. Furthermore, our investigation of the direct and indirect effects on energy consumption and CO2 emissions indicated that both total of direct energy consumption and CO2 emissions were higher than both total indirect energy consumption and CO2 emissions. Finally, some potential suggestions on reducing the energy consumption and CO2 emissions deduced from this study are discussed.

Keywords: Input-output analysis Linkage and multiplier effects Energy consumption CO2 emission Electricity production sector

Economic Growth, Carbon Abatement Technology and Decoupling Strategy – The Case of Taiwan

Chien-Ming Lee, Ever Romel Rosalez

Source: Volume: 17 , Issue: 6 , Pages: 1649-1657 DOI: 10.4209/aaqr.2016.11.0487

Greenhouse Gas (GHG) emissions decoupling from economic growth are imperative goals for sustainable development. This study combines decoupling index and Log Mean Divisia Index (LMDI) to study which major transformation is required in the way energy is produced, delivered and consumed in order to achieve decoupling in Taiwan. The results indicate that a high-energy price can improve the energy structure by inciting energy efficiency use and result in decoupling CO2 emissions from economic growth. Targeting CO2 emissions through early action is the best approach to acquire decoupling. An annual energy intensity decrease of 2.4% is key for Taiwan to achieve absolute decoupling by 2020. The study suggests that the Taiwan government should focus

on energy efficiency through investing in clean energy innovation at an early phase. Taiwan should consider national policies that are sensitive to effective economic strategies that enhance research and development and also invest in promoting energy efficiency in the economy-wide.

Keywords: Decoupling Energy efficiency Economic growth Early action CO2 emissions

Influences of Copper(II) Chloride Impregnation on Activated Carbon for Low-Concentration Elemental Mercury Adsorption from Simulated Coal Combustion Flue Gas

Cheng-Yen Tsai, Chun-Hsiang Chiu, Ming-Wei Chuang, Hsing-Cheng Hsi

Source: Volume: 17, Issue: 6, Pages: 1637-1648 DOI: 10.4209/aaqr.2016.10.0435

In this study, the Hg⁰ adsorption equilibrium and kinetics of a coconut-shell-based activated carbon impregnated with CuCl₂ were examined with respect to their resulting physical and chemical properties. Integrating the results from N₂ adsorption isotherm at 77 K, scanning electron microscopy, elemental analysis, X-ray photoelectron spectroscopy, and Hg⁰ adsorption experiments under N₂ and simulated coal-combustion flue gases conditions, it was found that HCl pretreatment could enhance Hg⁰ adsorption of crude activated carbon; the Hg⁰ adsorption capacities of crude and HCl-pretreated activated carbon under N₂ condition were 95.8 and 225.4 µg g⁻¹, respectively. Additionally, CuCl₂ impregnation further increased the adsorption capacity of crude. The Hg⁰ adsorption capacity of crude activated carbon with 8% CuCl₂ impregnation was 631.1 µg g⁻¹. However, the equilibrium Hg⁰ adsorption capacity decreased when Cu loading exceeded 8 wt%, suggesting that adequate forms of surface Cu, O and Cl interacting with flue gas components and Hg⁰, as well as the presence of pores with specific size ranges allowing rapid transport of the Hg molecules into the interior of the activated carbon and as energy sinker govern the overall chemisorption process. Pseudo-second-order kinetic model could best describe the adsorption behaviors of tested samples under both test conditions, indicating that Hg⁰ adsorbed on the activated carbon surface could be explained by bimolecular reaction mechanisms.

Keywords: Mercury Adsorbent Impregnation CuCl₂ Coal-combustion flue gas

Analysis of Long-Range Transport Effects on PM_{2.5} during a Short Severe Haze in Beijing, China

Weilin Yang, Guochen Wang, Chunjuan Bi

Source: Volume: 17, Issue: 6, Pages: 1610-1622 DOI: 10.4209/aaqr.2016.06.0220

Comprehensively using Inverse Distance Weighted (IDW) analysis, Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model, three-dimensional (3D) cluster analysis, Weight Potential Source Concentration Function (WPSCF) analysis and other statistic methods, we

mainly studied about the spatio-temporal variation, long-range transport and potential source regions of PM_{2.5} in Beijing during a short severe haze from Dec05 to Dec11, 2015. The results showed that the concentration of PM_{2.5} decreased from south to north of Beijing. PM_{2.5} accumulation in the short-severe haze had high correlation with calm and steady meteorological condition (high relative humidity (RH), low wind speed (WS), low boundary-layer temperature (BLT) and surface air pressure (SAP)). In addition, air-flow in different heights (500 m, 1500 m and 3000 m) had different effects on the haze episode and the air flows at 500 m had the greatest contribution of the air pollution. The potential sources were mainly from the desert in northwest of Beijing and the built-up areas in Jing-Jin-Ji zone. Higher WPSCF values (> 0.7) were mainly distributed in Hebei, west Shandong province (around 0.5) and south Tianjin (around 0.5).

Keywords: PM_{2.5} Spatio-temporal distribution Meteorological parameters HYSPLIT Potential sources

The Effects of Height on the Accumulation of n-Alkanes and Polycyclic Aromatic Hydrocarbons (PAHs) in Air-Conditioning Filter Dust from High-Rise Apartments

Ji-Zhong Wang , Cheng Qian, Jia-Qin Liu, Xiu-Feng Ni, Shu-Chuan Peng

Source: Volume: 17, Issue: 6, Pages: 1582-1591 DOI: 10.4209/aaqr.2017.03.0119

To assess the effect of floor levels of high-rise apartment buildings on the accumulation of contaminants in indoor environment, residential air-conditioner filter dust (ACFD) samples from the 1st, 10th, 20th and 30th floors of a high-rise apartment building were collected for the determination of n-alkanes and polycyclic aromatic hydrocarbons (PAHs). The results show that both n-alkanes and PAHs in the residential ACFD were ubiquitous but varied greatly in concentrations. The total concentrations of 27 n-alkanes ($\Sigma 27AK$) and 16 PAHs ($\Sigma 16PAH$) ranged from 1.35 to 9290 $\mu\text{g g}^{-1}$ and 278–34200 ng g^{-1} , respectively. Source apportionment revealed that n-alkanes were from mixed sources combining fossil fuel combustion, natural emission and solid biomass burning, but PAHs were mainly from indoor sources. Furthermore, the diagnostic ratio of paired low weight molecular PAH species may change during transportation and accumulation. Significantly higher concentrations of $\Sigma 27AK$ and $\Sigma 16PAH$ were observed in the samples from low floor levels (the 1st and 10th floors) compared to those from high floor levels (the 20th and 30th floors). The results of classification and regression tree analysis clearly suggested floor level is the most important factor influencing the accumulation of $\Sigma 27AK$ and $\Sigma 16PAH$ in the ACFD. Our findings imply that people living on lower floor levels have greater exposure risks to PAHs associated with indoor dust.

Keywords: High-rise apartment Floor level Air-conditioner filter dust n-alkanes PAHs

Identification and Characterization of Size-Segregated Bioaerosols at Different Sites in Delhi

Himanshu Lal, Bipasha Ghosh, Anshul Srivastava, Arun Srivastava

Source: Volume: 17, Issue: 6, Pages: 1570-1581 DOI: 10.4209/aaqr.2015.05.0331

Ambient levels of culturable bioaerosol were measured at four different sites of Delhi, India in six size ranges ($> 7.0 \mu\text{m}$, $7.0\text{--}4.7 \mu\text{m}$, $4.7\text{--}3.3 \mu\text{m}$, $3.3\text{--}2.1 \mu\text{m}$, $2.1\text{--}1.1 \mu\text{m}$, $< 1.1 \mu\text{m}$). The study also accounted the seasonal variation (monsoon, post monsoon, winter and pre-monsoon) of the air microbes. The sampling was carried out for three different fractions of bioaerosols viz. fungi, gram positive and negative bacteria during August 2010 to April 2011 using a six-stage viable cascade impactor sampler. Unlike gram positive and negative bacteria, the concentration of fungal bioaerosol found in different stages at each site seems to follow a typical pattern in all four season. The typical pattern of concentration depicts that majority of the fungal species found in the diameter range of $3.3\text{--}2.1 \mu\text{m}$, which coincides with the penetration range in the secondary bronchi of the lungs in the human body. This reveals that majority of the immunotoxic and allergic fungi found at this stage are mostly prone to affect the secondary bronchi in human lungs when inhaled. At all four sites maximum fungal concentration ($1740.5\text{--}3224.7 \text{ CFU m}^{-3}$), gram-positive bacterial concentration ($2790.6\text{--}9428.3 \text{ CFU m}^{-3}$) and gram-negative bacterial concentration ($1990.3\text{--}7609 \text{ CFU m}^{-3}$) were found in post monsoon season. In the majority of the sites, minimum concentrations were found in monsoon period which probably may be due to rain wash during the sampling. For all the three bioaerosol fractions no particular relationship pattern was found to exist between their respective concentrations with temperature and relative humidity (RH). However, higher range of variation was observed at higher concentration levels and lower range of variation at low concentration levels for all the three bioaerosol fractions. Most of the fungal bioaerosol identified such as *Penicillium* sp., *Alternaria* sp. and *Aspergillus* sp. are associated with immunotoxic and allergic diseases.

Keywords: Bioaerosol Air Pollution Fungi Bacteria Delhi

Part I: PM_{2.5} and Polychlorinated Dibenzo-p-dioxins and Dibenzofurans (PCDD/Fs) in the Ambient Air of Southern China

Haiyan Tang, Kangping Cui, Jin Xing, Jinning Zhu, Wen-Jhy Lee, John Kennedy Mwangi, Yu-Cheng Lee

Source: Volume: 17, Issue: 6, Pages: 1550-1569 DOI: 10.4209/aaqr.2017.03.0117

The atmospheric PM_{2.5}, PM_{2.5}/PM₁₀, PCDD/Fs-WHO2005-TEQ, and PCDD/F (polychlorinated dibenzo-p-dioxins and dibenzofuran) phase distributions of 23 cities in southern China, during 2014–2016, were investigated in this study. In general, the cities with higher latitudes had higher PM_{2.5} concentrations than those with lower latitudes. During 2014–2016, the lowest three-year average concentrations of PM_{2.5} occurred at Sanya and Haikou and were 16.4 and $21.7 \mu\text{g m}^{-3}$, respectively; while the highest concentrations of PM_{2.5} was occurred at Wuhan and Luzhou and were 68.8 and $63.1 \mu\text{g m}^{-3}$, respectively. During 2015–2016, the PM_{2.5} concentrations of most of

cities decreased, but those of five cities (Chengdu, Luzhou, Nanchang, Qujing and Quanzhou) increased, indicating that the air quality of these five cities was still not well controlled. The average RM values of the 23 cities were 5.20, 4.49 and 4.13 in 2014, 2015 and 2016, respectively, which revealed that the PM_{2.5} concentrations in the cities of southern China slowly decreased, although they were still far above the WHO air quality regulated standard (10 µg m⁻³). In general, a city with a higher PM_{2.5} concentration was also had a higher PM_{2.5}/PM₁₀ ratio. Among the 23 cities, the six highest three-year averages of total-PCDD/Fs-WHO2005-TEQ concentrations were 0.0665, 0.0633, 0.0625, 0.0600, 0.0528 and 0.0526 pg-WHO2005-TEQ m⁻³ in Chengdu, Wuhan, Nanjing, Hefei, Luzhou and Hangzhou, respectively. During 2014, the six cities (Hefei, Nanjing, Wuhan, Guiyang, Shanghai and Chengdu) with the lowest temperatures in winter (an average of 5.4°C), their average particle phase fractions of total-PCDD/Fs-WHO2005-TEQ that were approximately 76%, 53%, 71% and 93% in the spring, summer, fall and winter, respectively; while, the six cities (Haikou, Fuzhou, Guangzhou, Nanning, Nanchang and Changsha) with the highest temperatures in summer (an average of 16.5°C), had average particle phase fractions of total-PCDD/Fs-WHO2005-TEQ that were approximately 61%, 42%, 57% and 81% in the spring, summer, fall and winter, respectively. The results of this study provide information showing the trends of both atmospheric PM_{2.5} and PCDD/Fs in the cities of southern China. In addition, this study provided the overview relating to the PM_{2.5} and PCDD/Fs in ambient air of southern China, which was not reported in previous studies. The results of this study were of great importance to present the trends of air quality in China. It is also useful for the establishment of control strategies in the future.

Keywords: PM_{2.5} PCDD/Fs PM_{2.5}/PM₁₀ ratio Phase distribution Particle-bound Southern China

Optical Properties, Chemical Composition and the Toxicological Potential of Urban Particulate Matter

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Source: Volume: 17, Issue: 6, Pages: 1515-1526 DOI: 10.4209/aaqr.2016.09.0395

This paper discusses the diurnal variation and the interdependences between the physical, chemical and toxicological characteristics of atmospheric carbonaceous particulate matter (CPM) and co-emitted gaseous components. Measurements were carried out at two different urban sites during a 2-month period. On-line measured parameters were optical absorption coefficients (OAC), total number concentration (TNC), mass concentration of CPM and the concentration of gaseous species (CO, NO_x and BTEX). Off-line analyses were carried out on filters collected with 6-hour time resolution. The concentrations of elemental carbon (EC), organic carbon (OC), total carbon (TC), levoglucosan (LG) and polycyclic aromatic hydrocarbons (PAH) were determined. The ecotoxicity of CPM was assessed by the *Vibrio fischeri* marine bioluminescence inhibition bioassay (ISO 21338:2010). We found ($r > 0.498$) positive and wavelength dependent correlation between the CPM related parameters based on optical response (OAC, AAE) and thermal stability (TC, EC, OC, OC/TC). We also revealed weak ($r = 0.309$) or moderate ($r = 0.448$) correlation between the AAE and the ecotoxicity data indicating that carbonaceous fraction of the ambient particulate matter has ecotoxicological impact. Based on the determined correlations, we propose the applicability of the

AAE determined by multi wavelength photoacoustic measurements as a possible candidate for first-screening the toxicological impact of optically active carbonaceous ambient particulate matter. The strengths and the limitations of this methodology are both discussed here.

Keywords: Aerosol Angström Exponent Ecotoxicity Photoacoustic spectroscopy Polycyclic aromatic hydrocarbons

Application of WRF Model for Air Quality Modelling and AERMOD – A Survey

Awkash Kumar , Rashmi S. Patil, Anil Kumar Dikshit, Rakesh Kumar

Source: Volume: 17 , Issue: 7 , Pages: 1925-1937 DOI: 10.4209/aaqr.2016.06.0265

Meteorology plays a crucial role in air quality. The presence of uncertainties of a significant nature in the meteorological profile used during air quality model simulation has the potential to affect negatively the results of the simulations. This paper describes a most recent version of the meteorological model called Weather Research and Forecasting (WRF) model and its importance in air quality. The performance of WRF depends upon the intended application and parameterization scheme of physics options. WRF model is also applied to investigate the simulation results with various land surface models (LSMs) and Planetary Boundary Layer (PBL) parameterizations and various set of microphysics options. It predicts various meteorological spatial parameters like mixing layer height, temperature, humidity, rain fall, cloud cover and wind. The WRF results are integrated with air quality model (AQM) and the AQM depends upon the performance of WRF. It has been applied for evaluation of national pollution control policy, behaviour of plume rise, property of aerosols, prediction of Ozone, SO₂, NO_x, PM₁₀, PM_{2.5} etc. using AQM for various sources. The effect of topography and different seasons on the concentration of pollutants in the atmosphere has also been studied using AQM. AQM AERMOD has also been reviewed with various other AQM models such as ADMS-Urban and CALPUFF. AERMOD has been used for different time scales, health risk assessment, evaluation of various control strategies, Environmental Impact Assessment (EIA) studies and emission factor estimation. This paper presents the importance of meteorological model to AQM as well as many applications of AQM to demonstrate various scientific questions and policies.

Keywords: Meteorological model WRF model Air quality modeling AERMOD Urban region Atmospheric dispersion

Evaluation of Model Performance for Forecasting Fine Particle Concentrations in Korea

Young Sung Ghim , Yongjoo Choi, Soontae Kim, Chang Han Bae, Jinsoo Park, Hye Jung Shin

Source: Volume: 17 , Issue: 7 , Pages: 1856-1864 DOI: 10.4209/aaqr.2016.10.0446

The performance of a modeling system consisting of WRF model v3.4.1 and CMAQ model v4.7.1 for forecasting fine particle concentrations were evaluated using measurement data at the surface. Twenty-four hour averages of PM_{2.5} and its major components at Bulgwang (located in the northwest of Seoul) during the period February 2012 through January 2013 were compared with predicted concentrations as well as hourly averages of inorganic ions measured at Yongin (located to the southeast of Seoul) in spring 2012. The mean fractional bias (MFB) of -0.37 for PM_{2.5} at Bulgwang fell just outside the goal of -0.3, the level of accuracy that the best model can be achieved. Negative values of MFB, especially in winter, along with the correlation coefficient of 0.61 between measured and predicted concentrations showed that the model performance at Bulgwang was closer to that for Europe than that for North America. However, underestimation of SO₄²⁻ and overestimation of NO₃⁻ were similarly observed at Bulgwang as in the United States. Although diurnal variations in the measured values showed distinctive features at Yongin according to the classified patterns, most variations in the predicted values typically showed a peak early in the morning followed by an increase at night.

Keywords: CMAQ/WRF Mean fractional bias PM_{2.5} Major components Temporal variations

Emission Characteristics of Gas-Fired Boilers in Beijing City, China: Category-Specific Emission Factor, Emission Inventory, and Spatial Characteristics

Xiao Yan, Guangwu Song , Jing Yan , Zhiyun Luo, Xuesong Sun, Chunwang Wei, Rui Zhang, Guohao Li, Qi Ding, Di Zhang

Source: Volume: 17 , Issue: 7 , Pages: 1825-1836 DOI: 10.4209/aaqr.2016.11.0506

Gas-fired boilers are the main stationary sources of NO_x in Beijing. However, the understanding of gas-fired boilers is limited. In the present study, the emission characteristics of NO_x, SO₂, and CO from gas-fired boilers in Beijing were established using category-specific emission factors (EFs) from field measurements. To obtain category-specific EFs, boilers were classified through influence analysis. Factors such as combustion mode, boiler type, and installed capacity were considered critical for establishing EFs because they play significant roles in pollutant formation. The EFs for NO_x, CO, and SO₂ ranged from 1.42–6.86 g m⁻³, 0.05–0.67 g m⁻³ and 0.03–0.48 g m⁻³. The emissions of NO_x, SO₂, and CO for gas-fired boilers in Beijing were 11121 t, 468 t, and 222 t in 2014, respectively. The emissions were spatially allocated into grid cells with a resolution of 1 km × 1 km, and the results indicated that top emitters were in central Beijing. The uncertainties were quantified using a Monte Carlo simulation. The results indicated high uncertainties in CO (-157% to 154%) and SO₂ (-127% to 182%) emissions, and relatively low uncertainties (-34% to 34%) in NO_x emission. Furthermore, approximately 61.2% and 96.8% of the monitored chamber combustion boilers (CCBs) met the standard limits for NO_x and SO₂, respectively. Concerning NO_x, low-NO_x burners and NO_x emission control measures are urgently needed for implementing of stricter standards. Adopting terminal control measures is unnecessary for SO₂, although its concentration occasionally exceeds standard limits, because reduction of its concentration can be achieved thorough control of the sulfur content of natural gas at a stable low level. Furthermore, the atmospheric combustion boilers (ACBs) should be substituted with CCBs, because ACBs have a higher emission despite lower gross installed capacity. The results of this study will enable in understanding and controlling emissions from gas-fired boilers in Beijing.

Keywords: Classification of boilers Uncertainty analysis GIS-based approach Operating load Monte Carlo simulation

PM2.5-Bound Polycyclic Aromatic Hydrocarbons (PAHs), Oxygenated-PAHs and Phthalate Esters (PAEs) inside and outside Middle School Classrooms in Xi'an, China: Concentration, Characteristics and Health Risk Assessment

Jingzhi Wang, Benjamin Guinot, Zhibao Dong, Xiaoping Li, Hongmei Xu, Shun Xiao, Steven Sai Hang Ho, Suixin Liu, Junji Cao

Source: Volume: 17, Issue: 7, Pages: 1811-1824 DOI: 10.4209/aaqr.2017.03.0109

In China, the exposure of children to particulate toxics, like organics, has been poorly investigated mainly due to the technical challenges in sampling and analysis. This article reports indoor and outdoor concentrations of PM2.5-bound polycyclic aromatic hydrocarbons (PAHs), oxygenated-PAHs (OPAHs) and phthalate esters (PAEs) monitored for 13 days in May 2012 in two classrooms, A and B, of a middle school at Xi'an, China. Outdoors, the average PM2.5 mass was 96.9 $\mu\text{g m}^{-3}$, while indoor concentrations ranged between 154.7 $\mu\text{g m}^{-3}$ (A) and 120.2 $\mu\text{g m}^{-3}$ (B). Total PAEs, dominated by bis(2-ethylhexyl)phthalate (DEHP) and di-n-butyl phthalate (DBP), were found at much higher concentrations than PAHs and OPAHs, and their outdoor versus indoor distribution followed that of PM2.5, ranging from 622.0 ng m⁻³ outdoors, to 808.6 (A) and 864.7 ng m⁻³ (B) indoors. Concentrations of total PAHs were about 50 ng m⁻³ outdoors and indoors, while OPAHs were observed at concentrations of 17.7 outdoors and 15.9 (A) and 19.8 ng m⁻³ (B) indoors. High molecular weight PAHs (i.e., 4-ring, 5-ring and 6-ring) generally accounted for about 80%. Variations of PAHs levels indoors were closely associated with the ventilation and the occupancy rate of the classrooms. Activities on the playground also influenced the indoor organic pollutant concentrations. Intense PAEs sources were evidenced, but outdoor sources also influenced the I/O ratios. Both the PAHs and PAEs inhalation risk estimations demonstrated that there is a non-negligible potential cancer risk for children in their school environment.

Keywords : Indoor/Outdoor PAHs/OPAHs/PAEs PM2.5 Schoolchildren Health risks

Atmospheric Deposition of Polychlorinated Dibenzo-p-dioxins and Dibenzofurans in Two Cities of Southern China

Jinning Zhu, Haiyan Tang, Jin Xing, Wen-Jhy Lee, Ping Yan, Kangping Cui

Source: Volume: 17, Issue: 7, Pages: 1798-1810 DOI: 10.4209/aaqr.2017.05.0177

Atmospheric deposition is an important pathway for air pollutants entering the environment. In order to better understand both dry and wet deposition of PCDD/Fs in ambient air, two cities – Guangzhou and Nanjing in Southern China, were investigated. The monthly dry deposition fluxes of total-PCDD/Fs-WHO2005-TEQ were in the range of 60.6–560 and 104–1160 pg WHO2005-TEQ m⁻² month⁻¹ during 2014 in Guangzhou and Nanjing, respectively. In addition, it was found that the monthly dry deposition velocities of particle phase PCDD/Fs-TEQ ranged between 0.49 and 0.98 cm s⁻¹ (averaged 0.69 cm s⁻¹) and between 0.44 and 0.8 cm s⁻¹ (averaged 0.52 cm s⁻¹) in Guangzhou and Nanjing, respectively. The average scavenging ratios of total-PCDD/Fs-WHO2005-TEQ were 20480 and 30947 in Guangzhou and Nanjing, respectively. The total (dry + wet) deposition fluxes in Nanjing ranged between 135 and 1250, and averaged 643 pg WHO2005-TEQ m⁻² month⁻¹, approximately 1.38–2.23 times of magnitude higher than those in Guangzhou, which ranged between 97.7 to 559 pg WHO2005-TEQ m⁻² month⁻¹ and averaged 254 pg WHO2005-TEQ m⁻² month⁻¹. The results of this study provide useful information for both further studies and environmental control strategies aimed at persistent organic compounds (POPs).

Keywords: Dry deposition Wet deposition PCDD/Fs PM10 PM2.5 Southern China

Pollution Characteristics Revealed by Size Distribution Properties of Aerosol Particles at Urban and Suburban Sites, Northwest China

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Source: Volume: 17, Issue: 7, Pages: 1784-1797 DOI: 10.4209/aaqr.2016.07.0330

High temporal resolution (5 min) particle size distribution data (0.5–20 µm) were collected using aerodynamic particle sizer at an urban (Lanzhou) and a suburban (Yuzhong) site at Lanzhou, northwest China from 1st August to 31st October 2010. Variations of particle concentrations and properties of volume size distributions (PVSD) were analyzed and urban pollution characteristics were investigated using PVSDs and chemical analysis. The average particle number, surface area and volume concentrations for size range 0.5–10 µm were 280.54 ± 270.92 cm⁻³, 331.04 ± 316.95 µm² cm⁻³ and 93.01 ± 127.75 µm³ cm⁻³, respectively, at the urban site, which were 2.87, 1.50 and 1.62 times higher than those at the suburban site. Compared with the suburban site, shifts of accumulation mode (0.5–1.0 µm) to a smaller size and the coarse mode (1.0–10 µm) to a larger size of the PVSDs were observed at the urban site, which may be related to elevated fossil fuel burning and municipal construction or fugitive dust, respectively, in urban area. K-means cluster analysis was used to group the PVSD into six clusters representing the effect of different sources and meteorological conditions. PVSDs at the urban site were dominated by clusters affected by local anthropogenic sources and secondary aerosols, which was characterized by bimodal with peaks at accumulation mode and coarse mode, respectively, while those affected by construction works, wind-borne dust, and dust events were dominated by coarse mode. Chemical composition analysis of PM2.5 samples collected on days representing different clusters confirmed the assignment of clusters to different sources.

Keywords: Lanzhou Atmospheric particles Cluster analysis Volume concentration Sources

A Study on Distance Transport of PM_{2.5} to Xianlin in Nanjing, China and its Source Areas

Feng Cheng, Yong Zha , Jiahua Zhang , Junliang He, Shiyong Yan

Source: Volume: 17 , Issue: 7 , Pages: 1772-1783 DOI: 10.4209/aaqr.2016.08.0366

With rapid economic development in China, the concentration of particulate matters emitted into the atmosphere has become increasingly higher. As an important city in the Yangtze River delta, Nanjing City in China has a high PM_{2.5} concentration level, due partly to the long-range transport of PM_{2.5} from elsewhere. It is important to study the transport pathways and PM_{2.5} source areas as the results can serve as scientific evidence for prevention of regional air pollution. In this study, we analyzed the 2013–2016 PM_{2.5} concentration data at Xianlin in Nanjing City, China. After being clustered to determine the transport pathways, backward trajectory data underwent potential source contribution function (PSCF) and trajectory sector analysis so as to identify potential PM_{2.5} source areas. A new index called pollution source strength was proposed to indicate the level of pollution in an administrative region. Our results indicate that the concentration of PM_{2.5} at Xianlin in the city and externally sourced PM_{2.5} are high in winter, but much lower in summer. The external source areas also show some spatiotemporal regularities. External PM_{2.5} source areas lie chiefly to the south and southwest of Xianlin, Nanjing City in spring, but mostly to the southeast in summer, to the southeast and northwest in autumn, and to the northwest and northeast with a broader distribution in winter.

Keywords: PM_{2.5} Distance transport Backward trajectory PSCF Trajectory sector analysis

Ozone and Secondary Organic Aerosol Formation of Toluene/NO_x Irradiations under Complex Pollution Scenarios

Linghong Chen, Kaiji Bao, Kangwei Li, Biao Lv, Zhier Bao, Chao Lin, Xuecheng Wu, Chenghang Zheng , Xiang Gao, Kefa Cen

Source: Volume: 17 , Issue: 7 , Pages: 1760-1771 DOI: 10.4209/aaqr.2017.05.0179

Toluene is one of the most important precursor contributors to ozone and secondary organic aerosol (SOA), both of which greatly affect the air quality and human health. In this study, the effects of toluene on ozone and SOA formation were investigated in the presence of NO_x in the CAPS-ZJU (Complex Air Pollution Study-Zhejiang University) smog chamber. Three comparison groups of experiments were conducted under the gas-phase environments of toluene/NO_x, toluene/isoprene/NO_x, and toluene/ethylene/NO_x. The ozone concentration and physical properties of SOA such as mass concentration, aerosol yield, effective density, extinction and scattering were measured simultaneously. A toluene-dependent mechanism of ozone formation was found at ratios of toluene and NO_x between 3.1 and 11.3 with the initial NO_x concentration about 30 ppb. With further increase of the toluene concentration, the maximum value of ozone concentration remained almost stabilized. The maximum SOA yields decreased with increase of toluene, while the SOA effective density was concentrated at 1.3–1.4 g cm⁻³. The presence of isoprene or ethylene can promote the formation of ozone and SOA. The SOA nucleation was delayed

under different initial toluene concentrations and the yield was reduced at the same mass concentration. A linear increase of extinction and scattering was observed with the increase of SOA mass concentration in both the toluene/isoprene/NO_x and toluene/ethylene/NO_x systems. A rapid increase of single scattering albedo reflects the process of SOA nucleation and growth. In addition, organic aerosol oxidation products tend to carboxylic acids in toluene/isoprene/NO_x system according to Van Krevelen.

Keywords: Smog chamber Toluene Ozone Secondary organic aerosol Nox

Impact of Atmospheric Flow Conditions on Fine Aerosols in Sydney, Australia

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Source: Volume: 17 , Issue: 7 , Pages: 1746-1759 DOI: 10.4209/aaqr.2017.02.0083

We apply a simple objective measure of an airshed's degree of ventilation and determine the impact on PM_{2.5} observations at Lucas Heights, Sydney, Australia. We extend the analysis of previous studies, which considered total PM_{2.5}, by: using positive matrix factorisation to split the aerosol mass by source type; and using Radon-222 measurements as an independent indicator of ventilation and mixing. For this coastal airshed we found that for 64% of the time, conditions could be classified into four categories: local recirculation (LRC; 15%), stagnation (19.5%), regional recirculation (RRC; 10.9%), or ventilation (18.6%). Mean PM_{2.5} concentrations under recirculation (in this study separated into; LRC and RRC) were 33% higher than under stagnation and can be double that of concentrations under ventilation. Since the combination of LRC and RRC events account for around 26% of all events, recirculation effects on PM_{2.5} concentrations are significant. However, we found that airshed ventilation doesn't affect PM_{2.5} concentrations from all sources evenly. Considering the three main sources of total PM_{2.5} at this site (vehicle exhaust 26.3%, secondary sulfate 23.7% and aged industrial sulfur 20.6%), conditions leading to the highest concentrations differ. The highest vehicle exhaust concentrations occur under LRC, the highest aged-industrial-sulphur concentrations occur under RRC, and secondary sulfur had similarly high concentrations under LRC and RRC. Under LRC the concentration from vehicle exhaust can be up to a factor of 3.9 greater than under ventilation. On a seasonal basis, RRC flow is most likely to occur in summer and spring (the warmer months of the year when sea breezes are more likely), whereas LRC conditions are more likely to occur in autumn and winter. These findings support those of previous studies, indicating that re-circulation can have a significant effect on PM_{2.5} concentrations in coastal airsheds, and the degree of impact can vary by source type.

Keywords: Aerosols Circulation Stagnation Positive Matrix Factorisation Ventilation

Organic Nanoaerosol in Coal Mines: Formation Mechanism and Explosibility

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Source: Volume: 17, Issue: 7, Pages: 1735-1745 DOI: 10.4209/aaqr.2016.12.0533

The mechanism of aerosol formation in coal mines during the operation of a longwall shearer was studied using a diffusion battery, optical counter, and by means of Transmission Electron Microscopy (TEM). The aerosol number concentration was measured to be $(2-5) \times 10^5 \text{ cm}^{-3}$. The aerosol size spectrum contained three modes, at about 10, 100, and 1000 nm. The first mode relates to single (primary) particles formed by the homogeneous nucleation of supersaturated organic vapor. This vapor is formed by the evaporation of organic matter from coal due to the release of frictional heat at the interface between the cutting pick and coal. The second mode relates to the particles that are aggregates formed by coagulation of primary particles. The third mode relates to the particles formed by direct grinding of coal by the cutting picks.

The laboratory studies of organic aerosol formation in a flow coal grinding machine showed that the size spectrum and morphology of aerosol from the grinding machine were close to those in the coal mine. This fact confirms the thesis that the release of frictional heat is the driving force for the formation of organic aerosol. The analysis of gas-phase products in the outflow of the grinding machine showed that along with aerosol formation, gas products such as CO, CO₂, CH₄, C₂H₆, H₂O are released from coal due to the frictional heat. Methane and ethane concentrations in the flow reached 10 and 5 vol. %, respectively.

To demonstrate the explosibility of organic aerosol, the combustion of organic aerosol in the air was studied. It was shown that the lower explosive limit for organic aerosol is less than 50 g m⁻³. A conclusion is made that the formation of organic aerosol is to be taken into account when estimating the safety limits in coal mines.

Keywords: Aerosol generation Agglomeration Carbonaceous aerosols Nucleation Homogeneous Air pollution

Chemical Composition of Diesel/Biodiesel Particulate Exhaust by FTIR Spectroscopy and Mass Spectrometry: Impact of Fuel and Driving Cycle

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Source: Volume: 17, Issue: 7, Pages: 1717-1734 DOI: 10.4209/aaqr.2017.04.0127

The growing concern about air quality and the impact exhaust particles can have on the environment has resulted in the increased use of alternative fuels. A sampling campaign from a conventional heavy diesel engine operated in typical transient cycle or steady-state condition, and running on diesel, 30% biodiesel in diesel, and 100% biodiesel was carried out. The particulate composition was characterized using Fourier Transform Infrared (FTIR) spectroscopy, Two-step Laser Mass Spectrometry (L2MS), Secondary Ion Mass Spectrometry (SIMS), thermo-optical

analysis, and capillary electrophoresis. Elemental carbon is demonstrated to decrease from diesel to 100% biodiesel, in agreement with the evolution of aromatic bands and the MS abundance of Cn-fragments, while organic carbon exhibits a constant level irrespective of the working regime. Aliphatic, aromatic, carboxyl, carbonyl, hydroxyl functionalities, and nitro compounds are found to depend on the engine-working regime. Mass spectra are mainly characterized by alkyl fragments (C_nH_{2n+1}⁺), associated to normal and branched alkanes, PAHs and their alkylated derivatives. The addition of biodiesel to diesel changes the particulate composition towards more oxygenated constituents, such as carbonyl groups attributed to methyl ester CH₃O⁺ fragments of unburned biodiesel. Fuel-specific fragments have been identified, such as C₃H₇O⁺ for diesel, and C₂H₃O₂⁺ and CH₃O⁻ for biodiesel. Nitrogenized compounds are revealed by -NO₂ functionalities and N-containing fragments. Principal Component Analysis (PCA) was successfully applied to discriminate the engine operating conditions, with a higher variance given by the fuel, thus allowing to better evaluate the environmental impacts of alternative energy source emissions.

Keywords: Diesel engine Particulate emission FTIR Mass spectrometry Environmental pollution

The Construction and Application of a Multipoint Sampling System for Vehicle Exhaust Plumes

Xianbao Shen, Zhiliang Yao , Kebin He , Xinyue Cao, Huan Liu

Source: Volume: 17, Issue: 7, Pages: 1705-1716 DOI: 10.4209/aaqr.2017.02.0076

To study the formation process of secondary organic aerosols (SOA) in a vehicle exhaust plume near the exhaust gas discharge outlet, a new multipoint sampling system was established. The system has five sampling heads and includes a particulate matter multi-channel film sampling system, a CO₂/CO analyzer system, a volatile organic compound (VOC) sampling system, a particulate matter real-time analyzer system, and sensor interfaces. The vehicle exhaust near the exhaust nozzle can be sampled at multiple locations simultaneously using the new multipoint sampling system. Additionally, the system can be used to measure and analyze variations in the fine particulate matter, including the carbonaceous and ionic components, and organic compounds in the plume near the exhaust nozzle. This paper introduces the construction and application of the multipoint sampling system. The motor vehicle exhaust multipoint sampling system is reliable and can accurately capture the characteristics of the exhaust plume near the discharge outlet area. Changes in the CO₂ concentration were used to determine whether exhaust was accurately collected at the sampling points. The relationship between the dilution rate and distance was calculated on the basis of on-road test results using the following equation: DR = 21.4X^{1.16}. This equation can be used for modeling purposes, especially in comparisons of model results and observations and in the evaluation of dispersion models.

Keywords: Multipoint Sampling system Exhaust plume PM_{2.5} components

Ozone Catalytic Oxidation of Gaseous Toluene over MnO₂-Based Ozone Decomposition Catalysts Immobilized on a Nonwoven Fabric

Kazuhiko Sekiguchi , Yuki Kurita, Kenshi Sankoda, Norikazu Namiki, Fumio Yasui, Hajime Tamura

Source: Volume: 17, Issue: 8, Pages: 2110-2118 DOI: 10.4209/aaqr.2017.01.0045

Degradation of toluene gas by ozone catalytic oxidation (OZCO) by using a MnO₂-based ozone decomposition catalyst (ODC) was investigated to clarify the reactive site of ODC material with O₃. An optimum structure for the ODC to remove O₃ and toluene were proposed. For honeycomb ODC, toluene degradation by OZCO occurred only around the entrance of the honeycomb ODC, and we expected that a thinner ODC would increase the toluene degradation efficiency. A nonwoven fabric on which ODC was immobilized was developed to decompose O₃ and volatile organic compounds simultaneously. The toluene degradation ratio and the mineralization of toluene to CO₂ were determined to evaluate the performance of the fabric. Furthermore, the effects of relative humidity and O₃ concentration on the decomposition and mineralization ratios were also investigated with or without 254 nm UV irradiation (UV254). The fabric decomposed and mineralized toluene to CO₂, even at low O₃ concentrations. Although high humidity reduced the degradation ratio of toluene, UV254 irradiation improved the recovery of the degradation ratio and increased the mineralization ratio.

Keywords: Ozone decomposition catalyst (ODC) Ozone catalytic oxidation (OZCO) Toluene gas Nonwoven fabric UV irradiation

Particle Size Distribution of Soot from a Laminar/Diffusion Flame

Jian Wu, Linghong Chen , Jianwu Zhou, Xuecheng Wu, Xiang Gao, Gérard Gréhan, Kefa Cen

Source: Volume: 17, Issue: 8, Pages: 2095-2109 DOI: 10.4209/aaqr.2017.06.0216

A practical method is proposed to determine the inflame soot particle size distribution via an explicit solution derived from time-resolved laser-induced incandescence (TiRe-LII). Three appropriate time intervals are selected from the TiRe-LII decay signal. The equivalent mean sizes as well as relative ratios of number densities for three classes of monodisperse particles are determined with the mono-exponential fit to each interval, which allows an explicit solution for the particle size distribution. Simulations show that inversed log-normal distributions from the explicit solution are coincident with the input parameters in terms of trend, and there exist critical time intervals where inversed results are most insensitive to the variation of interval length. The error of inversion is critically dependent on the geometric standard deviation but weakly dependent on the count median particle diameter. Influences of experimental conditions on the inversed error are additionally evaluated. The results show that flame temperature has a significant impact on the error of inversion. Thus, a database of the error as a function of flame temperature at a fixed aggregate size is established. The error database allowed the inversed results from experimental TiRe-LII signals to be readily corrected at various flame locations by interpolation. The corrected inversed log-normal distributions were consistent in trend with those determined from the

established non-linear regression method, and the modeling LII signals reproduced agree with the experimental data. The small deviation of the results potentially stemmed from the statistical noise contained in recorded LII signals.

Keywords : Soot particle size Laser-induced incandescence Explicit solution Error database

Evaluation of $\delta^{13}\text{C}$ in Carbonaceous Aerosol Source Apportionment at a Rural Measurement Site

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Source: *Volume: 17 , Issue: 8 , Pages: 2081-2094 DOI: 10.4209/aaqr.2016.09.0392*

The stable isotope of carbon, ^{13}C , has been used in several studies for source characterization of carbonaceous aerosol since there are specific signatures for different sources. In rural areas, the influence of different sources is complex and the application of $\delta^{13}\text{C}$ for source characterization of the total carbonaceous aerosol (TC) can therefore be difficult, especially the separation between biomass burning and biogenic sources. We measured $\delta^{13}\text{C}$ from 25 filter samples collected during one year at a rural background site in southern Sweden. Throughout the year, the measured $\delta^{13}\text{C}$ showed low variability (-26.73 to -25.64‰). We found that the measured $\delta^{13}\text{C}$ did not correlate with other commonly used source apportionment tracers (^{14}C , levoglucosan). $\delta^{13}\text{C}$ values showed lower variability during the cold months compared to the summer, and this narrowing of the $\delta^{13}\text{C}$ values together with elevated levoglucosan concentrations may indicate contribution from sources with lower $\delta^{13}\text{C}$ variation, such as biomass or fossil fuel combustion. Comparison of two Monte Carlo based source apportionment models showed no significant difference in results when $\delta^{13}\text{C}$ was incorporated in the model. The insignificant change of redistributed fraction of carbon between the sources was mainly a consequence of relatively narrow range of $\delta^{13}\text{C}$ values and was complicated by an unaccounted kinetic isotopic effect and overlapping $\delta^{13}\text{C}$ end-member values for biomass burning and biogenic sources.

Keywords: End-member distributions Biomass burning Biogenic aerosol

Exposure Assessment of Hexavalent Chromium for a Powder Coating Spray Painter Associated with the Development of Lung Cancer

Kyeongmin Lee , Donguk Park, Boowook Kim, Jungah Shin

Source: *Volume: 17 , Issue: 8 , Pages: 2076-2080 DOI: 10.4209/aaqr.2016.05.0193*

A 63-year-old man who diagnosed with lung cancer had worked for 13.5 years (1999–2013) as a spray painter. The Occupational Lung Diseases Institute conducted retrospective exposure assessment to examine whether a spray painter job he had performed could be associated with the development of lung cancer. We investigated lung cancer carcinogens in his work environment. The safety data sheet of six powder coating products showed that powder coatings contained 1–10% of

hexavalent chromium. In addition, our quantitative analysis of powder coating samples also showed that the hexavalent chromium contents quantified in the yellow-green and red powder coating samples were 0.27% and 0.95%, respectively. In order to estimate his exposure level of hexavalent chromium, we measured a personal exposure level of hexavalent chromium for a spray painter in accordance with the National Institute for Occupational Safety and Health #7605 method. The results showed that the spray painter was exposed to the high level of hexavalent chromium ($216.9 \mu\text{g m}^{-3}$). Furthermore, we estimated that he was likely exposed to several lung cancer carcinogens such as crystalline silica or asbestos over the approximately 24 years at various construction sites prior to assuming a job as a spray painter. Therefore, we concluded that his lung cancer was caused by substantial exposure to several lung cancer carcinogens over approximately 37.5 years. Particularly, exposure characteristic to hexavalent chromium could substantially contribute to the development of lung cancer, despite of the sole case of exposure assessment.

Keywords: Lung cancer Exposure assessment Powder coatings Spray painter Hexavalent chromium

Fungal Bioaerosol Exposure and Its Effects on the Health of Mushroom and Vegetable Farm Workers in Taiwan

Yenni Gustiani Tarigan, Ruey-Yu Chen, Hsiu-Chen Lin, Chia-Yi Jung, Kraiwuth Kallawicha, Ta-Pang Chang, Po-Chen Hung, Chih-Yong Chen, Hsing Jasmine Chao

Source: Volume: 17, Issue: 8, Pages: 2064-2075 DOI: 10.4209/aaqr.2016.09.0401

Workers from specific occupational settings may be exposed to high fungal bioaerosol concentrations, causing detrimental health effects. Therefore, we conducted a study to evaluate the characteristics and health effects of fungal bioaerosols present on agricultural farms. By using IOM inhalable dust samplers, personal and area samples of airborne fungi were collected from five agricultural farms—two mushroom and three vegetable farms. A standardized questionnaire and spirometry were used to evaluate workers' health. The Kruskal–Wallis test was used to examine the distributions of fungal and environmental factors among the different farms, and regression analyses were performed to evaluate the effects of personal bioaerosol exposure on workers' health. In the personal samples, the geometric mean concentrations ranged from 4.3×10^3 to 3.0×10^4 CFU m^{-3} for total culturable fungi and from 4.2×10^3 to 1.2×10^5 spores m^{-3} for total fungal spores. The total fungal spore concentrations differed significantly among the personal samples ($p = 0.026$), but not among the area samples, from the five farms. The culturable fungal concentrations among the five farms did not differ significantly in the personal or area samples. Decreased lung functions of the workers were significantly associated with the concentrations of total fungi and several fungal taxa such as Ascospores, Fusarium, and Periconia. This study demonstrated that exposure to high fungal bioaerosol concentrations reduced the lung functions of the mushroom and vegetable farm workers. Superior ventilation and appropriate personal protection equipment are required to reduce occupational biohazards.

Keywords: Bioaerosols Personal exposure assessment Occupational biohazard Lung function

Ambient Fungal Spore Concentration in a Subtropical Metropolis: Temporal Distributions and Meteorological Determinants

Kraiwuth Kallawicha, Yi-Chen Chen, H. Jasmine Chao, Wei-Chiang Shen, Bing-Yu Chen, Yu-Chen Chuan, Yue Leon Guo

Source: Volume: 17, Issue: 8, Pages: 2051-2063 DOI: 10.4209/aaqr.2016.10.0450

Ambient particles comprise approximately 25% of fungal spores, which cause adverse health outcomes such as respiratory diseases, allergy, and infection. In this study, we investigated temporal variations and distributions of ambient fungal spores in an urban area of the Taipei metropolis for over 1 year. A Burkard 7-day volumetric spore trap was used to collect air samples. Samples collected daily were stained, counted, and identified on the basis of morphological characteristics. The associations between fungal spores and environmental parameters were then evaluated through multiple regression analysis. Daily monitoring data revealed a large variation in fungal spore concentrations. Specifically, fungal spores peaked during summer months (June–August) and declined during winter months (December–early March); moreover, the average concentration of total fungal spores was $3,607.97 \pm 3,181.81$ spores m^{-3} . Ascospores were the most prevalent taxon that was recovered from the samples, followed by basidiospores, *Aspergillus*/*Penicillium*, and *Cladosporium*. Multiple regression analysis revealed that meteorological parameters were the main predictors of fungal concentrations. Temperature, wind speed, and humidity were consistently correlated with total fungi and major fungal taxa, and sunlight had a negative association with ascospores. Among the atmospheric pollutants, particulate matter with an aerodynamic diameter $\leq 10 \mu m$ (PM₁₀) and ozone were positively associated with fungal spores. Carbon monoxide (CO) at lag day 1 had a negative association with basidiospores. This is the first study to characterize daily concentrations and determinants of ambient fungal spores in an urban area of Taipei metropolis. The obtained data can be used to evaluate the health impact of fungal spore exposure on the residents of the Taipei metropolitan area.

Keywords: Fungal spores Bioaerosols Temporal distribution Air pollution

Persistent Organic Pollutant Reductions from a Diesel Engine Generator Fueled with Waste Cooking Oil-based Biodiesel Blended with Butanol and Acetone

Jen-Hsiung Tsai, Sheng-Lun Lin, Shui-Jen Chen, Guo-Ping Chang-Chien, Bo-Cheng Jheng, Kuo-Lin Huang, Chih-Chung Lin, Juei-Yu Chiu

Source: Volume: 17, Issue: 8, Pages: 2041-2050 DOI: 10.4209/aaqr.2017.07.0233

This investigation focuses on the effects on emissions of persistent organic pollutants (POPs) (polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs), polychlorinated biphenyls (PCBs), polybrominated dibenzo-p-dioxins and dibenzofurans (PBDD/Fs) and polybrominated diphenyl ethers (PBDEs)) from a diesel engine fuelled by 20 vol% waste cooking oil-based biodiesel (W20) blended with various fractions of dehydrate/hydrous butanol (B/B') and acetone (A/A'). The emission concentrations of the POPs were in the order PBDE >> PBDD/F > PCB > PCDD/F,

regardless of the blending fuel or engine load. The POP with highest concentration was PBDE, being 2–3 times that of the others. Conversely, the magnitude of emitted toxicity followed the order PCDD/F > PCB \approx PBDD/F, while PCDD/F emissions had about 10 times the toxicity concentrations of PCBs and PBDD/Fs. Among the dioxin compounds, the emissions of PCDDs represented 46–73% (average 57%) and 50–72% (average 59%) of total PCDD/F mass and toxicity concentrations, respectively, and were which and were thus significantly higher than those of PCDFs. The non-ortho-PCB contributed almost all toxicity (\sim 100%) of 14 dioxin-like-PCBs, even though its contribution in mass was only 9–32% (average 16%) among the congeners. Similarly, PBDFs accounted for \sim 100% of toxicity of PBDD/Fs. Additionally, deca-BDEs contributed to most of the mass emissions of PBDEs (47.0–90.5%, 82.4% in average), while nona-BDEs and tri- to octa-BDEs only contributed 10% and 8%, respectively. The reductions of the absolute mass concentrations of POPs from W20 were in the order PBDEs \gg PBDD/Fs > PCDD/Fs \approx PCBs for all multi-component diesel blends. The reduction fractions of POPs were in the order PCDD/F > PCB \approx PBDD/F > PBDE, and those of TEQ were PCDD/F > PCB > PBDD/F. Thus, the addition of butanol and acetone, whether pure or hydrous, could further lower the POP emissions from W20.

Keywords: Generator engine Waste cooking oil-based biodiesel Butanol Acetone Persistent organic pollutants

Part II: PM_{2.5} and Polychlorinated Dibenzo-p-dioxins and Dibenzofurans (PCDD/Fs) in the Ambient Air of Northern China

Jin Xing, Kangping Cui, Haiyan Tang, Wen-Jhy Lee, Lin-Chi Wang, Jinning Zhu, Qianli Huang

Source: Volume: 17, Issue: 8, Pages: 2010-2026 DOI: 10.4209/aaqr.2017.06.0211

During 2014–2016, this study investigated the atmospheric PM_{2.5}, RM, PM_{2.5}/PM₁₀, PCDD/Fs-WHO2005-TEQ, and PM_{2.5}-bound total-PCDD/Fs-WHO2005-TEQ content of 22 cities in northern China. In general, the more highly industrialized cities had higher PM_{2.5} concentrations. The lowest three-year average concentrations of PM_{2.5} occurred at Lhasa and Qiqihar, and were 25.2 and 36.7 $\mu\text{g m}^{-3}$, respectively, while the highest concentrations of PM_{2.5} occurred at Baoding and Shijiazhuang, and were 106 and 102 $\mu\text{g m}^{-3}$, respectively. From 2015 to 2016, the PM_{2.5} concentrations of most cities decreased, but those of several others (Shijiazhuang, Taiyuan, Yinchuan, Lhasa, Sinning, Urumqi, Weinan and Xian) increased, suggesting that the air quality of these was still not well controlled. The average of RM values of the 22 cities were 7.2, 6.5, and 6.1 in 2014, 2015 and 2016, respectively, which means the PM_{2.5} concentrations in northern China were much higher than the WHO air quality regulated standard (10 $\mu\text{g m}^{-3}$). A city with a higher PM_{2.5} concentration always had a higher PM_{2.5}/PM₁₀ ratio. Among the 22 cities, the six highest three-year averages of total-PCDD/Fs-WHO2005-TEQ concentrations were 0.107, 0.102, 0.095, 0.092, 0.085 and 0.077 pg-WHO2005-TEQ m^{-3} in Shijiazhuang, Baoding, Zhengzhou, Jinan, Linyi and Xian, respectively; the six lowest three-year averages of total-PCDD/Fs-WHO2005-TEQ concentrations were 0.036, 0.037, 0.045, 0.055, 0.056 and 0.060 pg-WHO2005-TEQ m^{-3} in Qiqihar, Lhasa, Dalian, Harbin, Changchun and Hohhot, respectively. The PM_{2.5}-bound total PCDD/Fs-WHO2005-TEQ content of 12 cities (six cities with higher PM_{2.5} concentration and six with lower PM_{2.5} concentration), during 2014, ranged between 0.444 and 1.000 ng-WHO2005-TEQ g^{-1} and averaged 0.672 ng-WHO2005-TEQ g^{-1} . The PM_{2.5} concentrations, RM values and PCDD/Fs-WHO2005-TEQ concentrations in the cities of northern China are higher than those in the south,

indicating that the air quality in the north is worse than in the south. The results of this study provide a theoretical basis for proposing air pollution control strategies and improving the atmospheric environment in China.

Keywords: PM2.5 PCDD/Fs PM2.5/PM10ratio TEQ Northern China Cities

Mobile Monitoring of Personal NO_x Exposures during Scripted Daily Activities in Chicago, IL

Jiayao Xu, Han Jiang, Haoran Zhao, Brent Stephens

Source: Volume: 17, Issue: 8, Pages: 1999-2009 DOI: 10.4209/aaqr.2017.02.0063

Elevated ambient concentrations of nitrogen oxides (NO_x), including nitric oxide (NO) and nitrogen dioxide (NO₂), are associated with a wide range of adverse human health effects. Most studies have investigated these associations using ambient NO₂/NO_x measurements from fixed-site monitors or modeled ambient NO₂/NO_x concentrations. However, the majority of personal exposures to NO₂/NO_x occur in a variety of different microenvironments in which people spend most of their time. Previous studies have reported widely varying correlations between personal exposures and ambient NO₂/NO_x concentrations over various timescales. To add to the knowledge base of how personal NO/NO₂/NO_x exposures vary spatially, temporally, and within different microenvironments in an urban environment, we conducted roll-around mobile monitoring of NO/NO₂/NO_x with 1-minute resolution during 14 days of scripted activities in and around Chicago, IL. Activities involved time spent in three primary microenvironments: outdoors, indoors inside various building types, and in multiple modes of transportation including walking, personal vehicle, and public transit. Measurement were conducted at a higher time resolution than most prior microenvironmental monitoring studies using a recently developed direct UV absorbance NO/NO₂/NO_x monitor that is designed to minimize interferences that have been observed in some field campaigns using chemiluminescence monitors. The individual microenvironmental categories with the highest median NO_x concentrations included four indoor environments and a variety of public transit environments. The individual transportation microenvironments with the highest median NO_x concentrations were found aboard regional trains, largely driven by high NO from diesel locomotives. Correlations between microenvironmental NO/NO₂/NO_x measurements and simultaneous records from the nearest ambient monitor were extremely low, with coefficients of determination below 0.05 for each NO_x constituent. These data further illustrate the limitations of relying on ambient site regulatory monitors to characterize personal NO/NO₂/NO_x exposures and provide further evidence that personal monitoring is critical for accurately assessing personal exposure to NO_x.

Keywords: Nitrogen oxides Human exposure Personal exposure Mobile samplers Indoor air pollution

Pollution Characteristics and Source Apportionment of PM_{2.5}-Bound n-Alkanes in the Yangtze River Delta, China

Zhenyu Hong, Youwei Hong, Han Zhang, Jinsheng Chen, Lingling Xu, Junjun Deng, Wenjiao Du, Yanru Zhang, Hang Xiao

Source: Volume: 17, Issue: 8, Pages: 1985-1998 DOI: 10.4209/aaqr.2016.12.0566

PM_{2.5}-bound n-alkanes in Shanghai (SH), Nanjing (NJ) and Ningbo (NB) cities from November 2014 to August 2015 were investigated. Averaged concentrations of the total 25 n-alkanes (Σ n-alkanes, C₁₆–C₄₀) in SH, NJ and NB were 97.4 ± 73.9 , 83.8 ± 57.1 and 187.1 ± 87.1 ng m⁻³, respectively. Obvious spatial and seasonal variations were attributed to the differences of emission sources and meteorological conditions. Analysis of the diagnostic ratios and specific molecular markers of n-alkanes suggested that high plant wax and vehicle emissions were the major sources of n-alkanes in the YRD region. Strong inputs of microbial components in summer were found and attributed to the emission from plankton in the ocean. The annual average contributions of higher plant wax to n-alkanes (%wax) in SH, NJ and NB were estimated to be 47.5%, 50.1% and 34.5%, respectively. Anthropogenic sources were responsible for the n-alkanes in NB, while biogenic sources contributed much more n-alkanes in NJ and SH. Based on the result of backward analysis, the emissions of n-alkanes in NB and NJ were mainly from local sources when the air masses came from the sea and south China with low n-alkanes concentrations. When the air masses originated from north China, the transport of contaminant aggravated the pollution of n-alkanes in SH.

Keywords: n-alkanes Fine particulate matter (PM_{2.5}) Pollution characteristics Source apportionment Yangtze River Delta

Particulate Size Distribution and Sources Evaluation of n-Alkanes during Long-Term Haze Episode around Chaohu Lake, Eastern

Qiu-Ping Xu, Ji-Zhong Wang, Jia-Qin Liu, Shu-Chuan Peng

Source: Volume: 17, Issue: 8, Pages: 1975-1984 DOI: 10.4209/aaqr.2017.05.0189

n-Alkanes (from nC₁₆ to nC₃₂) associated with particulate matters were determined in the ambient air around Chaohu Lake, Eastern China, from October to December in 2014 during a long-term haze episode. The total concentrations of particle bounded n-alkanes varied from 332 to 2500 ng m⁻³, with the homologues of nC₂₄–nC₃₀ the most abundant species. Spatial analysis revealed that low concentrations of n-alkanes existed at the sites close to Chaohu Lake, while high concentrations were generally at locations distant from the lake. For all aggregated fractions, most n-alkanes were distributed in fine particles with the mean geometric mean diameter (GMD) varying from 3.0 ± 0.6 μ m for nC₁₆ to 2.1 ± 0.6 μ m for nC₃₂. Short chain n-alkanes were accumulated in coarse particles with a unimodal distribution, but long chain aliphatic hydrocarbons appeared to have a bimodal distribution in fine and coarse particles. The mass size distribution of individual n-alkane homologue was predominantly influenced by its volatility; thus GMDs were well correlated with the logarithmically transformed subcooled liquid vapor pressures (P_{Lo}, Pa) of n-alkanes at each sampling site, following the equation: $GMDs = mgLogP_{Lo} + bg$. Furthermore, mg and bg

obtained from all locations tended to exhibit a significant linear correlation. This suggests that all saturated aliphatic hydrocarbons follow a similar accumulation mode during a haze episode, which allows us to predict the size distribution and GMD of a compound based on its PLo.

Keywords: Chaohu Lake n-alkanes particulate matter size distribution geometric mean diameter

Coarse-Particle Passive-Sampler Measurements and Single-Particle Analysis by Transmitted Light Microscopy at Highly Frequented Motorways

Zhaoxue Tian, Volker Dietze, Frank Sommer, Anja Baum, Uwe Kaminski, Jan Sauer, Christoph Maschowski, Peter Stille, Kuang Cen, Reto Gieré

Source: *Volume: 17, Issue: 8, Pages: 1939-1953 DOI: 10.4209/aaqr.2017.02.0064*

Measuring and characterizing airborne particulate matter (PM) is an important research area because PM can lead to impacts on health and to visibility reduction, material damage and groundwater pollution. In regard to road dust, suspension and re-suspension and the contribution of non-exhaust PM to total traffic emissions are expected to increase as a result of predicted climate scenarios. European environmental regulations have been enforced to reduce exhaust particle emissions from road traffic, but little attention has been paid to reducing non-exhaust coarse particle emissions due to traffic. Therefore, a monitoring program for coarse PM has been initiated in early 2013 to assess the predicted increase in the abundance of non-exhaust particles. Particle sampling was performed with the passive-sampler technique Sigma-2. The subsequent single-particle analysis allows for characterization of individual particles, determination of PM size distribution, and calculation of PM mass concentrations. Two motorways near Cologne (Köln), Germany were selected as sampling sites, and the experimental setup in the field was realized with a so-called twin-site method. The present study reports single-particle analysis data for samples collected between May 31, 2013 and May 30, 2014. Coarse PM, generated through multi-source mechanisms, consists of, e.g., tire-wear, soot aggregates, and mineral dust. The highest mass concentration occurs at both motorways in spring, and the observed PM mainly contains traffic-abrasion particles. The field measurements show that the minimum PM concentration was found in the 5 to 12°C temperature range, whereas the maximum concentration was observed in both the -5 to 5°C and the 12 to 24°C ranges, in agreement with previous laboratory measurements. Correlation between super-coarse (dp 10–80 µm, geometric equivalent diameter) PM concentration and precipitation displays a significant increase in concentration with decreasing number of precipitation events (dry weather periods).

Keywords: Passive-sampling Individual particle characterization Particle size distribution Seasonal meteorology Traffic impact

Atmospheric Research

Comparison of atmospheric PM_{2.5}-bounded mercury species and their correlation with bromine and iodine at coastal urban and island sites in the eastern China

NaChengLianDuanaGuangliXiuaMengfeiZhaoaGuanleiQian

Source: Volume 183, 1 January 2017, Pages 17-25

<https://doi.org/10.1016/j.atmosres.2016.08.009>

A year-long observation of PM_{2.5}-bounded mercury (PBM) and its species was conducted at a urban site (Shanghai, Xuhui; XH) and an island site (Shengsi, SS) in eastern China from September 2014 to August 2015. The seasonal variation of mercury species including hydrochloric soluble particle-phase mercury (HPM), element soluble particle-phase mercury (EPM) and residual soluble particle-phase mercury (RPM), as well as particulate halogen (Br, I) were determined. HPM concentration is the highest at urban, but RPM showed the largest fraction at island. Higher mass contents of all PM_{2.5}-bounded mercury species were found at island site than those at urban site, which indicated atmospheric mercury is more easily scavenged by particles at ocean atmosphere. Additionally, the correlation between bromine and mercury was stronger at urban site than that at island site, while iodine had the stronger correlation with mercury at island site than that urban site. These results showed marine aerosols played an important role to the transport of mercury.

Keywords: Mercury, PM_{2.5}, Urban site, Island site, Bromine, Iodine

Carbonaceous particles and aerosol mass closure in PM_{2.5} collected in a port city

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Source: Volume 183, 1 January 2017, Pages 245-254

<https://doi.org/10.1016/j.atmosres.2016.08.022>

Mass concentrations of PM_{2.5}, mineral dust, organic carbon (OC) and elemental carbon (EC), water-soluble organic carbon (WSOC), sea salts and anthropogenic metals have been studied in a city-port of south Italy (Brindisi). This city is characterized by different emission sources (ship, vehicular traffic, biomass burning and industrial emissions) and it is an important port and industrial site of the Adriatic sea. Based on diagnostic ratios of carbonaceous species we assess the presence of biomass burning emissions (BBE), fossil fuel emissions (FFE) and ship emission (SE). Our proposed conversion factors from OC to OM are higher than those reported in the literature for urban site: the reason of this could be due to the existence of aged combustion aerosols during the sampling campaign (WSOC/OC = 0.6 ± 0.3).

Keywords: PM_{2.5} Diagnostic ratios, Aerosol organic mass, Mass reconstruction

Seasonal variation of near surface black carbon and satellite derived vertical distribution of aerosols over a semi-arid station in India

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Source: Volume 184, 1 February 2017, Pages 77-87
<https://doi.org/10.1016/j.atmosres.2016.09.003>

Extensive measurements of aerosol black carbon mass concentration (BC) and vertical profiles of atmospheric aerosols have been carried out using Aethalometer and CALIPSO level – 2 satellite data from December 2012 to November 2014 over a semi-arid station, Anantapur. We found a bimodal distribution in the mass concentrations of BC aerosols on a diurnal scale. A sharp peak was observed during morning rush hours (7:00 to 8:00 LT) almost an hour after the local sunrise. After which, a broad nocturnal peak was found during ~ 21:00 to 22:00 LT. The seasonal mean BC concentrations (Mixed layer height (ML)) were found to be $3.45 \pm 1.44 \mu\text{g}/\text{m}^3$ ($676 \pm 117 \text{ m}$), $2.55 \pm 0.85 \mu\text{g}/\text{m}^3$ ($1215 \pm 190 \text{ m}$), $1.22 \pm 0.31 \mu\text{g}/\text{m}^3$ ($1134 \pm 194 \text{ m}$) and $1.75 \pm 0.70 \mu\text{g}/\text{m}^3$ ($612 \pm 135 \text{ m}$), during the winter, summer, monsoon and post-monsoon respectively. The vertical profiles of aerosol extinction coefficient and back scattering ratio profiles were derived from Cloud Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) showed a strong seasonal variation with aerosols mostly confined below 2 km during the post-monsoon and winter seasons whereas in the other two seasons, the aerosol layer expands beyond 6 km. Depolarization ratios (> 0.2) are higher during summer and monsoon at higher altitude regions demonstrate the presence of dust particles, which contribute to the large aerosol extinction at higher levels. These results are further supported by the backward trajectory cluster analysis.

Keywords: Black carbon, Mixed layer height, Extinction coefficient, Depolarization ratio, Hysplit analysis

Water soluble inorganic species of PM₁₀ and PM_{2.5} at an urban site of Delhi, India: Seasonal variability and sources

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Source: Volume 184, 1 February 2017, Pages 112-125
<https://doi.org/10.1016/j.atmosres.2016.10.005>

Comprehensive data of 2 years (2013–2014) of water soluble inorganic species (WSIS) in the particulate matter (PM₁₀: mean: $233.0 \pm 124.6 \mu\text{g m}^{-3}$ and PM_{2.5}: mean: $108.0 \pm 86.5 \mu\text{g m}^{-3}$)

have been used to study seasonal effect on the variation of total WSIS concentration, composition variability of inorganic aerosols and extent to which secondary formation of sulfate and nitrate aerosol occurred from their precursor gases. Mean concentrations of total WSIS in PM10 and PM2.5 were $82.12 \pm 72.15 \mu\text{g m}^{-3}$ and $54.03 \pm 49.22 \mu\text{g m}^{-3}$, respectively during the study period. Concentrations of total WSIS (PM10: $140.11 \pm 90.67 \mu\text{g m}^{-3}$; PM2.5: $74.41 \pm 47.55 \mu\text{g m}^{-3}$) during winter season was recorded higher than summer, monsoon and spring seasons. Significant correlation ($p < 0.01$) between NH_4^+ and Cl^- , SO_4^{2-} , NO_3^- in PM10 and PM2.5, respectively indicates NH_4^+ as the major cation species for the neutralization of acidic components in the winter season. On the contrary, in summer season Ca^{2+} , Mg^{2+} , Na^+ and K^+ were the alkaline species responsible for the neutralization of acidic components in the PM10 samples. Principal Component Analysis (PCA) showed that secondary aerosol, biomass burning and soil driven dust were the possible sources that explained $\sim 70\%$ of the total variance. Cluster analysis and Concentration Weighted Trajectory (CWT) analysis for different season depicts the advection of air masses over the continental landmasses of Afghanistan (summer season), northwestern region of Pakistan (summer and winter season), marine region (monsoon season) and adjoining states of Delhi. These air masses from different regions could be the cause of an increase in PM10 and PM2.5 aerosol over the study site.

Keywords: Particulate matter, Water soluble inorganic species, PCA, Cluster analysis, CWT analysis

Smog chamber study on aging of combustion soot in isoprene/SO₂/NO_x system: Changes of mass, size, effective density, morphology and mixing state

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Source: Volume 184, 1 February 2017, Pages 139-148

<https://doi.org/10.1016/j.atmosres.2016.10.011>

Atmospheric soot aging process is always accompanied by secondary particle formation, which is a comprehensive environmental issue that deserves great attention. On one hand, aging of primary soot could change its own physicochemical properties; on the other hand, complex air pollution caused by pollutant emission from various sources (e.g., vehicle exhausts, coal-fired flue gases and biogenic VOCs emission) may contribute to secondary particle formation onto primary particle surface. In this study, aging of combustion soot in isoprene/SO₂/NO_x system was investigated under controlled laboratory conditions in several smog chamber experiments. During the evolution of soot, several physical properties such as mass, size, effective density, morphology and mixing state were determined simultaneously by an integrated aerosol analytical system of Scanning Mobility Particle Sizer (SMPS), Differential Mobility Analyzer-Aerosol Particle Mass Analyzer-Condensation Particle Counter (DMA-APM-CPC) and Transmission Electron Microscopy coupled with Energy-dispersive X-ray Spectrometry (TEM/EDX) techniques. Here, based on the experimental results of soot aging under different gas-phase composition and relative humidity (RH), we firstly proposed possible aging pathways of soot in isoprene/SO₂/NO_x system. A synergetic effect was speculated to exist between SO₂ and isoprene on soot aging process, which led to more secondary particle formation. At the same time, TEM/EDX analysis showed that a competitive mechanism between H₂SO₄(g) and isoprene oxidation vapor may exist: H₂SO₄(g) firstly condensed onto fresh soot, then an acceleration of isoprene oxidation products formed onto

H₂SO₄ pre-coated soot. In isoprene/SO₂/NO_x system, high RH conditions could contribute to soot aging and new particle formation. The changes of effective density and dynamic shape factor of soot also indicated that high RH conditions could accelerate soot aging process, and led chain-like soot into more spherical morphology, which was further confirmed from the STEM image. Moreover, it was found that volume equivalent coating thickness (Δr_{ve}) could also be applied to normalized characterize soot aging parameters like diameter growth factor (G_{fd}) and mass growth factor (G_{fm}) in a complex reaction system like isoprene/SO₂/NO_x. Our results revealed the dual mechanism (competitive effect&cooperative effect) of isoprene and SO₂ on photochemical aging of soot, which is of significance for improving understanding of complex air pollution in China.

Keywords: Smog chamber, Complex air pollution, Soot aging, SOA, Heterogeneous reaction

Aerosol black carbon quantification in the central Indo-Gangetic Plain: Seasonal heterogeneity and source apportionment

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Source: Volume 185, 1 March 2017, Pages 13-21
<https://doi.org/10.1016/j.atmosres.2016.10.001>

Two years of aerosol spectral light absorption measurements, using filter based technique, from the central Indo-Gangetic plain (IGP), Gorakhpur (26.75°N, 83.38°E, 85 m amsl), are analyzed to study their seasonal behavior and to quantify their magnitude in terms of absorbing aerosols loading and source speciation. Spectral absorption analysis reveals a four-fold enhancement in absorption in winter (W) and post-monsoon (PoM) seasons at UV wavelengths as compared to IR wavelengths on account of increased biomass burning aerosol contribution to total absorbing aerosol load. Aerosols from the biomass sources contribute ~ 28% during W and PoM seasons as against ~ 16% in pre-monsoon (PM) and monsoon (M) seasons to the total absorbing aerosol content. A Mode shift in the distribution of the Absorption Ångström exponent (α) from 1.3 to 1.6 from PM-M seasons to PoM-W seasons signifies change in source type of absorbing aerosols from fossil fuel to biomass burning and their relative source strength. Due to near stagnant wind conditions combined with shallow boundary layer height, where air masses travelling to the central IGP are confined to a smaller volume, in W and PoM seasons, local sources assume more prominence rather than long-range transport of aerosols. Long-term measurements of aerosols physicochemical and radiative properties from this measurement location will enhance our understanding of the complex aerosol system over the IGP and its climatic implications.

Keywords: Indo-Gangetic Plain, Black carbon, Biomass burning, Fossil fuel, Absorption Ångström exponent

Urban air quality in a mid-size city — PM2.5 composition, sources and identification of impact areas: From local to long range contributions

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Source: Volume 186, 1 April 2017, Pages 51-62
<https://doi.org/10.1016/j.atmosres.2016.11.011>

Urban air quality represents a major public health burden and is a long-standing concern to European citizens. Combustion processes and traffic-related emissions represent the main primary particulate matter (PM) sources in urban areas. Other sources can also affect air quality (e.g., secondary aerosol, industrial) depending on the characteristics of the study area. Thus, the identification and the apportionment of all sources is of crucial importance to make effective corrective decisions within environmental policies.

The aim of this study is to evaluate the impacts of different emissions sources on PM2.5 concentrations and compositions in a mid-size city in the Po Valley (Treviso, Italy). Data have been analyzed to highlight compositional differences (elements and major inorganic ions), to determine PM2.5 sources and their contributions, and to evaluate the influence of air mass movements. Non-parametric tests, positive matrix factorization (PMF), conditional bivariate probability function (CBPF), and concentration weighted trajectory (CWT) have been used in a multi-chemometrics approach to understand the areal-scale (proximate, local, long-range) where different sources act on PM2.5 levels and composition.

Results identified three levels of scale from which the pollution arose: (i) a proximate local scale (close to the sampling site) for traffic non-exhaust and resuspended dust sources; (ii) a local urban scale (including both sampling site and areas close to them) for combustion and industrial; and (iii) a regional scale characterized by ammonium nitrate and ammonium sulfate. This approach and results can help to develop and adopt better air quality policy action.

Keywords: Urban area, Sources, PMF, Atmospheric circulation

Source contributions to PM2.5 in Guangdong province, China by numerical modeling: Results and implications

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Source: Volume 186, 1 April 2017, Pages 63-71
<https://doi.org/10.1016/j.atmosres.2016.11.007>

As one of the most populous and developed provinces in China, Guangdong province (GD) has been experiencing regional haze problems. Identification of source contributions to ambient PM2.5 level is essential for developing effective control strategies. In this study, using the most up-to-date emission inventory and validated numerical model, source contributions to ambient PM2.5 from eight emission source sectors (agriculture, biogenic, dust, industry, power plant, residential, mobile and others) in GD in 2012 were quantified. Results showed that mobile sources are the dominant

contributors to the ambient PM_{2.5}(24.0%) in the Pearl River Delta (PRD) region, the central and most developed area of GD, while industry sources are the major contributors (21.5% ~ 23.6%) to those in the Northeastern GD (NE-GD) region and the Southwestern GD (SW-GD) region. Although many industries have been encouraged to move from the central GD to peripheral areas such as NE-GD and SW-GD, their emissions still have an important impact on the PM_{2.5} level in the PRD. In addition, agriculture sources are responsible for 17.5% to ambient PM_{2.5} in GD, indicating the importance of regulations on agricultural activities, which has been largely ignored in the current air quality management. Super-regional contributions were also quantified and their contributions to the ambient PM_{2.5} in GD are significant with notable seasonal differences. But they might be overestimated and further studies are needed to better quantify the transport impacts.

Keywords: Source apportionment, Guangdong province, Super-regional impact, Seasonal variations

Model analysis of soil dust impacts on the boundary layer meteorology and air quality over East Asia in April 2015

Source: Volume 187, 1 May 2017, Pages 42-56
<https://doi.org/10.1016/j.atmosres.2016.12.008>

An online coupled meteorology-chemistry-aerosol model (WRF-Chem) is used to quantify the impact of soil dust on radiative forcing, boundary layer meteorology and air quality over East Asia. The simulation is conducted from 14 to 17 April 2015, when an intense dust storm originated in the Gobi Desert and moved through North China. An integrated comparison analysis using surface observations, satellite, and lidar measurements demonstrates the excellent performance of the WRF-Chem model for meteorological parameters, pollutant concentrations, aerosol optical characteristics, and the spatiotemporal evolution of the dust storm. The maximum aerosol optical depth induced by dust aerosols is simulated to exceed 3.0 over the dust source areas and 1.5 over the downwind regions. Dust has a cooling effect (-1.19 W m^{-2}) at the surface, a warming effect ($+0.90 \text{ W m}^{-2}$) in the atmosphere and a relatively small forcing (-0.29 W m^{-2}) at the top of the atmosphere averaged over East Asia from 14 to 17 April 2015. Due to the impact of dust aerosols, the near-surface air temperature is decreased by 0.01°C and 0.06°C in the daytime and increased by 0.13°C and 0.14°C at night averaged over the dust sources and the North China Plain (NCP), respectively. The changes in relative humidity are in the range of -0.38% to $+0.04\%$ for dust sources and -0.40% to $+0.27\%$ for NCP. The maximum decrease in wind speed of $\sim 0.1 \text{ m s}^{-1}$ is found over NCP. The planetary boundary layer height during the daytime exhibits maximum decreases of 16.34 m and 41.70 m over dust sources and NCP, respectively. The pollutant concentrations are significantly influenced by dust-related heterogeneous chemical reactions, with a maximum decrease of 1.66 ppbV for SO₂, 7.15 ppbV for NO_y, 35.04 $\mu\text{g m}^{-3}$ for NO₃⁻, and a maximum increase of 9.47 $\mu\text{g m}^{-3}$ for SO₄²⁻ over the downwind areas.

Keywords: Dust, Boundary layer meteorology, Air quality, Heterogeneous reaction, WRF-Chem

Characteristics of lightning activity in tropical cyclones developed over North Indian Ocean basin during 2010–2015

Source: Volume 187, 1 May 2017, Pages 16-32
<https://doi.org/10.1016/j.atmosres.2016.12.003>

The characteristics of lightning activity in tropical cyclones (TCs) over North Indian Ocean (NIO) are presented using sample of 21 TCs developed during 2010–2015 using TRMM and World Wide Lightning Location Network (WWLLN) datasets along with information from annual reports of Regional Specialized Meteorological Center (RSMC), New Delhi. The microphysical features such as Polarization Corrected Brightness Temperature (PCT), attenuation corrected reflectivity factor, Ice Water Path (IWP) play a pivotal role in development of convective systems within TCs. The TCs exhibited systematic variation in lightning flashes per day within 300 km of estimated center during their life cycle irrespective of their severity and flash rate within 300 km of storm center. The lightning flashes ranged from 1 to 3500 flashes per day during pre-cyclone stage, 100 to 8000 flashes per day during cyclone stage and 0 to 4300 flashes per day during post-cyclone stage. The TCs produced maximum flash density in eyewall region (20–40 km) and outer rainband region (200–260 km). The WWLLN recorded 165,512 flashes within 300 km of TC centers during their life cycle. The maximum flashes occurred during cyclone stage followed by pre-cyclone and post-cyclone. The time variation of flash rate for all cyclones was episodic and primarily peaked during late night and early morning hours. The diurnal variation of lightning flashes during TCs due to variation in detection efficiency of WWLLN also controls temporal distribution of lightning activity. During rapid intensification, TCs produced profuse lightning flashes per hour. Episodic lightning flashes per hour occurred during weakening stage and prior to landfall. It is evident that outbreaks of lightning flashes prior to maximum intensity change as manifested in maximum sustained wind speed and fall in estimated central pressure (ECP) indicate potential predictive value of lightning activity for TC intensity change.

Keywords: Tropical , cyclone, Lightning, Microphysics

A study on aerosol-cloud condensation nuclei (CCN) activation over eastern Himalaya in India

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Source: Volume 189, 1 June 2017, Pages 69-81
<https://doi.org/10.1016/j.atmosres.2017.01.015>

Simultaneous measurements of condensation a nucleus (CN) and cloud condensation nuclei (CCN) has been performed over a high altitude site Darjeeling (27°01'N, 88°15'E, 2200 m asl) at eastern part of Himalaya in India. The study was carried out during dry seasons (October 2015–May 2016) to investigate the temporal variability of CN and CCN concentrations and the major factors controlling CN-CCN activation. CCN concentrations measured at 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8 and 0.9% super saturations have been reported in this study. The number concentrations of CN ranged between 694 and 23,643 cm⁻³ with an average of 6563 ± 2160 cm⁻³ whereas that of CCN (at 0.5% super saturation) ranged between 262 and 13,382 cm⁻³ with an average of

1761 ± 856 cm⁻³ during the entire study period. CN and CCN showed prominent monthly and seasonal variations controlled by local emissions, meteorological factors and long-range transport. Indo-Gangetic Plain (IGP) was found to be the most contributing region for CN and CCN over Darjeeling mainly during winter and premonsoon. A clear indication of the contribution of aerosols from plain land regions driven by up-slope valley wind was observed during premonsoon from diurnal variability of CN and CCN. Overall, 30–32% aerosols were observed to activate to CCN during winter and premonsoon whereas 24% activation was observed during postmonsoon. Chemical nature and hence solubility of aerosols controlled CCN activation more in night-time than day-time as observed from the correlations between activation ratios and hygroscopicity parameter, *k*. Strong seasonal dependence of CCN and activation ratio on super saturation and aerosol loading was observed. We observed higher CN and CCN loading and lower activation ratio over Darjeeling compared to western Himalayan high altitude station suggesting higher influence of local and long-range aerosols over eastern Himalaya and significant difference in chemical nature of aerosols between these two parts of Himalaya.

Keywords: Cloud condensation nuclei, Eastern Himalaya, Activation ratio, Darjeeling

Source apportionment of ambient PM₁₀ and PM_{2.5} in Haikou, China

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Source: Volume 190, 1 July 2017, Pages 1-9
<https://doi.org/10.1016/j.atmosres.2017.01.021>

In order to identify the sources of PM₁₀ and PM_{2.5} in Haikou, 60 ambient air samples were collected in winter and spring, respectively. Fifteen elements (Na, Mg, Al, Si, K, Ca, Ti, V, Cr, Mn, Fe, Ni, Cu, Zn and Pb), water-soluble ions (SO₄²⁻ and NO₃⁻), and organic carbon (OC) and elemental carbon (EC) were analyzed. It was clear that the concentration of particulate matter was higher in winter than in spring. The value of PM_{2.5}/PM₁₀ was > 0.6. Moreover, the proportions of TC, ions, Na, Al, Si and Ca were more high in PM₁₀ and PM_{2.5}. The SOC concentration was estimated by the minimum OC/EC ratio method, and deducted from particulate matter compositions when running CMB model. According to the results of CMB model, the resuspended dust (17.5–35.0%), vehicle exhaust (14.9–23.6%) and secondary particulates (20.4–28.8%) were the major source categories of ambient particulate matter. Additionally, sea salt also had partial contribution (3–8%). And back trajectory analysis results showed that particulate matter was greatly affected by regional sources in winter, while less affected in spring. So particulate matter was not only affected by local sources, but also affected by sea salt and regional sources in coastal cities. Further research could focuses on establishing the actual secondary particles profiles and identifying the local and regional sources of PM at once by one model or analysis method.

Keywords: PM₁₀ and PM_{2.5}, Chemical composition, Source apportionment, back trajectory analysis

Source apportionment of PM_{2.5} size distribution and composition data from multiple stationary sites using a mobile platform

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Source: Volume 190, 1 July 2017, Pages 21-28
<https://doi.org/10.1016/j.atmosres.2017.02.008>

Several source apportionment studies considering multiple sites showed spatial variability of source contributions. However, setting up multiple fixed sites to collect comprehensive chemical speciation data is resource demanding. In this study, field campaigns were conducted at multiple receptor sites in the Mailiao and Taishi townships in Yunlin County using a mobile platform to demonstrate the feasibility of receptor modeling with particle size distribution and PM_{2.5} speciation data. Sources of air pollutants to all of the monitoring sites were identified and quantified using a modified positive matrix factorization (PMF) model. Modeling results indicated that a mixed source dominated by secondary aerosol was the largest contributor to PM_{2.5} at most sites. Adding VOC measurements with high time resolution helped to improve the source separation. Different patterns of source contributions among sites and seasons were observed showing both spatial heterogeneity and seasonal variation.

Keywords: Source apportionment, Mobile platform, Spatial variation, Positive matrix factorization (PMF), Multiple time resolution

Characterization of size-resolved urban haze particles collected in summer and winter at Taiyuan City, China using quantitative electron probe X-ray microanalysis

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Source: Volume, 1 July 2017, Pages 29-42
<https://doi.org/10.1016/j.atmosres.2017.02.005>

The aim of the study is to characterize the size-resolved urban haze particles and investigate their modification in morphology and composition in summer and winter using the semi-quantitative electron probe X-ray microanalysis (EPMA) based on both scanning and transmission electron microscopies equipped with ultrathin-window energy dispersive X-ray spectrometers (SEM-EDX and TEM-EDX). The haze and non-haze particles were collected through a seven-stage May cascade impactor on Dec. 29–30, 2009 and Jan. 8–9 and July 11–14, 2010 in Taiyuan, a typical inland city in the North China Plain. Approximately 3752 atmospheric particles in the size ranges of 4–2 μm , 2–1 μm , 1–0.5 μm , and 0.5–0.25 μm in aerodynamic diameter were measured and identified according to their secondary electron or TEM images and elemental atomic concentrations calculated through a Monte Carlo simulation program. Results show that on the haze days many reacted or aged mineral dust particles were encountered, in which the sulfate-containing ones outnumbered the

nitrate-containing ones in the winter samples while it was on the contrary in the summer samples, suggesting different haze formation and evolution mechanisms in summer and winter. Furthermore, in the haze events (especially in summer), many CNOS-rich particles, likely mixtures of water-soluble organic carbon with $(\text{NH}_4)_2\text{SO}_4$ or NH_4HSO_4 , were observed not only in the submicron but also in the super-micron fractions. The simultaneous observation of the fresh and aged CNOS-rich particles in the same SEM or TEM images implied that the status and components of secondary particles were complicated and changeable. The significant increase of both elemental concentration ratios of $[\text{N}]/[\text{S}]$ and $[\text{C}]/[\text{S}]$ in the aged ones compared to the fresh ones indicated that NH_4NO_3 and secondary organic matter were likely absorbed onto $(\text{NH}_4)_2\text{SO}_4$ or NH_4HSO_4 particles and mixed with them. K-rich, Fe-rich, and heavy metal-containing particles in TEM-EDX measurement were detected more in the winter haze samples than in the summer ones, suggesting that they tend to be smaller in size and mainly derive from anthropogenic biomass burning and coal combustion. It was concluded that the combined use of SEM-EDX and TEM-EDX can identify both submicron and super-micron urban haze particles in a straightforward way and trace their modifications in size, shape, mixing state, and chemical compositions in different seasons, helping address their evolution processes and hazards on human health.

Keywords: Urban haze particles, Taiyuan, Electron probe X-ray microanalysis, SEM-EDX, TEM-EDX

Carbonaceous species in PM_{2.5} and PM₁₀ in urban area of Zhengzhou in China: Seasonal variations and source apportionment

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Source: Volume, 15 July 2017, Pages 1-11

<https://doi.org/10.1016/j.atmosres.2017.02.003>

PM_{2.5} and PM₁₀ samples were simultaneously collected in an urban site in Zhengzhou, China from October 2014 to July 2015 representing the four seasons. Organic carbon (OC), elemental carbon (EC), and non-polar organic compounds including n-alkanes (C₈–C₄₀) and polycyclic aromatic hydrocarbons (PAHs) were quantified. The characteristics of their concentrations, seasonal variations, and sources of n-alkanes and PAHs were investigated. Diagnostic ratios and positive matrix factorization (PMF) were used to characterize carbonaceous species, identify their possible sources, and apportion the contributions from each possible source. The concentrations of the components exhibited distinct seasonal variation, that is, the concentrations are high in winter and low in summer. This finding could be associated with increase in air pollutant emissions during heating season and stable weather condition. The estimated total carbonaceous aerosol accounts for 32% of PM_{2.5} and 30% of PM₁₀. Hence, carbonaceous compounds were the major components of particulate matter in the study area. Moreover, OC, EC, PAHs, and n-alkanes preferentially accumulated into fine particles. The carbonaceous components exhibited high correlation in PM_{2.5} and PM₁₀, thereby indicating that their sources were similar. The PMF results revealed that the main sources of PAHs were coal combustion (40%) and motor vehicles (29%); n-alkanes were mainly from burning of fossil fuel (48%). These sources were consistent with the diagnostic ratios obtained. This study provides guidance for improving air quality and reducing human exposure to toxic air pollutants.

Keywords: PM_{2.5}, PM₁₀, PAHs, n-Alkanes, PMF, Zhengzhou

Modeling approach for emissions reduction of primary PM_{2.5} and secondary PM_{2.5} precursors to achieve the air quality target

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Source: Volume 192, 1 August 2017, Pages 11-18
<https://doi.org/10.1016/j.atmosres.2017.03.018>

The study proposes an approach to quantify necessary emission reductions of various related pollutants to meet national PM_{2.5} air quality standard (AQS). Many scenarios of emission reductions are simulated to establish the relationships between the reduction ratios of air pollutants and improvements of atmospheric PM_{2.5} concentration for specific air basins in Taiwan. The air pollutants include primary PM_{2.5} and secondary PM_{2.5} precursor species, such as nitrogen oxides (NO_x), sulfur oxides (SO_x), volatile organic compounds (VOCs), and ammonia (NH₃). These relationships were used to estimate the required amounts of emission reduction for various pollutants to achieve the national PM_{2.5} AQS of annual average with 15 µg m⁻³. Three air basins of KPAB, YCNAB and CTAB with severe PM_{2.5} pollution in Taiwan were selected as case studies. The results follow: (1) The primary PM_{2.5} emission reductions and the PM_{2.5} concentration improvements exhibit linear relationships, and its effects are relatively significant. (2) Among the secondary PM_{2.5} precursor species, emission reductions of NO_x and NH₃ will have greater effectiveness, but higher proportions (> 50%) of emission reductions are needed. (3) In terms of mitigating PM_{2.5} pollution, the priorities for pollutant emission control should be in the order of primary PM_{2.5}, NO_x, and SO_x, while NMHC and NH₃ should not currently be a focus. (4) If three air basins of KPAB, YCNAB and CTAB with high PM_{2.5} levels are required to meet the AQS, it is necessary to reduce primary PM_{2.5}, NO_x and SO_x emissions simultaneously in case that LRT effect remains constant. Under the premise of meeting the AQS for the three air basins, the reduction of primary PM_{2.5}, NO_x and SO_x emission in Taiwan should be up to 76%, 93% and 20% respectively. Fortunately, many efforts have been and will be done to improve the PM_{2.5} concentration in China. Those efforts will mitigate significantly the loading of emission reduction and AQS should be more possible to achieve for all air basins in Taiwan.

Keywords: Fine particulate matter, Air quality standard, Emission reduction, Air quality modeling

Effect of nitrogen oxides (NO and NO₂) and toluene on SO₂ photooxidation, nucleation and growth: A smog chamber study

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Source: Volume 192, 1 August 2017, Pages 38-47
<https://doi.org/10.1016/j.atmosres.2017.03.017>

The formation and growth of new particles has recently been shown to have a significant influence on Chinese haze pollution, and sulfuric acid has long been recognized as a major contributor to new particle formation. In this study, four comparison groups of experiments related to SO₂ photooxidation, as well as aerosol nucleation and growth, have been conducted in the CAPS-ZJU (Complex Air Pollution Study-Zhejiang University) smog chamber. These were conducted either under SO₂/NO_x or SO₂/toluene gas-phase environments in the absence of seed particles. During aerosol nucleation and growth process, several physical properties such as mass, size and effective density were measured simultaneously by Scanning Mobility Particle Sizer (SMPS) and Differential Mobility Analyzer-Aerosol Particle Mass Analyzer-Condensation Particle Counter (DMA-APM-CPC). The effective density of new particles decreased from 1.8 to 1.35 g/cm³ as the particle size increased from 20 to 65 nm. The single particle mass showed good power-law relationship with mobility diameter, with an average mass-mobility exponent of 2.885. A new algorithm and a reference density of 1.38 g/cm³ based on size-resolved single particle mass (SPM) were proposed to calculate the mass concentration of new particles. Two methods based on Log Normal and Max Concentration were applied to derive particle growth rate (GR), and data merging from both methods was implemented to decrease calculation uncertainty. Meanwhile, both continuous nucleation and inhibition of further growth in sub-20 nm size range were observed in different experiments depending on composition, and possible reasons were analyzed. The presence of NO was found to suppress nucleation and subsequent aerosol growth; while the presence of NO₂ or toluene promoted it. It was concluded that decreasing NO_x (NO or NO₂) or increasing toluene may promote SO₂ photooxidation, nucleation and subsequent aerosol growth, all of which is significant for deeper understanding of complex air pollution in China.

Keywords: Smog chamber, New particle formation, Complex air pollution, Effective density, PM_{2.5}

Estimating Particulate Matter using satellite based aerosol optical depth and meteorological variables in Malaysia

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Source: Volume 193, 1 September 2017, Pages 142-162

<https://doi.org/10.1016/j.atmosres.2017.04.019>

The insufficient number of ground-based stations for measuring Particulate Matter < 10 µm (PM₁₀) in the developing countries hinders PM₁₀ monitoring at a regional scale. The present study aims to develop empirical models for PM₁₀ estimation from space over Malaysia using aerosol optical depth (AOD₅₅₀) and meteorological (surface temperature, relative humidity and atmospheric stability) data (retrieved or estimated) from Moderate Resolution Imaging Spectroradiometer (MODIS) during the period 2007–2011. The MODIS retrievals are found to be satisfactorily correlated with ground-based measurements at Malaysia. Multiple linear regressions (MLR) and Artificial Neural Network (ANN) techniques are utilized to develop the empirical models for PM₁₀ estimation. The model development and training are performed via comparison with measured PM₁₀ at 29 stations over Malaysia and reveal that the ANN provides slightly higher accuracy with R² = 0.71 and RMSE = 11.61 µg m⁻³ compared to the MLR method (R² = 0.66 and RMSE = 12.39 µg m⁻³). Stepwise regression analysis performed on the MLR method reveals that the MODIS AOD₅₅₀ is the most important parameter for PM₁₀ estimations (R² = 0.59 and RMSE = 13.61 µg m⁻³); however, the inclusion of the meteorological parameters in the MLR

increases the accuracy of the retrievals ($R^2 = 0.66$, $RMSE = 12.39 \mu g m^{-3}$). The estimated PM10 concentrations are finally validated against surface measurements at 16 stations resulting in similar performance from the ANN model ($R^2 = 0.58$, $RMSE = 10.16 \mu g m^{-3}$) and MLR technique ($R^2 = 0.56$, $RMSE = 10.58 \mu g m^{-3}$). The significant accuracy that has been attained in PM10 estimations from space allows us to assess the pollution levels in Malaysia and map the PM10 distribution at large spatial and temporal scales.

Keywords: PM10, Satellite remote sensing, Artificial Neural Network, Multiple linear regressions, Meteorology, Malaysia

Sub-micron particle number size distribution characteristics at two urban locations in Leicester

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Source: Volume, 15 September 2017, Pages 1-16
<https://doi.org/10.1016/j.atmosres.2017.04.021>

The particle number size distribution (PNSD) of atmospheric particles not only provides information about sources and atmospheric processing of particles, but also plays an important role in determining regional lung dose. Owing to the importance of PNSD in understanding particulate pollution two short-term campaigns (March–June 2014) measurements of sub-micron PNSD were conducted at two urban background locations in Leicester, UK. At the first site, Leicester Automatic Urban Rural Network (AURN), the mean number concentrations of nucleation, Aitken, accumulation modes, the total particles, equivalent black carbon (eBC) mass concentrations were 2002, 3258, 1576, 6837 # cm^{-3} , 1.7 $\mu g m^{-3}$, respectively, and at the second site, Brookfield (BF), were 1455, 2407, 874, 4737 # cm^{-3} , 0.77 $\mu g m^{-3}$, respectively. The total particle number was dominated by the nucleation and Aitken modes, with both consisting of 77%, and 81% of total number concentrations at AURN and BF sites, respectively. This behaviour could be attributed to primary emissions (traffic) of ultrafine particles and the temporal evolution of mixing layer. The size distribution at the AURN site shows bimodal distribution at ~ 22 nm with a minor peak at ~ 70 nm. The size distribution at BF site, however, exhibits unimodal distribution at ~ 35 nm. This study has for the first time investigated the effect of Easter holiday on PNSD in UK. The temporal variation of PNSD demonstrated a good degree of correlation with traffic-related pollutants (NO_x, and eBC at both sites). The meteorological conditions, also had an impact on the PNSD and eBC at both sites. During the measurement period, the frequency of NPF events was calculated to be 13.3%, and 22.2% at AURN and BF sites, respectively. The average value of formation and growth rates of nucleation mode particles were 1.3, and 1.17 $cm^{-3} s^{-1}$ and 7.42, and 5.3 $nm h^{-1}$ at AURN, and BF sites, respectively. It can suggest that aerosol particles in Leicester originate mainly from traffic and domestic heating emissions.

Keywords: Particle size distribution, Easter holiday, Temporal variation, New particle formation

Estimate of main local sources to ambient ultrafine particle number concentrations in an urban area

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Source: Volume, 15 September 2017, Pages 178-189

<https://doi.org/10.1016/j.atmosres.2017.04.036>

Quantifying and apportioning the contribution of a range of sources to ultrafine particles (UFPs, $D < 100$ nm) is a challenge due to the complex nature of the urban environments. Although vehicular emissions have long been considered one of the major sources of ultrafine particles in urban areas, the contribution of other major urban sources is not yet fully understood. This paper aims to determine and quantify the contribution of local ground traffic, nucleated particle (NP) formation and distant non-traffic (e.g. airport, oil refineries, and seaport) sources to the total ambient particle number concentration (PNC) in a busy, inner-city area in Brisbane, Australia using Bayesian statistical modelling and other exploratory tools. The Bayesian model was trained on the PNC data on days where NP formations were known to have not occurred, hourly traffic counts, solar radiation data, and smooth daily trend. The model was applied to apportion and quantify the contribution of NP formations and local traffic and non-traffic sources to UFPs. The data analysis incorporated long-term measured time-series of total PNC ($D \geq 6$ nm), particle number size distributions (PSD, $D = 8$ to 400 nm), PM_{2.5}, PM₁₀, NO_x, CO, meteorological parameters and traffic counts at a stationary monitoring site. The developed Bayesian model showed reliable predictive performances in quantifying the contribution of NP formation events to UFPs (up to 4×10^4 particles cm^{-3}), with a significant day to day variability. The model identified potential NP formation and no-formations days based on PNC data and quantified the sources contribution to UFPs. Exploratory statistical analyses show that total mean PNC during the middle of the day was up to 32% higher than during peak morning and evening traffic periods, which were associated with NP formation events. The majority of UFPs measured during the peak traffic and NP formation periods were between 30–100 nm and smaller than 30 nm, respectively. To date, this is the first application of Bayesian model to apportion different sources contribution to UFPs, and therefore the importance of this study is not only in its modelling outcomes but in demonstrating the applicability and advantages of this statistical approach to air pollution studies.

Keywords: Ambient ultrafine particles, Bayesian statistical model, Non-traffic sources, nucleated particle formation, urban area

Estimates of spatially and temporally resolved constrained black carbon emission over the Indian region using a strategic integrated modelling approach

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Source: Volume, 195, 1 October 2017, Pages 9-19

<https://doi.org/10.1016/j.atmosres.2017.05.007>

We estimated the latest spatially and temporally resolved gridded constrained black carbon (BC) emissions over the Indian region using a strategic integrated modelling approach. This was done extracting information on initial bottom-up emissions and atmospheric BC concentration from a general circulation model (GCM) simulation in conjunction with the receptor modelling approach. Monthly BC emission (83–364 Gg) obtained from the present study exhibited a spatial and temporal variability with this being the highest (lowest) during February (July). Monthly BC emission flux was considerably high ($> 100 \text{ kg km}^{-2}$) over the entire Indo-Gangetic plain (IGP), east and the west coast during winter months. This was relatively higher over the central and western India than over the IGP during summer months. Annual BC emission rate was 2534 Gg y^{-1} with that over the IGP and central India respectively comprising 50% and 40% of the total annual BC emissions over India. A high relative increase was observed in modified BC emissions (more than five times the initial emissions) over the most part of the IGP, east coast, central/northwestern India. The relative predominance of monthly BC emission flux over a region (as depicted from z-score distribution maps) was inferred being consistent with the prevalence of region- and season-specific anthropogenic activity.

Keywords: Black carbon, Emission inventory, Receptor modeling approach, Bottom-up and top-down emissions, Monthly BC emission flux, GCM simulation, z score distribution

Association of atmospheric pollution and instability indices: A detailed investigation over an Indian urban metropolis

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Source: *Volume, 196, 1 November 2017, Pages 83-96*

<https://doi.org/10.1016/j.atmosres.2017.04.033>

Convection has a significant role in maintaining the atmospheric dynamics and thermodynamics, particularly in the tropical regions during pre-monsoon season, which may be due to the changing patterns in atmospheric instability and pollution. A critical analysis is done on the variability of instability indices and their significant signature to meteorological parameters and atmospheric pollution over Indian region in the warming atmosphere during 2005–2015. The present study represents that the solid as well as gaseous pollutants, in combination, produce a damping force in suppressing convective activities over the eastern coastal regions of India. A significant anti-correlation ($r \sim -0.6$ to -0.8) between instability parameters [Convective Available Potential Energy (CAPE) and Lifted Index (LI)] and atmospheric pollutants [gaseous (NO_2 and SO_2) and solid (BC and $\text{PM}_{2.5}$)] has been obtained in the eastern coastal regions on a long-term basis. To improve the level of agreement between pollution and instability, a unitless and dimensionless index called KLURT index has been introduced, which provides correlation (r) value as high as ~ 0.6 . On a real time basis, KLURT index is found to be useful as an effective precursor of thunderstorm events. The final part of this study indicates a prediction technique using KLURT index which gives a high prediction efficacy of 75%, low FAR value, extremely good BS 0.06, an excellent bias (~ 0.96) and a good lead time of 1 h for a threshold value of 12.5 in terms of predicting intense convections at the urban location, Kolkata. Thus, the present study provides an appropriate means to manifest convection as a function of changing anthropogenic factors both in long and short-term basis with high correlation values and provides significant efficacy in predicting severe weather, thereby

demonstrating the usefulness of this hypothesis in various socio-economic aspects especially at the current tropical urban location.

Keywords: Atmospheric pollution, Instability, Precursor, Convection, KLURT

Characterization, sources and health risk analysis of PM_{2.5} bound metals during foggy and non-foggy days in sub-urban atmosphere of Agra

Awni Agarwal, Ankita Mangal, Aparna Satsangi, Anita Lakhani, K. Maharaj Kumari

Source: Volume, 197, 15 November 2017, Pages 121-131

<https://doi.org/10.1016/j.atmosres.2017.06.027>

A study was conducted at a campus site of Agra to determine the metals in fine mode particles (PM_{2.5}) and assess their associated health effects in adults and children during the period from 7th December 2015 to 17th February 2016. PM_{2.5} mass concentrations (190 µg/m³ and 132 µg/m³) exceeded the air quality standards of India by a factor of 3.2 and 2.2 during the foggy and non-foggy days, respectively. Total concentration varied from 11.9 µg/m³ to 40.6 µg/m³ during the fog period and from 5.7 µg/m³ to 25.8 µg/m³ during the non-foggy period for seventeen metals (including metals with concentration > 1 µg/m³, 0.1–1 µg/m³, 0.01–0.1 µg/m³ and < 0.01 µg/m³) with maximum contribution from K and minimum from Co. At this site in winter, coal and biomass combustion, vehicular emissions, crustal dust and iron producing industrial processes were identified as the major contributors to metals as determined by correlation analysis and Principal Component Analysis. Cr and Mn showed the highest carcinogenic and non-carcinogenic risks. Cumulated non-carcinogenic risk in children (HI: 3.90 for foggy days; HI: 2.74 for non-foggy days) was higher compared to the adults (HI: 1.81 for foggy days; HI: 1.29 for non-foggy days) during both periods while the cumulated effect of carcinogenic risk was higher for adults (ILCR: 2.25E – 04) than for children (ILCR: 1.31E – 04).

Keywords: PM_{2.5}, Heavy metals, Source Apportionment, Foggy days, Health risk Assessment

Determination of trace metals in TSP and PM_{2.5} materials collected in the Metropolitan Area of Monterrey, Mexico: A characterization study by XPS, ICP-AES and SEM-EDS

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Source: Volume, 196, 1 November 2017, Pages 8-22

<https://doi.org/10.1016/j.atmosres.2017.05.009>

The concentration levels of trace metals of toxicological importance were evaluated in the total suspended particles (TSP) and particulate matter smaller than 2.5 µm (PM_{2.5}) collected in the Metropolitan Area of Monterrey (MAM) in Mexico. Samples were characterized by inductively

coupled plasma atomic emission spectroscopy (ICP-AES), X-ray photoelectron spectroscopy (XPS) and scanning electron microscopy with an energy-dispersive spectroscopy system (SEM-EDS). In addition, the data were statistically treated by the methodology of Pearson Correlation (PC) and Principal Components Analysis (PCA) to identify the possible emitting sources. Surface analysis of the particulate matter (PM) by XPS revealed that the most abundant elements were Ca, Al, Na, Zn, Cu and Mg. The deconvolution of the Ca2p, Zn2p and Cu2p signals showed that the main contributors were CaCO₃, ZnO and Cu/Cu₂O, respectively. The bulk analysis of the PM by ICP-AES showed Fe, Cu and Zn as the most abundant elements. Fe-rich particles presented two different morphologies: the prismatic particles were associated with a natural origin, while the spherical particles with anthropogenic sources. The Zn and Cu were predominantly observed in the sampling stations with high vehicular traffic, and the emitting sources were associated with the burning of fuels from automobiles and the wear of the tires and brakes. The highest concentration of Pb was detected in the sampling station located near the industrial zones, and its cause was associated with the ceramic and glass industries, the burning of fuel oil in power plants and the production of lead-based batteries for automobiles.

Keywords : Trace metals , Total suspended particles , X-ray photoelectron spectroscopy , Principal components analysis

Association of atmospheric pollution and instability indices: A detailed investigation over an Indian urban metropolis

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Source: Volume 196, 1 November 2017, Pages 83-96

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Convection has a significant role in maintaining the atmospheric dynamics and thermodynamics, particularly in the tropical regions during pre-monsoon season, which may be due to the changing patterns in atmospheric instability and pollution. A critical analysis is done on the variability of instability indices and their significant signature to meteorological parameters and atmospheric pollution over Indian region in the warming atmosphere during 2005–2015. The present study represents that the solid as well as gaseous pollutants, in combination, produce a damping force in suppressing convective activities over the eastern coastal regions of India. A significant anti-correlation ($r \sim -0.6$ to -0.8) between instability parameters [Convective Available Potential Energy (CAPE) and Lifted Index (LI)] and atmospheric pollutants [gaseous (NO₂ and SO₂) and solid (BC and PM_{2.5})] has been obtained in the eastern coastal regions on a long-term basis. To improve the level of agreement between pollution and instability, a unitless and dimensionless index called KLURT index has been introduced, which provides correlation (r) value as high as ~ 0.6 . On a real time basis, KLURT index is found to be useful as an effective precursor of thunderstorm events. The final part of this study indicates a prediction technique using KLURT index which gives a high prediction efficacy of 75%, low FAR value, extremely good BS 0.06, an excellent bias (~ 0.96) and a good lead time of 1 h for a threshold value of 12.5 in terms of predicting intense convections at the urban location, Kolkata. Thus, the present study provides an appropriate means to manifest convection as a function of changing anthropogenic factors both in long and short-term basis with high correlation values and provides significant efficacy in predicting severe weather, thereby

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Keywords: Heavy metals, Source Apportionment, Foggy days , Health risk Assessment

High ozone episodes at a semi-urban site in India: Photochemical generation and transport

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Source: Volume 197, 15 November 2017, Pages 232-243

<https://doi.org/10.1016/j.atmosres.2017.07.014>

The present study focused on investigating the possible atmospheric conditions influencing the high ozone episodes over a semi-urban site (27.16° N, 78.08° E) in Agra, India during July 2014–June 2016. During the study period, average concentration of ozone was 32.3 ± 22.7 ppb, and concentrations of its precursors viz carbon monoxide (CO), nitrogen oxide (NO) and total nitrogen

oxides (NO_x) were 527.3 ± 482.7 ppb, 6.5 ± 8.2 ppb, and 12.1 ± 7.8 ppb, respectively. The maximum monthly average ozone concentration was observed in June (48.2 ± 31.0 ppb) and it was nearly three times higher than the minimum levels observed in December (16.7 ± 10.5 ppb). Considering high ozone episodes, 78 days exceed hourly ozone limit of 90 ppb specified by National Air Quality Standards (NAAQS, CPCB, 2009) for India while 75 days exceed the daily maximum 8 hourly ozone limit of 70 ppb or above (NAAQS, EPA, 2015). Trajstat model was used for air-mass cluster analysis during episodic days; five clusters of air-masses were identified and among them, the cluster from northwest direction had the maximum frequency (35.4%). During the study period, four different types of high ozone episodes were identified and explained using prevailing meteorology. The episodes were attributed to local photochemistry and/or transport.

Keywords : Ozone episode , Back-trajectory analysis , Photochemical generation , Long-range transport

Influence of air mass origins on optical properties and PM concentrations measured at a high mountain station located in the southwestern Mediterranean

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Source: Volume 197, 15 November 2017, Pages 244-254
<https://doi.org/10.1016/j.atmosres.2017.07.013>

The influence of air mass origins on aerosol optical properties and particulate matter (PM) concentrations measured from January 2014 to December 2015 at a high mountain station in the southwestern Mediterranean was analyzed. Mean values of extensive aerosol optical properties (scattering, backscattering and absorption coefficients) and PM mass fractions: σ_{sp} ($30.7 \pm 1.1 \text{ Mm}^{-1}$), σ_{bsp} ($3.9 \pm 0.1 \text{ Mm}^{-1}$), σ_{ap} ($2.4 \pm 0.1 \text{ Mm}^{-1}$), PM₁₀ ($13.1 \pm 0.6 \mu\text{g}\cdot\text{m}^{-3}$) and PM₁ ($4.0 \pm 0.1 \mu\text{g}\cdot\text{m}^{-3}$) were in the range of values reported in most studies carried out at high altitude locations. Nevertheless, a significant variation of these concentrations according to the air mass origin has been noticed. Transport from North Africa (NAF back-trajectories) and regional recirculations (REG) were the two air mass types in which the highest values of PM concentrations and optical properties were registered. Alternatively, the lowest values were recorded under Atlantic advections. Noticeable differences between NAF and REG categories were observed when intensive optical properties of aerosols were analyzed. During NAF scenarios the lowest value of SAE (0.91 ± 0.06) was obtained as well as the greatest AAE value (1.30 ± 0.02), with daily levels higher than 1.90. This result suggests that OC and hematite compounds were relevant in the absorption process during NAF events. However, during REG episodes the AAE mean value declined to 1.18 ± 0.01 , since the highest mean concentration of BC was recorded under this category, and the SAE value was maximum (1.70 ± 0.05) due to the smaller size of PM. It has also been documented that during the study period a great number of REG back-trajectories occurred just after the transport of air masses from North Africa. In these situations, AAE and SAE values showed slight variations due to the persistence of mineral dust in the local atmosphere.

Keywords: High mountain station, Optical properties of aerosols, Air mass origin, Dust outbreak, Regional circulation

Source region and sector contributions of atmospheric soot particle in a coalfield region of Dhanbad, eastern part of India

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Source: Volume 197, 15 November 2017, Pages 415-424
<https://doi.org/10.1016/j.atmosres.2017.07.020>

Black carbon (BC) aerosols affect the Earth's climate directly by interacting with the solar radiation and indirectly by modifying the lifetime and optical properties of clouds. However, our understanding of BC aerosols and their impacts on the climate are limited by lack of in situ measurements of BC, especially in the developing world. This study reports measurements of BC from Dhanbad, a coalfields area of eastern India, we analyze BC data at 370 and 880 nm during 2013 to gain insight into the emission sources affecting the study area. Our analysis indicates significantly higher absorption at the lower wavelength (ultraviolet). We estimate that ~ 33% of BC at Dhanbad comes from biomass/biofuel combustion and the remaining 67% from the fossil fuel combustion. Higher concentrations of BC_{370 nm} ($> 12 \mu\text{g m}^{-3}$) were observed when the air masses affecting Dhanbad originated far away in countries like Iran, Afghanistan, Pakistan, Oman, United Arab Emirates and passed over the Indo-Gangetic Plains (IGP) prior to arriving at the observation site. The source regions affecting BC_{880 nm} were localized over the IGP but BC_{880 nm} concentrations are 33% lower ($\sim 8 \mu\text{g m}^{-3}$) than BC_{370 nm}. The cluster analysis showed that the largest fraction (35 and 29%) of the air masses arriving at Dhanbad passed through the boundary layer of the central IGP and north-west IGP region during the post-monsoon season. Average values of BC_{370 nm} (16.0 and 20.0 $\mu\text{g m}^{-3}$) and BC_{880 nm} (9.5 and 10.0 $\mu\text{g m}^{-3}$) in the IGP influenced air masses were significantly higher than those arriving from other source regions. The Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) model were applied to understand the relative importance of different sources affecting Dhanbad. The variability of observed BC mass concentrations was captured fairly well by WRF-Chem with minor deviations from the measured values. Model results indicate that anthropogenic emissions account for more than 75% of the surface BC at Dhanbad. Biomass burning contribution peaks in March–April and October–November but remains less than 25%. Long-range transport estimated in terms of inflow from domain boundaries does not affect BC concentrations at Dhanbad significantly.

Keywords : Atmospheric aerosol , Anthropogenic , Soot particle , Source identification

The association of weather patterns with haze episodes: Recognition by PM2.5 oriented circulation classification applied in Xiamen, Southeastern China

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Source: Volume 197, 15 November 2017, Pages 425-436

<https://doi.org/10.1016/j.atmosres.2017.07.024>

Investigating the association between weather circulation patterns and high PM_{2.5} episodes is useful for interpreting the connection between physical weather and chemical weather. Principal component analysis (PCA) is often applied to decompose circulation modes but has limitations for studying high PM_{2.5} events related circulation patterns. This study describes an improved circulation classification integrated with PCA and *k*-means algorithm oriented to high local PM_{2.5}. The classification scheme was applied in Xiamen, southeastern China, when local PM_{2.5} exceeded 75 µg m⁻³ (the 24-hour limit Chinese Ambient Air Quality Grade II standard) during the winter 2013. Nine typical circulation patterns were classified. Circulation patterns related to the highest PM_{2.5} concentrations were associated with a negative pressure anomaly at 850 hPa over the Sea of Japan which yielded a strong transport of PM_{2.5} from northern China during windy days. The improved classification methodology links large-scale circulation to local PM_{2.5} in target city and is able to distinguish possibly different circulation patterns over continual haze episodes. This clustering method can be applied in any cities and would be useful for predicting chemical weather and serving local environmental policymakers.

Keywords : Haze , Principal component analysis , Circulation decomposition , Circulation classification

Atmospheric deposition of trace elements at urban and forest sites in central Poland – Insight into seasonal variability and sources

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Source: Volume 198, 1 December 2017, Pages 123-131

<https://doi.org/10.1016/j.atmosres.2017.07.033>

This paper includes the results of chemical composition of bulk deposition samples collected simultaneously at urban (Poznań city) and forest (Jeziory) sites in central Poland, between April 2013 and October 2014. Rainwater samples were analyzed for trace elements (As, Zn, Ni, Pb, Cu, Cr, Cd) and physicochemical parameters. Overall, three metals, i.e. Zn, Pb and Cu were the most abundant anthropogenic constituents of rainwater samples from both locations. In Poznań city, the rainwater concentrations of trace elements did not differ significantly between spring and summer. However, they were elevated and more variable during the cold season (fall and winter), suggesting strong contribution from local high-temperature processes related to coal combustion (commercial and residential sector). In contrast to the urban site, relatively low variability in concentrations was found for Cu, Ni, Zn at the forest site, where direct impact of emission from vehicle traffic and coal-fired combustion (power plants) was much lower. The bulk deposition fluxes of Ni, As, Pb and Zn at

this site exhibited a clear trend, with higher values during the cold season (fall and winter) than in spring and summer. At the urban site, the sums of total bulk deposition fluxes of Zn, Cu, Pb, Ni, As, Cr, Cd were as follows: 8460.4, 4209.2, 2247.4, 1882.1, 606.6, 281.6 and 31.4 $\mu\text{g m}^{-2}$. In addition, during the winter season, a significantly higher deposition flux of Cu and Zn were observed for rain (on average 103.8 and 129.4 $\mu\text{g m}^{-2}$, respectively) as compared to snow (19.7 $\mu\text{g Cu m}^{-2}$ and 54.1 $\mu\text{g Zn m}^{-2}$). This suggests that different deposition pattern of trace elements for rain, mixed and snow was probably the effect of several factors: precipitation type, changes in emission and favorable meteorological situation during rain events.

Keywords: Trace elements, Bulk deposition, Emission sources, Rainwater, Seasonal variation

Chemical characteristics of PM_{2.5} during summer at a background site of the Yangtze River Delta in China

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Source: Volume 198, 1 December 2017, Pages 163-172

<https://doi.org/10.1016/j.atmosres.2017.08.012>

With rapid economic development and urbanization, particular attention has been paid to atmospheric chemical studies in the Yangtze River Delta in China. PM_{2.5} samples were collected by a MiniVol™ air sampler in summer time at a background site of the Yangtze River Delta in China. Carbonaceous components, i.e., OC and EC, levoglucosan and water-soluble inorganic ions, including sulfate, nitrate, ammonium, etc., were quantified. The average concentration of PM_{2.5} in summer at Lin'an was $30.19 \pm 8.86 \mu\text{g m}^{-3}$, lower than previous studies reported, confirming that air pollution in China is improving, e.g., by emission control measures and subsequent reduction in PM emissions in China. Investigating the relationship among sulfate, nitrate and ammonium showed that SO₄²⁻ existed as (NH₄)₂SO₄, while NO₃⁻ may have been present as NaNO₃ and KNO₃. Based on molecular tracers, synoptic data as well as air mass back trajectory analysis, it was revealed that regional transport and stable synoptic conditions both play an important role in controlling the variations of aerosol chemical components. The comparison of chemical species between clean and hazy days showed that secondary organic and inorganic aerosols have different production processes. Secondary organic carbon (SOC) was much more important during clean days, while secondary inorganic aerosol species were readily produced and consequently became more important during polluted periods in Lin'an during summer time.

Keywords: Organic aerosol, Water-soluble inorganic ions, Haze, YRD

Temporal and spatial analyses of particulate matter (PM₁₀ and PM_{2.5}) and its relationship with meteorological parameters over an urban city in northeast China

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Source: Volume 198, 1 December 2017, Pages 185-193

<https://doi.org/10.1016/j.atmosres.2017.08.023>

Temporal and spatial characteristics of atmospheric particulate matter (PM₁₀ and PM_{2.5}) and its relationship with meteorology over Shenyang, a city in northeast China, were statistically analyzed using hourly and daily averaged PM mass concentrations measured at 11 locations and surface meteorological parameters, from January 2014 to May 2016. Using averaged data from 11 stations in Shenyang, it was found that the monthly mean PM_{2.5} mass concentrations were higher in winter ($97.2 \pm 11.2 \mu\text{g m}^{-3}$) and autumn ($85.5 \pm 42.9 \mu\text{g m}^{-3}$), and lower in spring ($62.0 \pm 14.0 \mu\text{g m}^{-3}$) and summer ($42.5 \pm 8.4 \mu\text{g m}^{-3}$), similar to the seasonal variation in PM₁₀ concentrations. The monthly ratios of PM_{2.5}/PM₁₀ ranged from 0.41 to 0.87, and were larger in autumn and winter but lowest in spring due to dust activities. PM pollution was concentrated mainly in the central, northern, and western areas of Shenyang in most seasons mainly due to anthropogenic activities such as traffic and residential emission and construction activity as well as natural dust emission. PM concentrations observed over different areas in all seasons generally exhibited two peaks, at 08:00–10:00 local time (LT) and 21:00–23:00 LT, with the exception of PM_{2.5} in summer, which showed only one peak during the daytime. In addition, PM₁₀ concentrations peaked around 14:00 LT during spring in the western area of Shenyang because of strong thermal and dynamic turbulence, resulting in elevated dust emissions from adjacent dust sources. The relationship between daily PM concentrations and meteorological parameters showed both seasonal and annual variation. Overall, both PM_{2.5} and PM₁₀ concentrations were negatively correlated with atmospheric visibility, with correlation coefficients (R) of 0.71 and 0.56, respectively. In most seasons, PM concentrations also exhibited negative correlations with wind speed, but showed positive correlations with air pressure, air temperature, and relative humidity. Strong wind speed favored the dispersion of PM pollution, but also aided the release of coarse dust particles in spring. High air pressure and downdrafts restrained the upward movement of atmospheric PM, resulting in an accumulation of particles in the boundary layer. High air temperature favored the transformation of secondary particles through photochemical processes in summer, but also resulted in efficient vertical dispersion of pollutants in autumn and winter, leading to an inverse relationship between temperature and PM concentrations. Large relative humidity usually caused increases in PM concentrations due to the hygroscopic effect of aerosols, but not for PM₁₀ in spring and summer, mainly due to the suppression of dust emissions under wet air conditions in spring and the effects of wet scavenging under high summer rainfall.

Keywords: Particulate matter, Mass concentration, Meteorological parameter, Statistical analysis, Northeast China

Atmospheric Environment

Enhanced concentrations of reactive nitrogen species in wildfire smoke

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Source: Volume 148, January 2017, Pages 8-15
<https://doi.org/10.1016/j.atmosenv.2016.10.030>

During the summer of 2012 the Hewlett Gulch and High Park wildfires burned an area of 400 km² northwest of Fort Collins, Colorado. These fires both came within 20 km of the Department of Atmospheric Science at Colorado State University, allowing for extensive measurements of smoke-impacted air masses over the course of several weeks. In total, smoke plumes were observed at the measurement site for approximately 125 h. During this time, measurements were made of multiple reactive nitrogen compounds, including gas phase species NH₃, NO_x, and HNO₃, and particle phase species NO₃⁻ and NH₄⁺, plus an additional, unspciated reactive nitrogen component that is measured by high temperature conversion over a catalyst to NO. Concurrent measurements of CO, levoglucosan and PM_{2.5} served to confirm the presence of smoke at the monitoring site. Significant enhancements were observed for all of the reactive nitrogen species measured in the plumes, except for NH₄⁺ which did not show enhancements, likely due to the fresh nature of the plume, the presence of sufficient regional ammonia to have already neutralized upwind sulfate, and the warm conditions of the summer measurement period which tend to limit ammonium nitrate formation. Excess mixing ratios for NH₃ and NO_x relative to excess mixing ratios of CO in the smoke plumes, $\Delta\text{NH}_3/\Delta\text{CO}$ (ppb/ppb) and $\Delta\text{NO}_x/\Delta\text{CO}$ (ppb/ppb), were determined to be 0.027 ± 0.002 and 0.0057 ± 0.0007 , respectively. These ratios suggest that smoldering combustion was the dominant source of smoke during our plume interceptions. Observations from prior relevant laboratory and field measurements of reactive nitrogen species are also briefly summarized to help create a more comprehensive picture of reactive nitrogen and fire.

Keywords: Biomass burning, Ammonia, Reactive nitrogen, Emissions

Solar and thermal radiative effects during the 2011 extreme desert dust episode over Portugal

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Source: Volume 148, January 2017, Pages 16-29
<https://doi.org/10.1016/j.atmosenv.2016.10.037>

This paper analyses the influence of the extreme Saharan desert dust (DD) event on shortwave (SW) and longwave (LW) radiation at the EARLINET/AERONET Évora station (Southern Portugal)

from 4 up to 7 April 2011. There was also some cloud occurrence in the period. In this context, it is essential to quantify the effect of cloud presence on aerosol radiative forcing. A radiative transfer model was initialized with aerosol optical properties, cloud vertical properties and meteorological atmospheric vertical profiles. The intercomparison between the instantaneous TOA shortwave and longwave fluxes derived using CERES and those calculated using SBDART, which was fed with aerosol extinction coefficients derived from the CALIPSO and lidar-PAOLI observations, varying OPAC dataset parameters, was reasonably acceptable within the standard deviations. The dust aerosol type that yields the best fit was found to be the mineral accumulation mode. Therefore, SBDART model constrained with the CERES observations can be used to reliably determine aerosol radiative forcing and heating rates. Aerosol radiative forcings and heating rates were derived in the SW (ARFSw, AHRSw) and LW (ARFLw, AHRLw) spectral ranges, considering a cloud-aerosol free reference atmosphere. We found that AOD at 440 nm increased by a factor of 5 on 6 April with respect to the lower dust load on 4 April. It was responsible by a strong cooling radiative effect pointed out by the ARFSw value (-99 W/m^2 for a solar zenith angle of 60°) offset by a warming radiative effect according to ARFLw value ($+21.9 \text{ W/m}^2$) at the surface. Overall, about 24% and 12% of the dust solar radiative cooling effect is compensated by its longwave warming effect at the surface and at the top of the atmosphere, respectively. Hence, larger aerosol loads could enhance the response between the absorption and re-emission processes increasing the ARFLw with respect to those associated with moderate and low aerosol loads. The unprecedented results derived from this work complement the findings in other regions on the modifications of radiative energy budget by the dust aerosols, which could have relevant influences on the regional climate and will be topics for future investigations.

Keywords :Dust , Aerosol vertical profiles , EARLINET AERONET Forcing

Characteristics of aerosol size distribution and vertical backscattering coefficient profile during 2014 APEC in Beijing

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Source: Volume 148, January 2017, Pages 30-41

<https://doi.org/10.1016/j.atmosenv.2016.10.020>

During the 2014 Asia-Pacific Economic Cooperation (APEC) conference period, Beijing's air quality was greatly improved as a result of a series of tough emission control measures being implemented in Beijing and its surrounding provinces. However, a moderate haze occurred during the period of 4–5 November. In order to evaluate the emission control measures and study the formation mechanism of the haze, a comprehensive field observation based on a supersite and a lidar network was carried out from 25 October 2014 to 20 January 2015. By investigating the variations in aerosol number concentration and mean backscattering coefficient before, during and after the APEC period, it was found that number concentration of accumulation mode and coarse mode particles experienced the most significant decrease by 47% and 68%, and mean backscattering coefficient below 1 km decreased by 34% during the APEC period. Being characterized as “rapidly accumulating and rapidly dispersing”, the moderate haze occurred during the APEC period was probably initiated by a wind direction change to south and an increase of wind speed to 4 m/s.

Sulfur dioxide involved plume nucleation without growth in size as well as a burst of particles ranging between 100 and 300 nm were observed simultaneously during the haze episode. The elevation of sulfur dioxide concentration and particle number concentration was highly correlated with the southerly wind, signifying the contribution of regional transport. It was observed by the lidar network that the aerosol backscattering coefficient increased in sequence among three sites along the southwest pathway, suggesting that aerosols might be transported from the southwest to the northeast of Beijing with a speed of approximately 17 km/h, which agreed with the movement of air masses modeled by Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT). The dual-wavelength lidar (355 and 532 nm) observation suggested that transportation of fine particles from high-level atmosphere (approximately 2 km) could be the potential sources of the haze. Our result showed that regional transport would contribute to haze formation in Beijing under such meteorological conditions, thus, to maintain the “APEC blue”, significant attention should be paid to controlling regional transport through the southwest pathway.

Keywords : Haze , APEC , Regional transport , Southwest pathway , Beijing

Effect of fiber material on ozone removal and carbonyl production from carpets

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Source: Volume 148, January 2017, Pages 42-48

<https://doi.org/10.1016/j.atmosenv.2016.10.034>

Indoor air quality is affected by indoor materials such as carpets that may act as sources and/or sinks of gas-phase air pollutants. Heterogeneous reactions of ozone with carpets may result in potentially harmful products. In this study, indoor residential carpets of varying fiber types were tested to evaluate their ability to remove ozone, and to assess their role in the production of carbonyls when exposed to elevated levels of ozone. Tests were conducted with six types of new unused carpets. Two sets of experiments were conducted, the first measured ozone removal and ozone deposition velocities, and the second measured primary carbonyl production and secondary production as a result of exposure to ozone. The tests were conducted using glass chambers with volume of 52 L each. Air exchange rates for all tests were 3 h⁻¹. The ozone removal tests show that, for the conditions tested, the polyester carpet sample had the lowest ozone removal (40%), while wool carpet had the greatest ozone removal (65%). Most carpet samples showed higher secondary than primary carbonyl emissions, with carpets containing polypropylene fibers being a notable exception. Carpets with polyester fibers had both the highest primary and secondary emissions of formaldehyde among all samples tested. While it is difficult to make blanket conclusions about the relative air quality merits of various carpet fiber options, it is clear that ozone removal percentages and emissions of volatile organic compounds can vary drastically as a function of fiber type.

Keywords : Indoor air quality , Ozone deposition velocity , Formaldehyde , Aldehydes , Measurements

Radiation fog chemical composition and its temporal trend over an eight year period

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Source: Volume 148, January 2017, Pages 49-61

<https://doi.org/10.1016/j.atmosenv.2016.10.031>

Radiation fog samples have been collected at a rural site in Central Pennsylvania from 2007 through 2015 in order to document chemical composition, assess concentration changes over time, and to provide insight into emission sources that influence the region. The collection of samples over multiple years makes this one of the few long duration radiation fog studies that have been completed. During the course of the campaign, 146 samples were obtained and analyzed for pH, major inorganic ions, low molecular weight organic acids, total organic carbon (TOC) and total nitrogen (TN). Ammonium (median concentration = 209 μN), sulfate (69 μN), calcium (51 μN), and nitrate (31 μN) were the most abundant inorganic ions, although these were present at much lower concentrations than for radiation fog studies conducted in other locations. Organic acids, of which formate (20 μM) and acetate (21 μM) were the most abundant, were closer in magnitude to measurements made during previous studies. Organic acids accounted for 15% of TOC, which had a median concentration of 6.6 mgC l⁻¹. The median concentration of TN was 3.6 mgN l⁻¹, 18% of which was determined to be organic nitrogen. Statistically significant decreasing trends from 2007 to 2015 were noted for sulfate, ammonium, chloride, and nitrate. For the same period, an increase in pH was observed. Seasonal trends were identified for a number of species as well. The partitioning of ammonia between the gas and aqueous phases was also investigated and found to deviate significantly from equilibrium.

Keywords :Radiation fog chemistry, Organic acids, Total organic carbon, Total nitrogen, Multi-year trends, Ammonia partitioning

Characteristics, sources and evolution of fine aerosol (PM₁) at urban, coastal and forest background sites in Lithuania

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Source: Volume 148, January 2017, Pages 62-76

<https://doi.org/10.1016/j.atmosenv.2016.10.038>

The chemical and isotopic composition of organic aerosol (OA) samples collected on PM₁ filters was determined as a function of desorption temperature to investigate the main sources of organic carbon and the effects of photochemical processing on atmospheric aerosol. The filter samples were collected at an urban (54°38' N, 25°18' E), coastal (55°55' N, 21°00' E) and forest (55°27' N, 26°00' E) site in Lithuania in March 2013. They can be interpreted as winter-time samples because the monthly averaged temperature was -4 °C.

The detailed chemical composition of organic compounds was analysed with a thermal desorption PTR-MS. The mass concentration of organic aerosol at the forest site was roughly by a factor of 30 lower than at the urban and coastal site. This fact could be an indication that in this cold month the biogenic secondary organic aerosol (SOA) formation was very low. Moreover, the organic aerosol collected at the forest site was more refractory and contained a larger fraction of heavy molecules with $m/z > 200$.

The isotopic composition of the aerosol was used to differentiate the two main sources of organic aerosol in winter, i.e. biomass burning (BB) and fossil fuel (FF) combustion. Organic aerosol from biomass burning is enriched in ^{13}C compared to OA from fossil fuel emissions. $\delta^{13}\text{COC}$ values of the OA samples showed a positive correlation with the mass fraction of several individual organic compounds. Most of these organic compounds contained nitrogen indicating that organic nitrogen compounds formed during the combustion of biomass may be indicative of BB. Other compounds that showed negative correlations with $\delta^{13}\text{COC}$ were possibly indicative of FF. These compounds included heavy hydrocarbons and were on the average less oxidized than the bulk organic carbon.

The correlation of $\delta^{13}\text{COC}$ and the O/C ratio was positive at low but negative at high desorption temperatures at the forest site. We propose that this might be due to photochemical processing of OA. This processing can lead to accumulation of carbon in the more refractory organic fraction that is depleted in ^{13}C compared with the less refractory organic fraction. Detailed laboratory experiments are necessary to further investigate the aging of aerosol particles before firm conclusions can be drawn.

Keywords : Aerosol, Isotopic composition, IRMS, PTR-MS

The impact of Mount Etna sulfur emissions on the atmospheric composition and aerosol properties in the central Mediterranean: A statistical analysis over the period 2000–2013 based on observations and Lagrangian modelling

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Source: Volume 148, January 2017, Pages 77–88

<https://doi.org/10.1016/j.atmosenv.2016.10.032>

The emission of gases and aerosols due to volcanic activity may impact significantly atmospheric composition, cloud occurrence and properties, and the regional and global climate. While the effects of strong explosive (stratospheric) eruptions are relatively well known, limited information on the impacts of small to moderate volcanic activities, including passive degassing, is available. In this paper, the downwind impact of Mount Etna's sulfur emissions on the central Mediterranean is investigated on a statistical basis over the period 2000–2013 using: (a) daily sulfur dioxide emission rates measured near crater at Mount Etna with ground-based ultraviolet spectrophotometers, (b) Lagrangian trajectories and simulated plume dispersion obtained with the FLEXPART (FLEXible PARTicle dispersion) model, and (c) long-term observations of column SO_2 concentration and aerosol Ångström exponent α at Lampedusa (35.5° N, 12.6° E). This

statistical analysis has allowed, for the first time, the characterization of decadal impact of Mount Etna's sulfur emissions on the sulfur dioxide and the aerosol microphysical/optical properties in the central Mediterranean. On average, statistically significant higher SO₂ concentrations and smaller aerosol sizes are present when air masses from Mount Etna overpass Lampedusa. Despite being upwind of Lampedusa for only 5% of the time, Mount Etna is potentially responsible for up to 40% and 20% of the SO₂ and α extreme values (exceedances of a fixed threshold), respectively, at this location. The most important factor determining this perturbation is the prevailing dynamics, while the magnitude of the SO₂ emission rates from Mount Etna appears to be likely important only for relatively strong emissions. The observed perturbations to the aerosol size distribution are expected to produce a direct regional radiative effect in this area.

Keywords : Volcanic emissions, Sulfur cycle, Secondary sulfate aerosols, Regional climate, Mediterranean, Mount Etna

Multi-tracer approach to characterize domestic wood burning in Athens (Greece) during wintertime

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Source: Volume 148, January 2017, Pages 89-101
<https://doi.org/10.1016/j.atmosenv.2016.10.011>

During the last years the atmosphere of the Great Athens Area (GAA) and other Greek cities is burdened from extended residential biomass burning for heating purposes. In this work, a series of near real-time and off-line biomass burning tracers are analyzed during intense wood burning events in Athens. The measurements were conducted at an urban background site located in the center of Athens, and in the heart of wood burning activities (winter 2013–2014). The measured tracers include high resolution measurements of non-sea salt potassium (nss-K⁺), wood burning black carbon (BC_{wb}), the m/z 60 fragment associated with levoglucosan and monosaccharide anhydrides (levoglucosan, mannosan and galactosan) determined on selected filter samples. The suitability of these tracers was evaluated when the prevailing meteorological conditions with low dispersion and deposition mechanisms (low wind speed, absence of precipitation) were associated with high biomass burning emissions at nighttime. During the severe smog periods, the levels of K⁺, BC_{wb}, m/z 60 and levoglucosan were up to 2.2 µg m⁻³, 12.5 µg m⁻³, 3.4 µg m⁻³ and 8.6 µg m⁻³, respectively, higher by a factor of at least two, relatively to the non smog periods due to biomass burning. Correlations between biomass burning tracers as well as between monosaccharide anhydrides provided information about the type of material and wood being burned.

Keywords : Urban pollution, Wood burning tracers, Ions, Potassium, Black carbon

Oxidative potential of on-road fine particulate matter (PM_{2.5}) measured on major freeways of Los Angeles, CA, and a 10-year comparison with earlier roadside studies

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Source: Volume 148, January 2017, Pages 102-114

<https://doi.org/10.1016/j.atmosenv.2016.10.042>

This study describes on-road measurements of fine particulate matter (PM_{2.5}) using a mobile instrumentation platform to assess the chemical composition and oxidative potential of PM_{2.5}, using the dithiothreitol (DTT) assay, over three representative roadways in the Los Angeles Basin: the I-110 and I-710 freeways, the Wilshire/Sunset boulevards as well as the main campus of the University of Southern California (USC), used as a reference urban background site. Samples were chemically analyzed for elemental carbon (EC), organic carbon (OC), polycyclic aromatic hydrocarbons (PAHs) and 50 elements. The cumulative mass fraction of the measured PAHs was highest on the freeways (0.16 ± 0.01 and 0.15 ± 0.01 ng/ μ g PM, on I-110 and I-710, respectively); which on average was 3 and 3.3-fold higher than at Wilshire/Sunset and USC site, respectively. Mass fractions of Ba, Cr, Cu, Mn, Ni, Pb, Sb and Zn, tracers of vehicular abrasion, were 3.8 ± 0.8 times higher on both freeways in comparison to Wilshire/Sunset. The observed intrinsic (normalized per PM mass) DTT activity was greatest on freeways, averaging 30.13 ± 3.15 nmol/min mg PM and being roughly 1.9 and 2.1 times higher than the values obtained at Wilshire/Sunset and USC, respectively.

Furthermore, comparison of our results with previous on-road and roadside studies conducted in the last decade in Los Angeles indicated an overall reduction in the contribution of carbonaceous species and PAHs (important tracers of exhaust emissions) to PM mass, especially on I-710 freeway with the higher heavy-duty diesel vehicle fraction, indicating the effectiveness of diesel vehicle emissions control policies implemented in recent years in California. In contrast, greater contributions of certain groups of metals and trace elements that are indicators of non-tailpipe emissions compared to previous studies provide evidence on the increasing importance of non-tailpipe emissions to the oxidative potential of on-road PM_{2.5} as vehicular exhaust emissions becomes cleaner. This finding was also reflected in the increased levels of on-road DTT activity by factors of 1.4–1.5 in comparison to the DTT activity of vehicular emissions estimated in previous dynamometer studies.

Keywords : Particulate matter, Oxidative potential, Dithiothreitol (DTT) assay, Tailpipe and non-tailpipe emissions, Metals and elements, EC and OC

Comparison of source apportionment of PM_{2.5} using receptor models in the main hub port city of East Asia: Busan

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Source: Volume 148, January 2017, Pages 115-127

<https://doi.org/10.1016/j.atmosenv.2016.10.055>

The contributions of various PM_{2.5} emission sources to ambient PM_{2.5} levels during 2013 in the main hub port city (Busan, South Korea) of East Asia was quantified using several receptor modeling techniques. Three receptor models of principal component analysis/absolute principal component score (PCA/APCS), positive matrix factorization (PMF), and chemical mass balance (CMB) were used to apportion the source of PM_{2.5} obtained from the target city. The results of the receptor models indicated that the secondary formation of PM_{2.5} was the dominant (45–60%) contributor to PM_{2.5} levels in the port city of Busan. The PMF and PCA/APCS suggested that ship emission was a non-negligible contributor of PM_{2.5} (up to about 10%) in the study area, whereas it was a negligible contributor based on CMB. The magnitude of source contribution estimates to PM_{2.5} levels differed significantly among these three models due to their limitations (e.g., PM_{2.5} emission source profiles and restrictions of the models). Potential source contribution function and concentration-weighted trajectory analyses indicated that long-range transport from sources in the eastern China and Yellow Sea contributed significantly to the level of PM_{2.5} in Busan.

Keywords: Receptor modeling, PM_{2.5}, PCA/APCS, PMF, CMB, PSCF, CWT, Busan

Evaluation of fire weather forecasts using PM_{2.5} sensitivity analysis

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Source: Volume 148, January 2017, Pages 128-138

<https://doi.org/10.1016/j.atmosenv.2016.09.010>

Fire weather forecasts are used by land and wildlife managers to determine when meteorological and fuel conditions are suitable to conduct prescribed burning. In this work, we investigate the sensitivity of ambient PM_{2.5} to various fire and meteorological variables in a spatial setting that is typical for the southeastern US, where prescribed fires are the single largest source of fine particulate matter. We use the method of principle components regression to estimate sensitivity of PM_{2.5}, measured at a monitoring site in Jacksonville, NC (JVL), to fire data and observed and forecast meteorological variables. Fire data were gathered from prescribed fire activity used for ecological management at Marine Corps Base Camp Lejeune, extending 10–50 km south from the PM_{2.5} monitor. Principal components analysis (PCA) was run on 10 data sets that included acres of prescribed burning activity (PB) along with meteorological forecast data alone or in combination with observations. For each data set, observed PM_{2.5} (unitless) was regressed against PCA scores from the first seven principal components (explaining at least 80% of total variance).

PM_{2.5} showed significant sensitivity to PB: $3.6 \pm 2.2 \mu\text{g m}^{-3}$ per 1000 acres burned at the investigated distance scale of ~10–50 km. Applying this sensitivity to the available activity data revealed a prescribed burning source contribution to measured PM_{2.5} of up to 25% on a given day. PM_{2.5} showed a positive sensitivity to relative humidity and temperature, and was also sensitive to wind direction, indicating the capture of more regional aerosol processing and transport effects. As expected, PM_{2.5} had a negative sensitivity to dispersive variables but only showed a statistically significant negative sensitivity to ventilation rate, highlighting the importance of this parameter to fire managers. A positive sensitivity to forecast precipitation was found, consistent with the practice of conducting prescribed burning on days when rain can naturally extinguish fires. Perhaps most importantly for land managers, our analysis suggests that instead of relying on the forecasts from a day before, prescribed burning decisions should be based on the forecasts released the morning of the burn when possible, since these data were more stable and yielded more statistically robust results.

Keywords : Prescribed fires, Fine particulate matter, PM_{2.5}, Sensitivity, Source apportionment, Forest ecosystem management

Size-segregated particulate matter inside residences of elderly in the Metropolitan Area of São Paulo, Brazil

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Source: Volume 148, January 2017, Pages 139-151

<https://doi.org/10.1016/j.atmosenv.2016.10.004>

The elderly population spend relatively more time indoors and is more sensitive to air pollution-related health risks but there is limited information on the quality of the air they breathe inside their residences. The objectives of this work are to (i) characterise mass of size-segregated particulate matter (PM) in elderly residences in Metropolitan Area of São Paulo (MASP) in Brazil, (ii) assess the impact of the meteorological parameters on the behaviour of indoor PM concentrations, (iii) evaluate the indoor and outdoor relationship of PM mass concentration, and (iv) estimate the respiratory deposition doses (RDD). To achieve these objectives, we measured mass concentrations of size-segregated particles in 59 elderly residences in MASP. The measurements were made in the 0.25–10 μm size range in 5 size bins using a Personal Cascade Impactor Sampler. We evaluated the mass concentration of particles using a gravimetric method and compared our PM₁₀ (sum of all size bins) and PM_{2.5} (sum of all size bins, except PM_{10–2.5}) concentrations against the 24 h mean guidelines recommended by World Health Organization (WHO). Our results show the mean PM₁₀ and PM_{2.5} measured in elderly residences in MASP as 35.2 and 27.4 $\mu\text{g m}^{-3}$, respectively. PM_{2.5} and PM_{<0.25} (particles with aerodynamic diameter of less than 0.25 μm) contributed 78% and 38% of total PM₁₀, respectively, clearly suggesting a significantly high exposure to fine particles by the elderly. About 13 and 43% of the measurements exceeded the WHO's PM₁₀ and PM_{2.5} guidelines, respectively. The samples were clustered into five groups to found the behaviour of indoor PM. The cluster representing the residences with higher PM concentration in all size bins are predominantly residences near the heavy traffic areas during the non-precipitation days. About 68% of residences showed the highest fraction of PM_{<0.25}, indicating a high concentration of ultrafine particles in these residences. We calculated

indoor/outdoor (I/O) rates and found them as 1.89 and 1.06 for PM_{2.5} and PM₁₀, respectively. About 77% and 40% of the residences had higher PM_{2.5} and PM₁₀ indoors than those in outdoor environments. During seated position, the RDD rates for coarse and fine particles for male elderly were found to be about 20% and 25% higher compared with female elderly, respectively. Our findings suggest a control of indoor sources in the elderly residences to limit adverse health effects of particulate matter, especially fine particles, on elderly.

Keywords :Size-segregated particulate matter, Elderly residences, Indoor air quality, Urban area of São Paulo, Indoor/outdoor ratio, Respiratory deposition

Clustered long-range transport routes and potential sources of PM_{2.5} and their chemical characteristics around the Taiwan Strait

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Source: Volume 148, January 2017, Pages 152-166

<https://doi.org/10.1016/j.atmosenv.2016.10.010>

This study investigated the spatiotemporal variation, chemical characteristics, source apportionment, and their contribution for clustered transport routes of atmospheric fine particle (PM_{2.5}) around the Taiwan Strait. Atmospheric PM_{2.5} was simultaneously collected at three selected sampling sites around the Taiwan Strait in the years of 2013–2015. Field sampling results showed that atmospheric PM_{2.5} concentrations varied with the clustered transport routes. Backward trajectory analyses suggested that PM_{2.5} concentrations under the northerly wind conditions were generally higher than those under the southerly wind conditions. Chemical analysis results showed that the most abundant chemical composition were secondary inorganic aerosols (SO₄²⁻, NO₃⁻, and NH₄⁺), natural crustal materials (Mg, Ca, Al, K, and Fe), and anthropogenic metals (Pb, Ni, and Zn). Moreover, high OC/EC ratios of PM_{2.5} were commonly observed at the west-side site located at the downwind of major stationary sources. Furthermore, primary organic carbons (POC) were always higher than secondary organic carbons (SOC) on both sides of the Taiwan Strait. The concentrations of chemical species from the trajectory NWW (southern China) were much higher than other trajectories. Integrating the results obtained from receptor modeling and backward trajectory simulation indicated that high PM_{2.5} concentrations were transported from North China, the eastern coast of China, Korea Peninsula, and South Japan. It was mainly attributed to the combination of the relatively elevated emissions from coal burning for space heating, and long-range transport (LONG-RANGE TRANSPORT) of PM_{2.5} from upwind sources. The source apportionment of secondary aerosols were in order of east-side site > offshore site > west-side site, suggesting that aged secondary particles could be formed during the transportation process by longer range and duration toward the east-side site of Taiwan Strait and the offshore site.

Keywords :Atmospheric fine particles Taiwan Strait, Clustered transport routes, Long-range transport, Source apportionment

On-road measurements of vehicle NO₂/NO_x emission ratios in Denver, Colorado, USA

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Source: Volume 148, January 2017, Pages 182-189
<https://doi.org/10.1016/j.atmosenv.2016.10.039>

Nitrogen oxides (NO_x = NO + NO₂) emitted by on-road combustion engines are important contributors to tropospheric ozone production. The NO_x fraction emitted as nitrogen dioxide (NO₂) is usually presumed to be small but can affect ozone production and distribution, and this fraction is generally not reported in emissions inventories. We have developed an accurate method for determination of this primary NO₂ emission and demonstrated it during measurement of on-road vehicle emission plumes from a mobile laboratory during July and August 2014 in the region between Denver and Greeley in Colorado. During a total of approximately 90 h of sampling from an instrumented mobile laboratory, we identified 1867 vehicle emission plumes, which were extracted using an algorithm that looks for rapid and large increases in measured NO_x. We find a distribution of NO₂/NO_x emissions similar to a log-normal profile, with an average emission ratio of 0.053 ± 0.002 per sampled NO_x plume. The average is not weighted by the total NO_x emissions from sampled vehicles, which is not measured here, and so may not represent the NO₂/NO_x ratio of the total NO_x emission if this ratio is a function of NO_x itself. Although our current data set does not distinguish between different engine types (e.g., gasoline, light duty diesel and heavy duty diesel), the ratio is on the low end of recent reports of vehicle fleet NO₂ to NO_x emission ratios in Europe.

Keywords: NO_x emissions, Direct NO₂ emissions

Emission characteristics of polycyclic aromatic hydrocarbons and nitro-polycyclic aromatic hydrocarbons from diesel trucks based on on-road measurements

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Source: Volume 148, January 2017, Pages 190-196
<https://doi.org/10.1016/j.atmosenv.2016.10.040>

Polycyclic aromatic hydrocarbon (PAH) and nitro-polycyclic aromatic hydrocarbon (NPAH) emissions from 18 diesel trucks of different sizes and with different emission standards were tested in Beijing using a portable emission measurement system (PEMS). Both the gaseous- and particulate-phase PAHs and NPAHs were quantified by high-performance liquid chromatography (HPLC) in the laboratory. The emission factors (EFs) of the total PAHs from light-duty diesel trucks (LDDTs), medium-duty diesel trucks (MDDTs) and heavy-duty diesel trucks (HDDTs) were 82229.11 ± 41906.06 , 52867.43 ± 18946.47 and 93837.35 ± 32193.14 µg/km, respectively, much higher than the respective values of total NPAHs from their counterpart vehicles. The gaseous

phase had an important contribution to the total PAHs and NPAHs, with a share rate of approximately 69% and 97% on average, respectively. The driving cycle had important impacts on the emissions of PAHs and NPAHs, especially for LDDTs and HDDTs. Higher emissions of PAHs and NPAHs were detected on non-highway roads compared to that on highways for these two types of vehicles. Compared to the results of different studies, the difference in the EFs of PAHs and NPAHs can reach several orders of magnitudes, which would introduce errors in the development of an emission inventory of PAHs and NPAHs.

Keywords : PAHs, NPAHs, Diesel vehicle, Emission factors, PEMS

Analysis of major air pollutants and submicron particles in New York City and Long Island

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Source: *Volume 148, January 2017, Pages 203-214*

<https://doi.org/10.1016/j.atmosenv.2016.10.043>

A year-long sampling campaign of major air pollutants and submicron particle number size distributions was conducted at two sites taken as representative of city-wide air quality in New York City and Long Island, respectively. A number of species were quantified with hourly time resolution, including particle number concentrations in 6 size ranges (20–30 nm, 30–50 nm, 50–70 nm, 70–100 nm, 100–200 nm, and >200 nm), nitrogen oxides, sulfur dioxide, ozone, carbon monoxide, methane, non-methane hydrocarbons, PM_{2.5} mass concentration and some PM major components (sulfate, organic and elemental carbon). Hourly concentrations of primary and secondary organic carbon were estimated using the EC tracer method. Data were matched with weather parameters and air parcel back-trajectories. A series of tools were thus applied to: (i) study the seasonal, weekly, diurnal cycles of pollutants; (ii) investigate the relationships amongst pollutants through correlation and lagged correlation analyses; (iii) depict the role of atmospheric photochemical processes; (iv) examine the location of the potential sources by mean of conditional bivariate probability function analysis and (v) investigate the role of regional transport of air masses to the concentrations of analyzed species. Results indicate that concentrations of NO_x, SO₂, CO, non-methane hydrocarbons, primary OC and EC are predominantly determined by local sources, but are also affected by regional transports of polluted air masses. On the contrary, the transport of continental polluted air masses has a main effect in raising the concentrations of secondary PM_{2.5} (sulfate and secondary organic carbon). By providing direct information on the concentrations and trends of key pollutants and submicron particle number concentrations, this study finally enables some general considerations about air quality status and atmospheric processes over the New York City metropolitan area.

Keywords : Air quality, New York City, Nitrogen oxides, Organic carbon, Sulfate, Particle size

Source apportionment of PM_{2.5} chemically speciated mass and particle number concentrations in New York City

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Source: Volume 148, January 2017, Pages 215-229

<https://doi.org/10.1016/j.atmosenv.2016.10.044>

The major sources of fine particulate matter (PM_{2.5}) in New York City (NYC) were apportioned by applying positive matrix factorization (PMF) to two different sets of particle characteristics: mass concentrations using chemical speciation data and particle number concentrations (PNC) using number size distribution, continuously monitored gases, and PM_{2.5} data. Post-processing was applied to the PMF results to: (i) match with meteorological data, (ii) use wind data to detect the likely locations of the local sources, and (iii) use concentration weighted trajectory models to assess the strength of potential regional/transboundary sources. Nine sources of PM_{2.5} mass were apportioned and identified as: secondary ammonium sulfate, secondary ammonium nitrate, road traffic exhaust, crustal dust, fresh sea-salt, aged sea-salt, biomass burning, residual oil/domestic heating and zinc. The sources of PNC were investigated using hourly average number concentrations in six size bins, gaseous air pollutants, mass concentrations of PM_{2.5}, particulate sulfate, OC, and EC. These data were divided into 3 periods indicative of different seasonal conditions. Five sources were resolved for each period: secondary particles, road traffic, NYC background pollution (traffic and oil heating largely in Manhattan), nucleation and O₃-rich aerosol. Although traffic does not account for large amounts of PM_{2.5} mass, it was the main source of particles advected from heavily trafficked zones. The use of residual oil had limited impacts on PM_{2.5} mass but dominates PNC in cold periods

Keywords : Source apportionment, Positive matrix factorization, PM_{2.5} mass concentration, Particle number concentration, New York City, Residual oil

Oxidative potential of subway PM_{2.5}

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Source: Volume 148, January 2017, Pages 230-238

<https://doi.org/10.1016/j.atmosenv.2016.10.045>

Air quality in subway systems is of interest not only because particulate matter (PM) concentrations can be high, but also because of the peculiarly metalliferous chemical character of the particles, most of which differ radically from those of outdoor ambient air. We report on the oxidative potential (OP) of PM_{2.5} samples collected in the Barcelona subway system in different types of stations. The PM chemical composition of these samples showed typically high concentrations of Fe, Total Carbon, Ba, Cu, Mn, Zn and Cr sourced from rail tracks, wheels, catenaries, brake pads and pantographs. Two toxicological indicators of oxidative activity, ascorbic acid (AA) oxidation (expressed as OPAA μg^{-1} or OPAA m^{-3}) and glutathione (GSH) oxidation

(expressed as OPGSH $\mu\text{g}-1$ or OPGSH $\text{m}-3$), showed low OP for all samples (compared with outdoor air) but considerable variation between stations (0.9–2.4 OPAA $\mu\text{g}-1$; 0.4–1.9 OPGSH $\mu\text{g}-1$). Results indicate that subway PM toxicity is not related to variations in PM_{2.5} concentrations produced by ventilation changes, tunnel works, or station design, but may be affected more by the presence of metallic trace elements such as Cu and Sb sourced from brakes and pantographs. The OP assays employed do not reveal toxic effects from the highly ferruginous component present in subway dust.

Keywords : Particulate matter , Oxidative potential, Toxicity, PM chemistry, Subway air quality, Airborne metals

Air quality early-warning system for cities in China

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Source: Volume 148, January 2017, Pages 239-257

<https://doi.org/10.1016/j.atmosenv.2016.10.046>

Air pollution has become a serious issue in many developing countries, especially in China, and could generate adverse effects on human beings. Air quality early-warning systems play an increasingly significant role in regulatory plans that reduce and control emissions of air pollutants and inform the public in advance when harmful air pollution is foreseen. However, building a robust early-warning system that will improve the ability of early-warning is not only a challenge but also a critical issue for the entire society. Relevant research is still poor in China and cannot always satisfy the growing requirements of regulatory planning, despite the issue's significance. Therefore, in this paper, a hybrid air quality early-warning system was successfully developed, composed of forecasting and evaluation. First, a hybrid forecasting model was proposed as an important part of this system based on the theory of “decomposition and ensemble” and combined with the advanced data processing technique, support vector machine, the latest bio-inspired optimization algorithm and the leave-one-out strategy for deciding weights. Afterwards, to intensify the research, fuzzy evaluation was performed, which also plays an indispensable role in the early-warning system. The forecasting model and fuzzy evaluation approaches are complementary. Case studies using daily air pollution concentrations of six air pollutants from three cities in China (*i.e.*, Taiyuan, Harbin and Chongqing) are used as examples to evaluate the efficiency and effectiveness of the developed air quality early-warning system. Experimental results demonstrate that both the accuracy and the effectiveness of the developed system are greatly superior for air quality early warning. Furthermore, the application of forecasting and evaluation enables the informative and effective quantification of future air quality, offering a significant advantage, and can be employed to develop rapid air quality early-warning systems.

Keywords : Air quality early-warning systems, Hybrid forecasting model, Data preprocessing, Bio-inspired optimization algorithm, Fuzzy evaluation

Regionalized PM2.5 Community Multiscale Air Quality model performance evaluation across a continuous spatiotemporal domain

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Source: Volume 148, January 2017, Pages 258-265

<https://doi.org/10.1016/j.atmosenv.2016.10.048>

The regulatory Community Multiscale Air Quality (CMAQ) model is a means to understanding the sources, concentrations and regulatory attainment of air pollutants within a model's domain. Substantial resources are allocated to the evaluation of model performance. The Regionalized Air quality Model Performance (RAMP) method introduced here explores novel ways of visualizing and evaluating CMAQ model performance and errors for daily Particulate Matter $\leq 2.5 \mu\text{m}$ (PM2.5) concentrations across the continental United States. The RAMP method performs a non-homogenous, non-linear, non-homoscedastic model performance evaluation at each CMAQ grid. This work demonstrates that CMAQ model performance, for a well-documented 2001 regulatory episode, is non-homogeneous across space/time. The RAMP correction of systematic errors outperforms other model evaluation methods as demonstrated by a 22.1% reduction in Mean Square Error compared to a constant domain wide correction. The RAMP method is able to accurately reproduce simulated performance with a correlation of $r = 76.1\%$. Most of the error coming from CMAQ is random error with only a minority of error being systematic. Areas of high systematic error are collocated with areas of high random error, implying both error types originate from similar sources. Therefore, addressing underlying causes of systematic error will have the added benefit of also addressing underlying causes of random error.

Keywords : Model performance evaluation, Regulatory modeling, PM2.5, CMAQ, Modeled data

Estimating accidental pollutant releases in the built environment from turbulent concentration signals

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Source: Volume 148, January 2017, Pages 266-281

<https://doi.org/10.1016/j.atmosenv.2016.10.050>

We present an inverse atmospheric model to estimate the mass flow rate of an impulsive source of pollutant, whose position is known, from concentration signals registered at receptors placed downwind of the source. The originality of this study is twofold. Firstly, the inversion is performed using high-frequency fluctuating, i.e. turbulent, concentration signals. Secondly, the inverse algorithm is applied to a dispersion process within a dense urban canopy, at the district scale, and a street network model, SIRANERISK, is adopted. The model, which is tested against wind tunnel experiments, simulates the dispersion of short-duration releases of pollutant in different typologies of idealised urban geometries. Results allow us to discuss the reliability of the inverse model as an operational tool for crisis management and the risk assessments related to the accidental release of toxic and flammable substances.

Keywords : Inverse model, Accidental release, Turbulent dispersion, Urban dispersion model, Concentration fluctuations

Role of surface wind and vegetation cover in multi-decadal variations of dust emission in the Sahara and Sahel

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Source: Volume 148, January 2017, Pages 282-296
<https://doi.org/10.1016/j.atmosenv.2016.10.051>

North Africa, the world's largest dust source, is non-uniform, consisting of a permanently arid region (Sahara), a semi-arid region (Sahel), and a relatively moist vegetated region (Savanna), each with very different rainfall patterns and surface conditions. This study aims to better understand the controlling factors that determine the variation of dust emission in North Africa over a 27-year period from 1982 to 2008, using observational data and model simulations. The results show that the model-derived Saharan dust emission is only correlated with the 10-m winds (W10m) obtained from reanalysis data, but the model-derived Sahel dust emission is correlated with both W10m and the Normalized Difference Vegetation Index (NDVI) that is obtained from satellite. While the Saharan dust accounts for 82% of the continental North Africa dust emission (1340–1570 Tg year⁻¹) in the 27-year average, the Sahel accounts for 17% with a larger seasonal and inter-annual variation (230–380 Tg year⁻¹), contributing about a quarter of the transatlantic dust transported to the northern part of South America. The decreasing dust emission trend over the 27-year period is highly correlated with W10m over the Sahara ($R = 0.92$). Over the Sahel, the dust emission is correlated with W10m ($R = 0.69$) but is also anti-correlated with the trend of NDVI ($R = -0.65$). W10m is decreasing over both the Sahara and the Sahel between 1982 and 2008, and the trends are correlated ($R = 0.53$), suggesting that Saharan/Sahelian surface winds are a coupled system, driving the inter-annual variation of dust emission.

Keywords : Dust emission, Surface wind, NDVI, GOCART model, North Africa, Sahel

Effects of synoptic weather on ground-level PM_{2.5} concentrations in the United States

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Source: Volume 148, January 2017, Pages 297-305
<https://doi.org/10.1016/j.atmosenv.2016.10.052>

It is known that individual meteorological factors affect the concentrations of fine particulate matter with aerodynamic diameters $\leq 2.5 \mu\text{m}$ (PM_{2.5}), yet the specific meteorological effects found

in previous studies are largely inconsistent and even conflicting. This study investigates influences of daily and short term changes in synoptic weather on ground-level PM_{2.5} concentrations in a large geographical area (75 cities across the contiguous United States (U.S.)) by using ten-year (2001–2010) spatial synoptic classification (SSC) data. We find that in the spring, summer, and fall the presence of the tropical weather types (i.e., dry-tropical (DT) and moist-tropical (MT)) is likely to associate with significantly higher levels of PM_{2.5} as compared to an all-weather-type-day average, and the presence of the polar weather types (i.e., dry-polar (DP) and moist-polar (MP)) is associated with significantly lower PM_{2.5} concentrations. The short-term (day to day) changes in synoptic weather types in a region are also likely to lead to significant variance in PM_{2.5} concentrations. For example, the largest increase in PM_{2.5} concentration occurs with the synoptic weather type changing from DP-to-MT. Conversely, a MT-to-DP weather type change results in the largest decrease in PM_{2.5} concentrations. Compared to air temperature, the effects of atmospheric moisture on PM_{2.5} concentration tend to be subtle, demonstrating that in conjunction with moderate temperature, neither the dry nor the moist air (except moist-moderate (MM) in summer) are associated with significantly high or low PM_{2.5} concentrations. Finally, we find that the effects of the synoptic weather type on PM_{2.5} concentrations may vary for different seasons and geographical areas. These findings suggest that interactions between atmospheric factors and seasonal and/or geographical factors have considerable impacts on the PM_{2.5} concentrations, and therefore should be considered in addition to the SSC when conducting environment health assessments.

Keywords : Spatial synoptic classification, PM_{2.5} concentrations, Season, Synoptic weather, Air quality

Transport of East Asian dust storms to the marginal seas of China and the southern North Pacific in spring 2010

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Source: Volume 148, January 2017, Pages 316-328

<https://doi.org/10.1016/j.atmosenv.2016.10.054>

The transport of a super-severe dust storm that occurred in East Asia from 19 to 22 March 2010 has been well documented by both model simulations and surface observations. We investigated the transport of this severe dust storm and several other spring 2010 dust storms using model simulations, backward trajectories, and measurements from the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO) satellite. The model simulations indicated that the emission of dust in spring 2010 was about 35% higher than that in spring 2006 and was twice that of the 44-year average from 1960 to 2003. The dust emissions over two major source regions (western China and the Gobi Desert) in spring 2010 accounted for 86% of the total amount of dust. The simulated depositional flux over the two major source regions was c.2.7–9.0 times that over two marginal seas (the Yellow Sea and the East China Sea) and more than two magnitudes higher than that over the North Pacific Subtropical Gyre. The mean extinction coefficient observed by the CALIPSO satellite over the two source regions was c.1.3–3.7 times that over the two marginal seas and one magnitude higher than that over the North Pacific. This was consistent with the result from our model, suggesting that this model is able to capture the major features of dust storms. The vertical profiles recorded by the CALIPSO satellite indicated that the dust aerosol mainly floated

from the ground to 13 km above ground level over the source regions and above the seas. A multiple dust layer appeared over the seas and the backward trajectories suggested that the dust aerosol in different layers may have been sourced from different regions.

Keywords : Dust storms, Transport, Dust emissions, Dust deposition, Vertical profiles, Extinction coefficients

Determination of n-alkanes, PAHs and nitro-PAHs in PM_{2.5} and PM₁ sampled in the surroundings of a municipal waste incinerator

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Source: Volume 149, January 2017, Pages 12-23

<https://doi.org/10.1016/j.atmosenv.2016.11.016>

This work investigates n-alkanes in the range C₁₄-C₃₂, polycyclic aromatic hydrocarbons (PAHs) and nitrated PAHs (nitro-PAHs) composition of PM_{2.5} and PM₁ collected in the surroundings of a municipal waste incinerator close to Bologna (Northern Italy). The particulate matter was sampled in eight stations: one of these was located inside the urban area of Bologna, while the others were placed in a domain of 8 × 9 km² around the incinerator plant. Two monitoring campaigns were carried out, in summer 2008 and in winter 2009. In each campaign, two stations were placed in zones of maximum impact of plant emissions and other two ones as their controls. The study of n-alkanes in atmospheric particulate was performed to understand the contribution of anthropogenic and biogenic sources, PAHs and nitro-PAHs were selected to investigate PM composition near an incinerator plant. Higher concentrations of PAHs and, to a less extent, of n-alkanes were measured in the cold season. This can be due to several reasons, namely, an increased combustion of fossil fuel during wintertime, stagnant atmospheric conditions and lower temperatures that not only favour gas-to-particles partitioning but also reduce atmospheric reactivity. The impact of combustion in the cold season was also confirmed by the higher percentage content of PAHs in PM₁ than in PM_{2.5}. On the contrary, higher amount of nitro-derivatives were measured in summer than in winter, suggesting that the contribution of secondary aerosol is not negligible in the hot season. The most abundant n-alkanes were the long-chain homologues (>C₂₇) deriving from anthropogenic sources as indicated by the Carbon Preference Indexes. In both seasons, the sites chosen as controls were characterized by higher concentrations of PAHs than the respective maxima suggesting that extra-incinerator sources, especially vehicles emissions, are the main responsible for PAHs in PM_{2.5}. On the contrary, in winter, PM₁ was enriched in PAHs in the site close to the incinerator with respect to its control.

Keywords : PM_{2.5}, PM₁, n-alkanes, PAHs, Nitro-PAHs, Incinerator plant

Effect of monitoring network design on land use regression models for estimating residential NO₂ concentration

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Source: Volume 149, January 2017, Pages 24-33

<https://doi.org/10.1016/j.atmosenv.2016.11.014>

Land-use regression (LUR) models are increasingly used to estimate exposure to air pollution in urban areas. An appropriate monitoring network is an important component in the development of a robust LUR model. In this study concentrations of NO₂ were simulated by a dispersion model at 'virtual' monitoring sites in 54 network designs of varying numbers and types of site, using a 25 km² area in Edinburgh, UK, as an example location. Separate LUR models were developed for each network. The LUR models were then used to estimate NO₂ concentration at all residential addresses, which were evaluated against the dispersion-modelled concentration at these addresses. The improvement in predictive capability of the LUR models was insignificant above ~30 monitoring sites, although more sites tended to yield more precise LUR models. Monitoring networks containing sites located within highly populated areas better estimated NO₂ concentrations across all residential locations. LUR models constructed from networks containing more roadside sites better characterised the high end of residential NO₂ concentrations but had increased errors when considering the whole range of concentrations. No particular composition of monitoring network resulted in good estimation simultaneously across all residential NO₂ concentration and of the highest NO₂ levels. This evaluation with dispersion modelling has shown that previous LUR model validation methods may have been optimistic in their assessment of the model's predictive performance at residential locations.

Keywords : Land-use regression model, Dispersion model, Exposure assessment

Particulate matter produced during commercial sugarcane harvesting and processing: A respiratory health hazard?

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Source: Volume 149, January 2017, Pages 34-46

<https://doi.org/10.1016/j.atmosenv.2016.11.012>

Emissions from sugarcane burning are known to impact on the respiratory health of sugar estate workers and local populations. Despite this, there have been few studies on occupational and ambient exposures and risks from airborne particulate matter (PM) associated with field burning and ash re-suspension. From workplace monitoring on sugarcane estates in two different South American countries in 2010 and 2011, median concentrations of airborne PM₁₀ (particulate matter nominally <10 µm in diameter) were found to be statistically much higher during pre-harvest sugarcane burning (1807 µg m⁻³) than during either sugarcane cutting after burning (~123 µg m⁻³) or in the sugarcane processing factory (~175 µg m⁻³). Median PM₁₀ measurements in ambient scenarios, for example in the sugarcane fields before the burning

or during 24 h measurements in neighboring villages (bordering the sugarcane plantation), were much lower, between 18 and 37 $\mu\text{g m}^{-3}$. From the analysis of size-selective samples of airborne PM₁₀, collected during sugarcane field burning, cutting and ambient periods, almost all (~96 wt %) fell within the 'respirable' fraction (<4 μm aerodynamic diameter), with a mass median aerodynamic diameter (MMAD) of 1.1 μm . Residual ash from field and bagasse burning, characterised using Scanning Electron Microscopy (SEM) with X-ray elemental analysis, was found to contain carbonaceous and silicate-dominated particles in the PM_{0.5} and PM_{0.5-2.5} size ranges and fibres from <10 to over 50 μm in length. Only a small proportion of the field burning ash (average 0.6 vol %) and bagasse ash (average 1.3 vol %) was in the respirable fraction. However, from grinding experiments, which simulate disaggregation as a result of disturbance during harvest or bagasse ash removal, the ash was fragile and easily broken down into thoracic particulate (<10 μm aerodynamic diameter) and, in some instances, created respirable-sized PM. From exposure calculations, the 8 h time weighted average (TWA) concentrations of PM₁₀, during the different measurement scenarios, were found to be below occupational exposure limits (OELs; 5000 $\mu\text{g m}^{-3}$ for respirable PM). Ambient PM₁₀ exposure of residents surrounding the sugarcane plantations was found to be below the WHO air quality guideline (50 $\mu\text{g m}^{-3}$ as a 24 h mean). The relative risk calculated for 'all cause' mortality from exposure of nearby residents to PM₁₀ generated by sugarcane burning was found to be 3%. The concentrations of PM₁₀ produced during the processing of sugarcane were high (up to 21.5 mg m^{-3}), which is concerning given that re-suspended particles of ash in the fields and processing plant have been previously shown to contain potentially toxic cristobalite. PM produced during sugarcane burning, and during extended periods of local exposure to the smoke and re-suspended ash, therefore, should be considered as both a potential acute and chronic respiratory health hazard. This issue will become increasingly important with the forecasted rise in sugarcane production for biofuels.

Keywords : Particulate matter, Sugarcane, Respiratory hazard, PM10, Occupational, Ambient

Multifractal analysis of interactive patterns between meteorological factors and pollutants in urban and rural areas

Hong-diHe

Source: Volume 149, January 2017, Pages 47-54

<https://doi.org/10.1016/j.atmosenv.2016.11.004>

This paper seeks to enhance understanding of the cross-correlation patterns between meteorological factors and pollutants. The observed databases of daily meteorological elements (temperature, humidity and wind speed), as well as pollutants (CO, NO_x, PM₁₀ and SO₂) levels during 2005–2014, is collected. Based on the database, the cross-correlation test is carried out firstly and the results indicate that cross-correlation behaviors exist statistically between them. Then the detrended cross-correlation analysis is performed for further analysis. With a detailed comparison, long-term cross-correlation behaviors are found to be more obvious in rural area. Beside, the influences of meteorological factors on multifractal property for pollutants are investigated. In contrast to humidity and wind speed, the long-term cross-correlation behaviors between temperature with pollutants are found to be more evident in both urban and rural areas. Furthermore, the difference of multifractal property for varied pollutants is explored. The strengths

of multifractal spectra between meteorological factors with PM10 are strongest while the corresponding values between meteorological factors with SO₂ are weakest. These findings successfully illustrate that the multifractal analysis is a useful tool for uncovering the interactive pattern in environmental issues.

Keywords : Cross-correlation behavior, Multifractality, Meteorological factors, Pollutants

Use of tethered sonde and aircraft profiles to study the impact of mesoscale and microscale meteorology on air quality

Source: Volume 149, January 2017, Pages 55-69

<https://doi.org/10.1016/j.atmosenv.2016.10.025>

Highly-resolved vertical profiles of ozone and reactive nitrogen in the lower troposphere were obtained using Millersville University's tethered balloon system and NASA's P-3B aircraft during the July 2011 Baltimore, MD/Washington DC and the September 2013 Houston, TX deployments of the NASA DISCOVER-AQ air quality field mission. The tethered balloon and surface measurement sites were located at Edgewood, MD and Smith Point, TX. The balloon profiles are used to connect aircraft data from the lowest portion of NASA's P-3B spirals (300 m AGL) to the surface thus creating complete profiles from the surface to 3–5 km AGL. The highest concentrations of surface ozone at these coastal sites resulted from mean flow transport of polluted air over an adjacent body of water followed by advection back over land several hours later, due to a bay or gulf breeze. Several meteorological processes including horizontal advection, vertical mixing, thermally direct circulation (i.e., bay, gulf, and, sea breezes) combined with chemical processes like photochemical production and deposition played a role in the local ozone maxima. Several small-scale, but highly polluted layers from the Chesapeake Bay advected landward over Edgewood, MD. The Houston Metro area was subject to large-scale recirculation of emissions from petrochemical sources by the Gulf of Mexico and Galveston Bay breezes.

Keywords : Ozone, Bay breeze, Gulf breeze, DISCOVER-AQ campaign, Pollution

A comparison study of carbonaceous aerosols in a typical North China Plain urban atmosphere: Seasonal variability, sources and implications to haze formation

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Source: Volume 149, January 2017, Pages 95-103

<https://doi.org/10.1016/j.atmosenv.2016.11.009>

For a better understanding of the formation of regional haze in the North China Plain (NCP) region, aerosol concentrations and compositions, trace gases and meteorological factors were

comprehensively measured for 1 year in Jinan, a typical city in the NCP. Through comparison studies between hazy & non-hazy days, and five years of changes, this paper presents the characterisation of seasonal variations and sources of carbonaceous aerosol, and their implications to haze formation in the NCP region. The contributions of carbonaceous aerosols ($\text{OC} \times 1.8 + \text{EC}$) to $\text{PM}_{2.5}$ ranged from 17.2 to 30.4% on hazy days and 20.5–29.7% on non-hazy days. The annual trends of organic carbon (OC) and elemental carbon showed obvious seasonality, with the lowest and highest concentrations in spring and winter, respectively. The ratio of secondary OC to OC on hazy days during all four seasons ranged from 43.8 to 63.4%, which was higher than that on non-hazy days. Different types of haze events are identified (e.g., combustion of coal and biomass), and the sources and formation mechanisms of carbonaceous aerosols are discussed. In winter, coal combustion and secondary formation were the main sources of carbonaceous aerosols. Biomass combustion contributed to the rapid enhancement of carbonaceous aerosols during summer and autumn. We found that the OC and secondary OC concentrations on haze-fog days during the winter of 2012–2013 in Jinan were much higher than those during the winter of 2007–2008, which was consistent with the ambient relative humidity. These results suggest that aqueous reaction played a significant role in the formation of secondary organic aerosols and contributed to regional haze formation in the NCP region.

Keywords : Haze, Carbonaceous aerosols, Secondary organic aerosol, Coal burning, Biomass burning

Assessing the effect of long-range pollutant transportation on air quality in Seoul using the conditional potential source contribution function method

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Source: Volume 150, February 2017, Pages 33-44

<https://doi.org/10.1016/j.atmosenv.2016.11.017>

It is important to estimate the effects of the long-range transport of atmospheric pollutants for efficient and effective strategies to control air quality. In this study, the contributions of trans-boundary transport to the mean concentrations of SO_2 , NO_2 , CO, and PM_{10} in Seoul, Korea from 2001 to 2014 were estimated based on the conditional potential source contribution function (CPSCF) method. Eastern China was found to be the major source of trans-boundary pollution in Seoul, but moderate sources were also located in northeastern China. The contribution of long-range transport from Japan was negligible. The spatial distributions of the potential source contribution function (PSCF) values of each pollutant showed reasonable consistency with their emission inventory and satellite products. The PSCF values of SO_2 and PM_{10} from eastern China were higher than those of NO_2 and CO. The mean concentrations of SO_2 , NO_2 , CO, and PM_{10} in Seoul for the period from 2001 to 2014 were 5.34, 37.0, and 619.1 ppb, and 57.4 $\mu\text{g}/\text{m}^3$, respectively. The contributions of long-range transport to the mean concentrations of SO_2 , NO_2 , CO, and PM_{10} in Seoul were 0.74, 3.4, and 39.0 ppb, and 12.1 $\mu\text{g}/\text{m}^3$, respectively, which are 14%, 9%, 6%, and 21% of the mean concentrations, respectively. The annual mean concentrations of SO_2 and NO_2 followed statistically significant increasing linear trends (0.5 and 1.6 ppb per decade, respectively), whereas the trends in the annual mean concentrations of CO and PM_{10} were statistically insignificant. The trends in the ratio of the increased concentrations associated with

long-range transport to the annual mean concentrations of the pollutants were statistically insignificant. However, the results indicate that the trans-boundary transport of SO₂, NO₂, CO, and PM₁₀ from eastern China consistently affected air quality in Seoul over the study period (2001–2014). Regionally, the effects of the long-range transport of pollutants from Beijing and Harbin-Changchun on air quality in Seoul have become more significant over this period.

Keywords : Long-range transport, PM₁₀, SO₂, CO, NO₂, Potential source contribution function

PM_{2.5} and aerosol black carbon in Suva, Fiji

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Source: Volume 150, February 2017, Pages 55-66

<https://doi.org/10.1016/j.atmosenv.2016.11.041>

Concentrations of particulate air pollution in Suva, Fiji, have been largely unknown and consequently, current strategies to reduce health risk from air pollution in Suva are not targeted effectively. This lack of air quality data is common across the Pacific Island Countries. A monitoring study, during 2014 and 2015, has characterised the fine particulate air quality in Suva, representing the most detailed study to date of fine aerosol air pollutants for the Pacific Islands; with sampling at City, Residential (Kinoya) and Background (Suva Point) sites. Meteorology for Suva, as it relates to pollutant dispersion for this period of time, has also been analysed. The study design enables the contribution of maritime air and the anthropogenic emissions to be carefully distinguished from each other and separately characterised. Back trajectory calculations show that a packet of air sampled at the Suva City site has typically travelled 724 km in the 24-h prior to sampling, mainly over open ocean waters; inferring that pollutants would also be rapidly transported away from Suva. For fine particulates, Suva City reported a mid-week PM_{2.5} of $8.6 \pm 0.4 \mu\text{g}/\text{m}^3$, averaged over 13-months of gravimetric sampling. Continuous monitoring (Osiris laser photometer) suggests that some areas of Suva may experience levels exceeding the WHO PM_{2.5} guideline of $10 \mu\text{g}/\text{m}^3$, however, compared to other countries, Fiji's PM_{2.5} is low. Peak aerosol particulate levels, at all sites, were experienced at night-time, when atmospheric conditions were least favourable to dispersion of air pollutants. Suva's average ambient concentrations of black carbon in PM_{2.5}, $2.2 \pm 0.1 \mu\text{g}/\text{m}^3$, are, however, similar to those measured in much larger cities. With any given parcel of air spending only seven minutes, on average, over the land area of Suva Peninsula, these black carbon concentrations are indicative that significant combustion emissions occur within Suva. Many other communities in the Pacific Islands, as well as in Africa, Asia and South America share similar climate and similar burning practices and as such are likely to experience similar aerosol black carbon loadings. These black carbon levels indicate the need for combustion emissions, particularly those from open burning and diesel usage, to be addressed in air policy.

Keywords : PM_{2.5}, Black carbon, Suva

Effects of future temperature change on PM_{2.5} infiltration in the Greater Boston area

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Source: Volume 150, February 2017, Pages 98-105

<https://doi.org/10.1016/j.atmosenv.2016.11.027>

Global temperature and the frequency of extreme weather events are projected to increase and affect indoor exposure to outdoor particulate matter (PM); however, no studies have quantitatively examined the effect of climate change on particle infiltration and indoor PM exposure.

Objective

To quantify the relationship between future changes in ambient temperature and fine particle (PM_{2.5}) infiltration in the Greater Boston area.

Methods

We assembled a large database of outdoor and indoor PM_{2.5} data from 340 homes, and used the indoor-outdoor sulfur ratio (*Sr*) as a surrogate for PM_{2.5} infiltration. We employed linear mixed-effects models to examine the relationship between *Sr* and ambient temperature for all homes in the database and a subgroup of naturally ventilated homes. We used projected temperature data from 1981 to 2000 and 2046–2065 to predict future changes in *Sr*.

Results

The summer-winter difference in *Sr* was calculated to be 30% and 54% for all homes and in the naturally ventilated subgroup, respectively. The largest future difference in *Sr* (21%) was linked to differences in prevalence of air conditioning. Furthermore, *Sr* was predicted to increase by 7% for naturally ventilated homes and 2% for all homes in summer, corresponding to an average increase of 2–3 °C in future temperature.

Conclusions

We found that increases in future temperature due to climate change will be associated with increased PM_{2.5} infiltration, particularly in summer. The predicted temperature-related changes in *Sr* can be used to characterize future health risk due to elevated indoor PM_{2.5} exposure through increased particle infiltration.

Keywords: Climate change, Indoor-outdoor sulfur ratio, Particle infiltration, Temperature

Short term effects of criteria air pollutants on daily mortality in Delhi, India

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Source: Volume 150, February 2017, Pages 210-219

<https://doi.org/10.1016/j.atmosenv.2016.11.044>

Various epidemiological studies conducted in different parts of the world have conclusively established that the adverse health effects are associated with common urban air pollutants. Although several recent studies revealed the poor air quality status in Delhi, but limited evidence of

the impact of criterion air pollutants on human health remains a big limitation for relevant policy changes. So we conducted a time series to estimate the short term effects of ambient air pollution on all-natural-cause mortality in Delhi for the period 2008 to 2010. The study examined the impact of criteria air pollutants [particulate matter less than 10 μm in diameter (PM₁₀), sulphur di-oxide (SO₂), Nitrogen di-oxide (NO₂), Carbon monoxide (CO) and Ozone (O₃)] on daily all-cause-mortality rate. A semi-parametric regression model was developed to estimate the short term effects of air pollutants on daily all-natural-cause-mortality adjusting nonlinear confounding of time, temperature and relative humidity. A significant association of all-natural-cause mortality in association with short-term exposure to particulate as well as the gaseous pollutants were observed. The study estimated 0.14% (95% CI 0.02%–0.26%) increase in all-cause-mortality for every 10 $\mu\text{g}/\text{m}^3$ increase in PM₁₀ concentration. Among the gaseous pollutants, NO₂ has been found to show most significant positive association of 1.00% (95% CI 0.07%–1.93%) increase in all-cause-mortality with every 10 $\mu\text{g}/\text{m}^3$ increase in daily NO₂ concentration. The effect of O₃ and CO has been observed to be significant after controlling the effects of NO₂. Analysis by different age groups reveals that particulate matter has maximum effect estimate in the age group ≥ 65 years (RR 1.002, 95% CI 1.000 to 1.004) whereas gaseous pollutants have been found to exhibit maximum effect estimate (RR 1.016, 95% CI 1.002 to 1.030) in the age group 5–44 years. The results of the present effect estimates appeared consistent with previous findings and can enhance the strength of the previous evidences to understand health burden associated with local air quality.

Keywords : Criteria pollutants, Mortality, Time series, Relative risks

Impact of anomalous forest fire on aerosol radiative forcing and snow cover over Himalayan region

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Source: Volume 150, February 2017, Pages 264-275

<https://doi.org/10.1016/j.atmosenv.2016.11.061>

Forest fires are very common in tropical region during February–May months and are known to have significant impact on ecosystem dynamics. Moreover, aerosols emitted from these burning activities significantly modulate the Earth's radiation budget. In present study, we investigated the anomalous forest fire events and their impact on atmospheric radiation budget and glaciated snow cover over the Himalayan region. We used multiple dataset derived from satellites [Moderate Resolution Spectroradiometer (MODIS) and Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO)] and reanalysis models [Global Fire Assimilation System (GFAS), Second Modern-Era Retrospective analysis for Research and Application (MERRA-2) and ERA-interim] to evaluate the effect of biomass burning aerosols on radiation budget. April 2016 is associated with anomalous fire activities over lower Himalayan region in the last fourteen years (2003–2016). The model estimated organic carbon (OC) and black carbon (BC) emission reaches up to $\sim 3 \times 10^4$ and $\sim 2 \times 10^3 \mu\text{g}/\text{m}^2/\text{day}$, respectively during the biomass burning period of April 2016. The meteorological data analysis accompanied with CALIOP aerosol vertical profile shows that these carbonaceous aerosols could reach up to $\sim 5\text{--}7$ km altitude and could be transported towards glaciated region of upper Himalayas. The large amount of BC/OC from biomass burning significantly modulates the atmospheric radiation budget. The estimated columnar heating rate

shows that these carbonaceous aerosols could heat up the atmosphere by $\sim 0.04\text{--}0.06$ K/day in April-2016 with respect to non-burning period (2015). The glaciated snow cover fractions are found to be decreasing by $\sim 5\text{--}20\%$ in 2016 as compared to long term mean (2003–2016). The combined analyses of various climatic factors, fires and associated BC emissions show that the observed snow cover decrease could be results of increased surface/atmospheric temperature due to combined effect of large scale climatic changes and BC absorption. Our results suggest that biomass burning can have significant effects on the Himalayan region, particularly in view of its importance in hydrological cycle and ecosystem.

Keywords : Forest fire, Biomass-burning, Black carbon, Organic carbon, Surface darkening, Himalayas

Effect of tillage and water management on GHG emissions from Mediterranean rice growing ecosystems

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Source: Volume 150, February 2017, Pages 303-312
<https://doi.org/10.1016/j.atmosenv.2016.11.020>

Paddy rice fields are an important source of greenhouse gases (GHG), especially methane. In the present work, we assessed the impact on GHG emissions of two main parameters of rice production: aerobic rice production was compared with traditional flooded rice production and conventional tillage (CT) was compared with short-term and long-term no-tillage (NT) management. A field experiment was performed over three years and the GHG emissions were measured during each year. Five treatments (3 replicates) were considered: **NTS7**: no-tillage over seven years and sprinkler irrigation; **NTS**: no-tillage and sprinkler irrigation; **CTS**: conventional tillage and sprinkler irrigation; **NTF**: no-tillage and flooding; **CTF**: conventional tillage and flooding. The use of sprinkler irrigation rather than flooding led to decreases in nitrous oxide and methane emissions of $\sim 40\%$ and more than 99% , respectively, over the 3-year experiment. The use of sprinkler irrigation compared with flooded irrigation reduced the global warming potential (GWP) about 40% and 36% in no-tillage and conventional tillage treatments, respectively. Treatment NTF decreased CH_4 emissions, relative to CTF, by $\sim 60\%$ over three years but the effect of NT on N_2O emissions was not clear: a decrease or no effect was mostly observed in the NT treatments, relative to CT. A decrease of $\sim 40\%$ in the total GHG emissions was observed in the NT treatments, relative to CT. No or small differences between NTS and NTS7 in terms of gaseous emissions were found. The short-term no-tillage and sprinkler irrigated treatment (NTS) gave lower yields than CTF in 2011 and 2012, but reached similar yields in the third year (NTS 8229 kg ha^{-1} ; CTF 8926 kg ha^{-1}), with average savings of 75% of the total amount of water applied in CTF. The NTS7 data showed that high yields (reaching 9805 kg ha^{-1} in 2012) and water savings are sustainable in the long term. Considering the yield-scaled GWP of the emissions, NT gave a decrease of up to 42% , relative to CT. However, the effect of water management on yield-scaled GWP depended on the soil management: yield-scaled GWP was higher with flooding when NT was used and lower when tillage was used. It can be concluded that, for aerobic rice production, NT is an efficient strategy to minimize GHG emissions while maintaining high levels of production.

An integrative approach for determination of air pollution and its health effects in a coal fired power plant area by passive sampling

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ÜzmezaAkifAriaGizemTuna
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Source: Volume 150, February 2017, Pages 331-345

<https://doi.org/10.1016/j.atmosenv.2016.11.025>

Ambient concentrations of nitrogen dioxide (NO₂), sulfur dioxide (SO₂), ozone (O₃) and volatile organic compounds (VOCs) were measured at several locations in Kütahya, a severely polluted city and also characterized as a thermal power plant city, in Turkey. Two-week extensive passive sampling campaigns were carried out in summer and winter at 108 sampling sites that were classified into three main groups as urban, rural and industrial. Spatial and seasonal distributions of the measured pollutants were evaluated employing Geographical Information System techniques. All pollutant concentrations showed an increasing pattern in winter, except for ozone. The concentrations of VOCs were substantially higher particularly at sampling sites with high traffic and population densities. Power plants were noted as important sources for VOCs since high concentrations were measured especially around the power plants. Highest NO₂ levels were observed in the city center while there was a general decrease in the concentration levels far away from the city center. Considerably higher SO₂ levels were observed in the settlements where local coal is used for residential heating. Seasonal variations in SO₂ concentrations were quite low around the thermal power plants indicating their important effect on atmospheric levels. A basic population exposure assessment was conducted for two largest settlements of the province (Kütahya city center and Tavşanlı) by combining population density maps with pollutant distribution maps of NO₂ and SO₂. Exposure to NO₂ and SO₂ were assessed separately according to a classification made for different degrees of exposure. Cancer risks associated with inhalation of benzene were also estimated. Higher risk values were obtained from the sampling sites with higher population densities, especially in winter. Risk values estimated for 95 sampling sites were higher than EPA's acceptable risk value (1×10^{-6})

Keywords : Volatile organic compounds (VOCs), Inorganic pollutants, Passive sampling, Power plant, Carcinogenic risk

Chemical characteristics of atmospheric PM_{2.5} loads during air pollution episodes in Giza, Egypt

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Source: Volume 150, February 2017, Pages 346-355

<https://doi.org/10.1016/j.atmosenv.2016.11.026>

Several types of pollution episodes, including dust storm (DSs), haze dust (HDs), straw rice combustions (SRCs) are common phenomena and represent severe environmental hazard in Egypt. This study provides the first comprehensive analysis of the chemical characteristics of aerosol during air pollution episodes at an urban area in Giza, Egypt. PM_{2.5} samples during various PM episodes during 2013–2014 were collected and analyzed. Results indicate that the highest PM_{2.5} mass concentrations were found during DSs (250 µg/m³), followed by HDs (130 µg/m³) and SRCs (103 µg/m³). Average PM_{2.5} mass concentrations were 1.91, 3.68 and 1.68 times higher than on normal days (NDs) during HDs, DSs and SRCs, respectively. The highest total water-soluble ions concentration was 61.1 µg/m³ during HDs, followed by SRCs (41.9 µg/m³) and DSs (35.2 µg/m³). SO₄²⁻ is the most abundant chemical components on the three PM episodes. Secondary inorganic ions (NO₃⁻, SO₄²⁻, and NH₄⁺) were enriched during HDs. The total secondary inorganic ions concentrations were 3.17, 1.39 and 1.75 times higher than NDs during HDs days, DSs days and SRCs days, respectively. PM from SRCs showed high K⁺ and Cl⁻. SO₄²⁻/K⁺, NO₃⁻/SO₄²⁻ and Cl⁻/K⁺ ratios proved effective as indicators for different pollution episodes. A Ca²⁺/Al ratio indicates that soil dust was dominant during DSs. Ion balance calculations indicated that PM_{2.5} from HDs was acidic, while the DSs and SRCs particles were alkaline and the NDs particle's was nearly neutral. The total crustal and anthropogenic metals concentrations were higher in DSs than other PM episodes and normal days. The enrichment factors values in PM episodes and normal days indicate that Fe and Mn in NDs, HDs, DSs and SRCs as well as Cr and Ni in DSs come mainly from crustal sources, whereas Cr, Ni, Co, Cu, Zn, Pb and Cd in PM episodes and NDs are anthropogenic.

Keywords : PM_{2.5}, Water-soluble ions, Mass balance, Heavy metals, Pollution episodes, Giza

Modelling and analysis of ozone concentration by artificial intelligent techniques for estimating air quality

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Source: Volume 150, February 2017, Pages 356-365

<https://doi.org/10.1016/j.atmosenv.2016.11.030>

High ozone concentration is an important cause of air pollution mainly due to its role in the greenhouse gas emission. Ozone is produced by photochemical processes which contain nitrogen oxides and volatile organic compounds in the lower atmospheric level. Therefore, monitoring and controlling the quality of air in the urban environment is very important due to the public health care. However, air quality prediction is a highly complex and non-linear process; usually several attributes have to be considered. Artificial intelligent (AI) techniques can be employed to monitor and evaluate the ozone concentration level.

The aim of this study is to develop an Adaptive Neuro-Fuzzy inference approach (ANFIS) to determine the influence of peripheral factors on air quality and pollution which is an arising problem due to ozone level in Jeddah city. The concentration of ozone level was considered as a

factor to predict the Air Quality (AQ) under the atmospheric conditions. Using Air Quality Standards of Saudi Arabia, ozone concentration level was modelled by employing certain factors such as; nitrogen oxide (NO_x), atmospheric pressure, temperature, and relative humidity. Hence, an ANFIS model was developed to observe the ozone concentration level and the model performance was assessed by testing data obtained from the monitoring stations established by the General Authority of Meteorology and Environment Protection of Kingdom of Saudi Arabia. The outcomes of ANFIS model were re-assessed by fuzzy quality charts using quality specification and control limits based on US-EPA air quality standards. The results of present study show that the ANFIS model is a comprehensive approach for the estimation and assessment of ozone level and is a reliable approach to produce more genuine outcomes.

Keywords : Air quality, Environmental factors, ANFIS, Modelling, Ozone level

Emissions of indoor air pollutants from six user scenarios in a model room

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Source: Volume 150, February 2017, Pages 389-394

<https://doi.org/10.1016/j.atmosenv.2016.11.033>

In this study six common user scenarios putatively influencing indoor air quality were performed in a model room constructed according to the specifications of the European Reference Room given in the new horizontal prestandard prEN 16516 to gain further information about the influence of user activities on indoor air quality. These scenarios included the use of cleaning agent, an electric air freshener, an ethanol fireplace and cosmetics as well as cigarette smoking and peeling of oranges. Four common indoor air pollutants were monitored: volatile organic compounds (VOC), particulate matter (PM), carbonyl compounds and CO₂. The development of all pollutants was determined during and after the test performance. For each measured pollutant, well-defined maximum values could be assigned to one or more of the individual user scenarios. The highest VOC concentration was measured during orange-peeling reaching a maximum value of 3547 µg m⁻³. Carbonyl compounds and PM were strongly elevated while cigarette smoking. Here, a maximum formaldehyde concentration of 76 µg m⁻³ and PM concentration of 378 µg m⁻³ were measured. CO₂ was only slightly affected by most of the tests except the use of the ethanol fireplace where a maximum concentration of 1612 ppm was reached. Generally, the user scenarios resulted in a distinct increase of several indoor pollutants that usually decreased rapidly after the removal of the source.

Keywords : Indoor air quality, Model room, VOC, Carbonyl compounds, Particulate matter, CO₂

Exposure assessment models for elemental components of particulate matter in an urban environment: A comparison of regression and random forest approaches

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Source: Volume 151, February 2017, Pages 1-11

<https://doi.org/10.1016/j.atmosenv.2016.11.066>

Exposure assessment for elemental components of particulate matter (PM) using land use modeling is a complex problem due to the high spatial and temporal variations in pollutant concentrations at the local scale. Land use regression (LUR) models may fail to capture complex interactions and non-linear relationships between pollutant concentrations and land use variables. The increasing availability of big spatial data and machine learning methods present an opportunity for improvement in PM exposure assessment models. In this manuscript, our objective was to develop a novel land use random forest (LURF) model and compare its accuracy and precision to a LUR model for elemental components of PM in the urban city of Cincinnati, Ohio. PM smaller than 2.5 μm (PM_{2.5}) and eleven elemental components were measured at 24 sampling stations from the Cincinnati Childhood Allergy and Air Pollution Study (CCAAPS). Over 50 different predictors associated with transportation, physical features, community socioeconomic characteristics, greenspace, land cover, and emission point sources were used to construct LUR and LURF models. Cross validation was used to quantify and compare model performance. LURF and LUR models were created for aluminum (Al), copper (Cu), iron (Fe), potassium (K), manganese (Mn), nickel (Ni), lead (Pb), sulfur (S), silicon (Si), vanadium (V), zinc (Zn), and total PM_{2.5} in the CCAAPS study area. LURF utilized a more diverse and greater number of predictors than LUR and LURF models for Al, K, Mn, Pb, Si, Zn, TRAP, and PM_{2.5} all showed a decrease in fractional predictive error of at least 5% compared to their LUR models. LURF models for Al, Cu, Fe, K, Mn, Pb, Si, Zn, TRAP, and PM_{2.5} all had a cross validated fractional predictive error less than 30%. Furthermore, LUR models showed a differential exposure assessment bias and had a higher prediction error variance. Random forest and other machine learning methods may provide more accurate exposure assessment.

Keywords : Elemental PM_{2.5}, Land use regression, Random forest

Large reductions in urban black carbon concentrations in the United States between 1965 and 2000

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Source: Volume 151, February 2017, Pages 17-23

<https://doi.org/10.1016/j.atmosenv.2016.11.001>

Long-term pollutant concentration trends can be useful for evaluating air quality effects of emission controls and historical transitions in energy sources. We employed archival records of coefficient of haze (COH), a now-retired measure of light-absorbing particulate matter, to re-construct historical

black carbon (BC) concentrations at urban locations in the United States (U.S.). The following relationship between COH and BC was determined by reinstating into service COH monitors beside aethalometers for two years in Vallejo and one year in San Jose, California: $BC (\mu\text{g m}^{-3}) = 6.7\text{COH} + 0.1$, $R^2 = 0.9$. Estimated BC concentrations in ten states stretching from the East to West Coast decreased markedly between 1965 and 1980: 5-fold in Illinois, Ohio, and Virginia, 4-fold in Missouri, and 2.5-fold in Pennsylvania. Over the period from the mid-1960s to the early 2000s, annual average BC concentrations in New Jersey and California decreased from 13 to 2 $\mu\text{g m}^{-3}$ and 4 to 1 $\mu\text{g m}^{-3}$, respectively, despite concurrent increases in fossil fuel consumption from 1.6 to 2.1 EJ (EJ = 1018 J) in New Jersey and 4.2 to 6.4 EJ in California. New Jersey's greater reliance on BC-producing heavy fuel oils and coal in the 1960s and early 1970s and subsequent transition to cleaner fuels explains why the decrease was larger in New Jersey than California. Patterns in seasonal and weekly BC concentrations and energy consumption trends together indicate that reducing wintertime emissions – namely substituting natural gas and electricity for heavy fuel oil in the residential sector – and decreasing emissions from diesel vehicles contributed to lower ambient BC concentrations. Over the period of study, declining concentrations of BC, a potent and short-lived climate warming pollutant, contrast increasing fossil fuel carbon dioxide (CO₂) emissions in the U.S. Declining BC emissions may have had the benefit of mitigating some atmospheric warming driven by increased CO₂ emissions with complementary health benefits.

Keywords : Concentration trends, Coefficient of haze, Elemental carbon, Air pollution, Energy consumption, Carbon dioxide emissions

Trend analysis of CO₂ and CH₄ recorded at a semi-natural site in the northern plateau of the Iberian Peninsula

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Source: Volume 151, February 2017, Pages 24-33

<https://doi.org/10.1016/j.atmosenv.2016.11.068>

CO₂ and CH₄ were recorded from October 2010 to February 2016 with a Picarro G1301 analyser at the centre of the upper plateau of the Iberian Peninsula. Large CO₂ values were observed during the vegetation growing season, and were reinforced by the stable boundary layer during the night. Annual CH₄ evolution may be explained by ecosystem activity and by the dispersion linked with the evolution of the boundary layer. Their trends were studied using an equation that considers one polynomial and one harmonic part. The polynomial part revealed an increasing trend from 0.8 to 2.3 ppm year⁻¹ for CO₂ and from 0.004 to 0.011 ppm year⁻¹ for CH₄. The harmonic part considered four harmonics whose amplitudes were noticeable for the first and second harmonics for CO₂ and for the first harmonic for CH₄. Long-term evolution was similar with alternative equations. Finally, seasonal study indicated summer minima for both gases, which may be explained by the lack of vegetation in this season. Harmonic analysis showed two maxima for CO₂, one in spring linked with vegetation growth, which decreased with time, and another in autumn related with the onset of plant activity after the summer, which increased with time. CH₄ presented only one maximum in winter and a short time with steady concentration in spring where the evolution of the boundary layer may play a noticeable role. The harmonic equation, which takes

into account all the observations, revealed opposite behaviour between CO₂, whose minima decreased, and CH₄, whose maxima increased.

Keywords : Carbon dioxide, Methane, Long-term analysis, Kernel smoothing, Local regression

The effects of marine vessel fuel sulfur regulations on ambient PM_{2.5} at coastal and near coastal monitoring sites in the U.S.

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Source: Volume 151, February 2017, Pages 52-61

<https://doi.org/10.1016/j.atmosenv.2016.12.012>

In August of 2012 the U.S. began implementing fuel sulfur limits on certain large commercial marine vessels within 200 nautical miles (nm) of its coasts as part of a North American Emissions Control Area (NA-ECA). The NA-ECA limited fuel sulfur use in these vessels to below 1% in 2012 and to below 0.1% starting in 2015. This work uses ambient PM_{2.5} monitoring data from the U.S. IMPROVE network and Positive Matrix Factorization (PMF) receptor modeling to assess the effectiveness of the NA-ECA at reducing ambient PM_{2.5} from high-sulfur residual fuel oil (RFO) use. RFO combustion emissions of PM_{2.5} are known to have a fairly unique vanadium (V) and nickel (Ni) trace metal signature. To determine if IMPROVE sites were affected by residual fuel oil combustion, V and Ni data from 65 IMPROVE sites in coastal States of the U.S. were analyzed from 2010 to 2011, the two years prior to NA-ECA implementation. 22 of these IMPROVE sites had a V and Ni correlation coefficient (r^2) greater than 0.65 and were selected for further analysis by PMF. The slopes of the correlations between V and Ni at these 22 sites ranged from 2.2 to 4.1, consistent with reported V:Ni emission ratios from RFO combustion. Each of the 22 IMPROVE sites was modeled independently with PMF, using the available PM_{2.5} chemical speciation data from 2010 to 2015. PMF model solutions for the 22 sites contained from 5 to 9 factors, depending on the site. At every site a PMF factor was identified that was associated with RFO combustion, however, 9 sites had PMF factors where RFO combustion was mixed with other aerosol sources. For the remaining 13 sites, PM_{2.5} from RFO combustion was analyzed for three time periods; 2010–2011 representing the time period prior to the NA-ECA implementation (pre-NA-ECA), 2013–2014 representing the time period where fuel sulfur was limited to 1.0% (NA-ECA 1.0% S), and 2015 representing the time period where fuel sulfur was limited to 0.1% (NA-ECA 0.1% S). All 13 sites indicated statistically significant reductions in the contribution of RFO combustion to PM_{2.5} between the pre-NA-ECA period and the two periods of fuel sulfur control. The average decrease in annual average PM_{2.5} from RFO combustion from the pre-NA-ECA to NA-ECA 1% S period was 50.2% (range, 29.0%–65.4%) and from the pre-NA-ECA to NA-ECA 0.1% S period was 74.1% (range, 33.0%–90.4%).

Keywords : PM_{2.5}, Marine vessel emissions, Positive matrix factorization, Source apportionment, Residual fuel oil, Emissions control area

Characteristics of volatile organic compounds (VOCs) from the evaporative emissions of modern passenger cars

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Source: Volume 151, February 2017, Pages 62-69

<https://doi.org/10.1016/j.atmosenv.2016.12.008>

Volatile organic compounds (VOCs) from vehicle evaporative emissions contribute substantially to photochemical air pollution. Yet, few studies of the characteristics of VOCs emitted from vehicle evaporative emissions have been published. We investigate the characteristics of 57 VOCs in hot soak, 24 h diurnal and 48 h diurnal emissions by applying the Sealed Housing Evaporative Determination unit (SHED) test to three modern passenger cars (one US Tier 2 and two China IV vehicles) using two different types of gasoline. The characteristics of the VOCs from the hot soak, 24 h diurnal and 48 h diurnal emissions were different due to their different emission mechanisms. In the hot soak emissions, toluene, isopentane/*n*-pentane, and 2,2,4-trimethylpentane were dominant species. In the 24 h and 48 h diurnal emissions, isopentane and *n*-pentane were dominant species. Toluene was the third most dominant component in the 24 h diurnal emissions but decreased by a mass of 42%–80% in the 48 h diurnal emissions. In the hot soak, 24 h diurnal and 48 h diurnal emissions, alkanes were generally the dominant hydrocarbons, followed by aromatics and olefins. However, owing to different evaporative emission mechanisms, the weight percentages of the aromatic hydrocarbons decreased and the weight percentages of the alkanes increased from the hot soak test to the 24 h diurnal and 48 h diurnal tests for each vehicle. The dominant contributors to the ozone formation potentials (OFPs) were also different in the hot soak, 24 h diurnal and 48 h diurnal emissions. The OFPs (g O₃/g VOC) of the hot soak emissions were higher than those of the 24 h and 48 h diurnal emissions. In addition, the combined effect of decreasing the olefin and aromatic contents of gasoline on vehicle evaporative emissions was investigated. The aromatics all decreased substantially in the hot soak, 24 h and 48 h diurnal emissions, and the total masses of the VOCs and OFPs decreased, with the greatest reduction occurring in the hot soak emissions when the fuel aromatic and olefin contents were reduced.

Keywords: *Evaporative emissions, Volatile organic compounds (VOCs), Ozone formation potential (OFP), Gasoline components*

Climatology of wintertime long-distance transport of surface-layer air masses arriving urban Beijing in 2001–2012

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Source: Volume 151, February 2017, Pages 70-81

<https://doi.org/10.1016/j.atmosenv.2016.12.003>

In this study, the FLEXPART-WRF coupled modeling system is used to conduct 12-year Lagrangian modeling over Beijing, China, for the winters of 2001–2012. Based on large trajectory tracking ensembles, the long-range air transport properties, in terms of geographic source regions within

the atmospheric planetary boundary layer (PBL) and large-scale ventilation, and its association with air quality levels were quantified from a climatological perspective. The results show the following: (1) The air masses residing in the near-surface layer over Beijing potentially originate from broader atmospheric boundary-layer regions, which cover vast areas with the backward tracking time elapsed. However, atmospheric transport from northeastern China and, to a lesser extent, from the surrounding regions of Beijing is important. (2) The evolution of air quality over Beijing is negatively correlated with large-scale ventilation conditions, particularly at a synoptic timescale. Thus, the simple but robust backward-trajectory ventilation (BV) index defined in this study could facilitate operational forecasting of severe air pollution events. (3) By comparison, the relatively short-range transport occurring over transport timescales of less than 3 days from southern and southeastern Beijing and its surrounding areas plays a vital role in the formation of severe air pollution events during the wintertime. (4) Additionally, an interannual trend analysis suggests that the geographic sources and ventilation conditions also changed, at least over the last decade, corresponding to the strength variability of the winter East Asian monsoon.

Keywords : Long-distance transport, Backward trajectory, Large-scale ventilation, Air pollution, Beijing city

Emission rates of particle number, mass and black carbon by the Los Angeles International Airport (LAX) and its impact on air quality in Los Angeles

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Source: Volume 151, February 2017, Pages 82-93

<https://doi.org/10.1016/j.atmosenv.2016.12.005>

This study describes a series of air monitoring measurements of particle number (PN), black carbon (BC) and PM_{2.5} mass concentrations in the vicinity of the Los Angeles International Airport (LAX) (roughly 150 m downwind of the LAX's south runways) as well as on-road measurements of the aforementioned pollutants using a mobile platform on three major freeways (i.e., I-110, I-105, and I-405) during May–July 2016. All measurements were performed in the “impact zone” of LAX with the predominant westerly winds from coast to inland. The overall impact of aircraft emissions from the LAX airport and its facilities in comparison to vehicular emissions from freeways on air quality was evaluated on a local scale (i.e. areas in the vicinity of the airport). PN concentration was, on average, 4.1 ± 1.2 times greater at the LAX site than on the studied freeways. Particle number emission factors for takeoffs and landings were comparable, with average values of 8.69×10^{15} particles/kg fuel and 8.16×10^{15} particles/kg fuel, respectively, and indicated a nearly 4-fold statistically significant reduction in PN emission factors for takeoffs during the past decade. BC emission factors were 0.12 ± 0.02 and 0.11 ± 0.01 g/kg fuel during takeoffs and landings, respectively. Additionally, the mean PM_{2.5} emission factor values for takeoffs and landings were also comparable, with values of 0.38 ± 0.04 and 0.40 ± 0.05 g/kg fuel, respectively. Within the impact zone of the airport, an area of roughly 100 km² downwind of the LAX, measurements indicated that the LAX daily contributions to PN, BC, and PM_{2.5} were approximately 11, 2.5, and 1.4 times greater than those from the three surrounding freeways. These results underscore the

significance of the LAX airport as a major source of pollution within its zone of impact comparing to freeway emissions.

Keywords : Aircraft emissions, Vehicular emissions, Ultrafine particles, Emission factor, LAX airport

Lung deposited surface area in Leicester urban background site/UK: Sources and contribution of new particle formation

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Source: Volume 151, February 2017, Pages 94-107
<https://doi.org/10.1016/j.atmosenv.2016.12.002>

Lung Deposited Surface Area (LDSA) has been identified as a potential metric for the correlation of a physical aerosol particle properties with health outcomes. Currently, there is little urban LDSA data. As a case study, we investigated measurements of LDSA (alveolar) concentrations in a mid-size European city. LDSA and associated measurements were carried out over 1.5 years at an urban background site in Leicester, UK. Average LDSA concentrations in the cold (November–April) and warm (May–October) seasons of UK were 37 and 23 $\mu\text{m}^2 \text{cm}^{-3}$, respectively. LDSA correlates well ($R^2 = 0.65\text{--}0.7$, $r = 0.77\text{--}0.8$) with traffic related pollutants, such as equivalent black carbon (eBC) and NOX. We also report for the first time in the UK the correlation between an empirically derived LDSA and eBC. Furthermore, the effect of wind speed and direction on the LDSA was explored. Higher LDSA concentrations are observed at low wind speeds ($1\text{--}2 \text{ m s}^{-1}$), owing to local traffic emissions. In addition, the diurnal variation of LDSA showed a second peak in the afternoon under warm and relatively clean atmospheric conditions, which can be attributed to photochemical new particle formation (NPF) and growth into the Aitken mode range. These NPF events increased the average background LDSA concentrations from 15.5 to 35.5 $\mu\text{m}^2 \text{cm}^{-3}$, although they might not be health-relevant. Overall, the results support the notion that local traffic emissions are a major contributor to observed LDSA concentrations with a clear seasonal pattern with higher values during winter.

Keywords : Aerosol, Seasonal variation, Traffic emissions, Lung deposited surface area

Simulating ozone concentrations using precursor emission inventories in Delhi – National Capital Region of India

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Source: Volume 151, February 2017, Pages 117-132
<https://doi.org/10.1016/j.atmosenv.2016.12.009>

This study simulates ground level ozone concentrations in a heavily populated and polluted National Capital Region (NCR- Delhi) in India. Multi-sectoral emission inventories of ozone precursors are prepared at a high resolution of $4 \times 4 \text{ km}^2$ for the whole region covering the capital city of Delhi along with other surrounding towns and rural regions in NCR. Emission inventories show that transport sector accounts for 55% of the total NO_x emissions, followed by power plants (23%) and diesel generator sets (7%). In NMVOC inventories, transport sector again accounts for 33%, followed by evaporative emissions released from solvent use and fuel handling activities (30%), and agricultural residue burning (28%). Refuse burning contributes to 73% of CO emissions mainly due to incomplete combustion, followed by agricultural residue burning (14%). These emissions are spatially and temporally distributed across the study domain and are fed into the WRF-CMAQ models to predict ozone concentrations for the year 2012. Model validations are carried out with the observed values at different monitoring stations in Delhi. The performance of the models over various metrics used for evaluation was found to be satisfactory. Summers and post-monsoon seasons were better simulated than monsoon and winter seasons. Simulations have shown higher concentrations of ozone formation during summers and lesser during winters and monsoon seasons, mainly due to varying solar radiation affecting photo-chemical activities. Ozone concentrations are observed lower at those locations where NO_x emissions are higher, and concentrations increase close to the boundary of study domain when compared to the center of Delhi city. Downwind regions to Delhi are influenced by the ozone formed due to plume of precursor emissions released from Delhi. Considering significant background contributions, regional scale controls are required for reducing ozone in NCR.

Keywords : Ozone, NCR, WRF-CMAQ modeling, Air quality

Characteristics of an open-cut coal mine fire pollution event

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Source: Volume 151, February 2017, Pages 140-151

<https://doi.org/10.1016/j.atmosenv.2016.12.015>

On 9 February 2014, embers from a nearby grass/shrub fire spotted into an unused part of the Hazelwood open-cut brown coal mine located in the Latrobe Valley of Victoria, Australia and started a fire that spread rapidly and extensively throughout the mine under strong south-westerly winds and burned over a period of 45 days. The close proximity of the town to the coal mine and the low buoyancy of the smoke plume led to the accumulation of dense smoke levels in the township of Morwell (population of 14,000) particularly under south-westerly winds. A maximum daily PM_{2.5} concentration of $731 \mu\text{g m}^{-3}$ and 8-h CO concentration of 33 ppm were measured at Morwell South, the closest residential area located approximately 500 m from the mine. These concentrations were significantly higher than national air quality standards. Air quality monitoring undertaken in the Latrobe Valley showed that smoke from the Hazelwood mine fire affected a wide area, with particle air quality standards also exceeded in Traralgon (population of 25,000) located approximately 13 km from the mine. Pollutant levels were significantly elevated in February, decreased in March once the fire abated and then returned to background levels once the fire was declared safe at the end of March.

While the smoke extent was of a similar order of magnitude to other major air pollution events worldwide, a closer look at emissions ratios showed that the open combustion of lignite brown coal

in the Hazelwood mine was different to open combustion of biomass, including peat. It suggested that the dominant combustion process was char combustion. While particle and carbon monoxide monitoring started approximately 4 days after the fire commenced when smoke levels were very high, targeted monitoring of air toxics only began on 26 February (17 days after the fire) when smoke levels had subsided. Limited research on emission factors from open-cut coal mine fires make it difficult to assess the likely concentrations of air toxics emitted during the initial more intense period of the fire.

Keywords : Mine fire, Smoke, Carbon monoxide, PM2.5, Air toxics

Atmospheric emissions of typical toxic heavy metals from open burning of municipal solid waste in China

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Source: Volume 152, March 2017, Pages 6-15

<https://doi.org/10.1016/j.atmosenv.2016.12.017>

Municipal solid waste (MSW) contains considerable hazardous components and the widely-distributed open MSW burning in heavily-populated urban areas can cause direct exposure of hazardous materials to citizens. By determining the best available representation of composition-varying and time-varying emission factors with fuzzy mathematics method and S-shape curves, a comprehensive atmospheric emission inventories of 9 typical toxic heavy metals (THMs, e.g. mercury (Hg), arsenic (As), lead (Pb), cadmium (Cd), chromium (Cr), selenium (Se), copper (Cu), zinc (Zn), and nickel (Ni)) from open MSW burning activities in China is established during the period of 2000–2013 for the first time. Further, the emissions in 2013 are allocated at a high spatial resolution of $0.5^{\circ} \times 0.5^{\circ}$ grid by surrogate indexes. The results show that 9 typical THMs emissions from open MSW burning are estimated at 21.25 t for Hg, 131.52 t for As, 97.12 t for Pb, 10.12 t for Cd, 50.58 t for Cr, 81.95 t for Se, 382.42 t for Cu, 1790.70 t for Zn, and 43.50 t for Ni, respectively. In terms of spatial variation, the majority of emissions are concentrated in relatively developed and densely-populated regions, especially for the eastern, central and southern regions. Moreover, future emissions are also projected for the period of 2015–2030 based on different scenarios of the independent and collaborative effects of control proposals including minimizing waste, improving MSW incineration ratio, and enhancing waste sorting and recycling, etc. The collaborative effect of the above proposals is expected to bring the most effective reduction to THMs emissions from open MSW burning in China except for Hg. The results will be supplementary to all anthropogenic emissions and useful for relevant policy-making and the improvement of urban air quality as well as human health.

Keywords : MSW, Toxic heavy metals, Open burning, Emission characteristics, Control proposals

Combined use of land use regression and BenMAP for estimating public health benefits of reducing PM2.5 in Tianjin, China

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Source: Volume 152, March 2017, Pages 16-23
<https://doi.org/10.1016/j.atmosenv.2016.12.023>

To assess the public health benefits of reducing PM_{2.5} in Tianjin, we created an annual air quality surface with a land use regression (LUR) model conducted at a high spatial resolution (1 km). The predictors included in the final model were population density, road length within a 1000 m buffer, industrial land area within a 2000m buffer and distance to the coast. The fitting R² and the leave-one-out-cross-validation (LOOCV) R² of the PM_{2.5} LUR models were 0.78 and 0.73, respectively, suggesting that the predicted PM_{2.5} concentrations fitted well with the measured values for the entire year. Daily air quality surfaces were established based on historic concentration data and interpolation method. We evaluated avoided cases of mortality and morbidity in Tianjin, assuming achievement of China's current air quality daily and annual standards (No. GB3095-2012). Reducing the daily average PM_{2.5} to the daily Class II standard (75 µg/m³), the avoided emergency department visits, the deaths for cardiovascular disease and the deaths for respiratory disease are 85,000 (95% confidence interval (CI), 17,000–150,000), 2000 (95% CI, 920–3100) and 280 (95% CI, 94–460) per year respectively, and the monetary values are 23–42 million yuan, 180–4800 million yuan and 25–670 million yuan per year in 2015 yuan year respectively. Reducing the annual average PM_{2.5} to the annual Class II standard (35 µg/m³), the avoided emergency department visits, the deaths for cardiovascular disease and the deaths for respiratory disease are 59,000 (95% CI, 12,000–110,000), 1400 (95% CI, 640–2100) and 200 (95% CI, 66–320) per year respectively, and the monetary values are 16–29 million yuan, 130 to 3400 million yuan and 18 to 480 million yuan per year in 2015 yuan year respectively.

Keywords : Land use regression, Public health benefit, BenMAP, PM_{2.5}, Tianjin

Responses of human health and vegetation exposure metrics to changes in ozone concentration distributions in the European Union, United States, and China

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Source: Volume 152, March 2017, Pages 123-145
<https://doi.org/10.1016/j.atmosenv.2016.12.025>

The impacts of surface ozone (O₃) on human health and vegetation have prompted O₃precursor emission reductions in the European Union (EU) and United States (US). In contrast, until recently, emissions have increased in East Asia and most strongly in China. As emissions change, the distribution of hourly O₃ concentrations also changes, as do the values of exposure metrics. The distribution changes can result in the exposure metric trend patterns changing in a similar direction as trends in emissions (e.g., metrics increase as emissions increase) or, in some cases, in opposite directions. This study, using data from 481 sites (276 in the EU, 196 in the US, and 9 in China), investigates the response of 14 human health and vegetation O₃ exposure metrics to changes in hourly O₃ concentration distributions over time. At a majority of EU and US sites, there was a reduction in the frequency of both relatively high and low hourly average O₃ concentrations. In contrast, for some sites in mainland China and Hong Kong, the middle of the distribution shifted

upwards but the low end did not change and for other sites, the entire distribution shifted upwards. The responses of the 14 metrics to these changes at the EU, US, and Chinese sites were varied, and dependent on (1) the extent to which the metric was determined by relatively high, moderate, and low concentrations and (2) the relative magnitude of the shifts occurring within the O₃ concentration distribution. For example, the majority of the EU and US sites experienced decreasing trends in the magnitude of those metrics associated with higher concentrations. For the sites in China, all of the metrics either increased or had no trends. In contrast, there were a greater number of sites that had no trend for those metrics determined by a combination of moderate and high O₃ concentrations. A result of our analyses is that trends in mean or median concentrations did not appear to be well associated with some exposure metrics applicable for assessing human health or vegetation effects. The identification of shifting patterns in the O₃ distribution and the resulting changes in O₃ exposure metrics across regions with large emission increases and decreases is an important step in examining the linkage between emissions and exposure metric trends. The results provide insight into the utility of using specific exposure metrics for assessing emission control strategies.

Keywords : Binning concentrations, Exposure metrics, Ozone distributions, NO_x scavenging, Shifting concentrations, Trends

Inhalation exposure and health risk levels to BTEX and carbonyl compounds of traffic policeman working in the inner city of Bangkok, Thailand

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Source: Volume 152, March 2017, Pages 111-120

<https://doi.org/10.1016/j.atmosenv.2016.11.062>

Benzene, toluene, ethylbenzene and xylenes (BTEX) and carbonyl compounds (CCs) are recognized traffic-related air pollutants in urban environments and are the focus of this study. In Bangkok, the BTEX and CC concentrations in both ambient air and personal exposure samples were studied during two periods (April–May and August–September 2014) at four different sampling sites around the Pathumwan District (three intersections and one T-junction). Traffic policemen, representing the high-exposure group for these toxic air pollutants, were observed, and the health risk to these workers was evaluated. Toluene was the predominant aromatic compound in the ambient and personal exposure samples. The maximum average ambient concentration of BTEX was 2968.96 µg/m³. Formaldehyde and acetaldehyde were the most abundant CCs at all of the sampling sites, with the greatest mean concentrations of these substances being 21.50 µg/m³ and 64.82 µg/m³, respectively. In the personal exposure samples, the highest levels of BTEX, formaldehyde and acetaldehyde concentrations were 2231.85 µg/m³, 10.61 µg/m³, and 16.03 µg/m³, respectively. In terms of risk assessment, benzene posed the greatest cancer risk (at the 95% CI), followed by toluene, acetaldehyde and formaldehyde (1.15E-02, 5.14E-03, 2.84E-04, and 2.52E-04, respectively). Three risk factors were investigated to reduce the total cancer risk levels: reducing the chemical concentration, exposure time and exposure duration. The use of a mask (chemical concentration) was the best way to reduce the risk to traffic police. However, the

risk value of benzene (average 1.57E-05) was still higher than an acceptable value when using a mask.

Keywords : BTEX, Carbonyl compounds, Health risk assessment, Traffic policemen, Bangkok

Responses of human health and vegetation exposure metrics to changes in ozone concentration distributions in the European Union, United States, and China

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Source: Volume 152, March 2017, Pages 123-145
<https://doi.org/10.1016/j.atmosenv.2016.12.025>

The impacts of surface ozone (O₃) on human health and vegetation have prompted O₃ precursor emission reductions in the European Union (EU) and United States (US). In contrast, until recently, emissions have increased in East Asia and most strongly in China. As emissions change, the distribution of hourly O₃ concentrations also changes, as do the values of exposure metrics. The distribution changes can result in the exposure metric trend patterns changing in a similar direction as trends in emissions (e.g., metrics increase as emissions increase) or, in some cases, in opposite directions. This study, using data from 481 sites (276 in the EU, 196 in the US, and 9 in China), investigates the response of 14 human health and vegetation O₃ exposure metrics to changes in hourly O₃ concentration distributions over time. At a majority of EU and US sites, there was a reduction in the frequency of both relatively high and low hourly average O₃ concentrations. In contrast, for some sites in mainland China and Hong Kong, the middle of the distribution shifted upwards but the low end did not change and for other sites, the entire distribution shifted upwards. The responses of the 14 metrics to these changes at the EU, US, and Chinese sites were varied, and dependent on (1) the extent to which the metric was determined by relatively high, moderate, and low concentrations and (2) the relative magnitude of the shifts occurring within the O₃ concentration distribution. For example, the majority of the EU and US sites experienced decreasing trends in the magnitude of those metrics associated with higher concentrations. For the sites in China, all of the metrics either increased or had no trends. In contrast, there were a greater number of sites that had no trend for those metrics determined by a combination of moderate and high O₃ concentrations. A result of our analyses is that trends in mean or median concentrations did not appear to be well associated with some exposure metrics applicable for assessing human health or vegetation effects. The identification of shifting patterns in the O₃ distribution and the resulting changes in O₃ exposure metrics across regions with large emission increases and decreases is an important step in examining the linkage between emissions and exposure metric trends. The results provide insight into the utility of using specific exposure metrics for assessing emission control strategies.

Keywords : Binning concentrations, Exposure metrics, Ozone distributions, NOx scavenging, Shifting concentrations, Trends

Comprehensive chemical characterization of industrial PM_{2.5} from steel industry activities

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Source: Volume 152, March 2017, Pages 180-190

Industrial sources are among the least documented PM (Particulate Matter) source in terms of chemical composition, which limits our understanding of their effective impact on ambient PM concentrations. We report 4 chemical emission profiles of PM_{2.5} for multiple activities located in a vast metallurgical complex. Emissions profiles were calculated as the difference of species concentrations between an upwind and a downwind site normalized by the absolute PM_{2.5} enrichment between both sites. We characterized the PM_{2.5} emissions profiles of the industrial activities related to the cast iron (complex 1) and the iron ore conversion processes (complex 2), as well as 2 storage areas: a blast furnace slag area (complex 3) and an ore terminal (complex 4). PM_{2.5} major fractions (Organic Carbon (OC) and Elemental Carbon (EC), major ions), organic markers as well as metals/trace elements are reported for the 4 industrial complexes. Among the trace elements, iron is the most emitted for the complex 1 (146.0 mg g⁻¹ of PM_{2.5}), the complex 2 (70.07 mg g⁻¹) and the complex 3 (124.4 mg g⁻¹) followed by Al, Mn and Zn. A strong emission of Polycyclic Aromatic Hydrocarbons (PAH), representing 1.3% of the Organic Matter (OM), is observed for the iron ore transformation complex (complex 2) which merges the activities of coke and iron sinter production and the blast furnace processes. In addition to unsubstituted PAHs, sulfur containing PAHs (SPAHS) are also significantly emitted (between 0.011 and 0.068 mg g⁻¹) by the complex 2 and could become very useful organic markers of steel industry activities. For the complexes 1 and 2 (cast iron and iron ore converters), a strong fraction of sulfate ranging from 0.284 to 0.336 g g⁻¹ and only partially neutralized by ammonium, is observed indicating that sulfates, if not directly emitted by the industrial activity, are formed very quickly in the plume. Emission from complex 4 (Ore terminal) are characterized by high contribution of Al (125.7 mg g⁻¹ of PM_{2.5}) but also, in a lesser extent, of Fe, Mn, Ti and Zn. We also highlighted high contribution of calcium ranging from 0.123 to 0.558 g g⁻¹ for all of the industrial complexes under study. Since calcium is also widely used as a proxy of the dust contributions in source apportionment studies, our results suggest that this assumption should be reexamined in environments impacted by industrial emissions

Keywords : Chemical profiles, PM_{2.5}, Steel industry, Organics markers, Trace elements

Ambient air quality measurements from a continuously moving mobile platform: Estimation of area-wide, fuel-based, mobile source emission factors using absolute principal component scores

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Source: Volume 152, March 2017, Pages 201-211
<https://doi.org/10.1016/j.atmosenv.2016.12.037>

We have applied the absolute principal component scores (APCS) receptor model to on-road, background-adjusted measurements of NO_x, CO, CO₂, black carbon (BC), and particle number (PN) obtained from a continuously moving platform deployed over nine afternoon sampling periods in Seattle, WA. Two Varimax-rotated principal component features described 75% of the overall variance of the observations. A heavy-duty vehicle feature was correlated with black carbon and particle number, whereas a light-duty feature was correlated with CO and CO₂. NO_x had moderate correlation with both features. The bootstrapped APCS model predictions were used to estimate area-wide, average fuel-based emission factors and their respective 95% confidence limits. The average emission factors for NO_x, CO, BC and PN (14.8, 18.9, 0.40 g/kg, and 4.3×10^{15} particles/kg for heavy duty vehicles, and 3.2, 22.4, 0.016 g/kg, and 0.19×10^{15} particles/kg for light-duty vehicles, respectively) are consistent with previous estimates based on remote sensing, vehicle chase studies, and recent dynamometer tests. Information on the spatial distribution of the concentrations contributed by these two vehicle categories relative to background during the sampling period was also obtained.

Keywords : Vehicle exhaust emission factors, Mobile monitoring, Principal component analysis, Traffic related air pollution, On-road air pollution

Impact of future climate policy scenarios on air quality and aerosol-cloud interactions using an advanced version of CESM/CAM5: Part I. model evaluation for the current decadal simulations

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Source: Volume 152, March 2017, Pages 222-239
<https://doi.org/10.1016/j.atmosenv.2016.12.035>

A version of the Community Earth System Model modified at the North Carolina State University (CESM-NCSU) is used to simulate the current and future atmosphere following the representative concentration partway scenarios for stabilization of radiative forcing at 4.5 W m^{-2} (RCP4.5) and radiative forcing of 8.5 W m^{-2} (RCP8.5). Part I describes the results from a comprehensive evaluation of current decadal simulations. Radiation and most meteorological variables are well simulated in CESM-NCSU. Cloud parameters are not as well simulated due in part to the tuning of model radiation and general biases in cloud variables common to all global chemistry-climate

models. The concentrations of most inorganic aerosol species (i.e., SO_4^{2-} , NH_4^+ , and NO_3^-) are well simulated with normalized mean biases (NMBs) typically less than 20%. However, some notable exceptions are European NH_4^+ , which is overpredicted by 33.0–42.2% due to high NH_3 emissions and irreversible coarse mode condensation, and Cl^- , that is negatively impacted by errors in emissions driven by wind speed and overpredicted HNO_3 . Carbonaceous aerosols are largely underpredicted following the RCP scenarios due to low emissions of black carbon, organic carbon, and anthropogenic volatile compounds in the RCP inventory and efficient wet removal. This results in underpredictions of $\text{PM}_{2.5}$ and PM_{10} by 6.4–55.7%. The column mass abundances are reasonably well simulated. Larger biases occur in surface mixing ratios of trace gases in CESM-NCSU, likely due to numerical diffusion from the coarse grid spacing of the CESM-NCSU simulations or errors in the magnitudes and vertical structure of emissions. This is especially true for SO_2 and NO_2 . The mixing ratio of O_3 is overpredicted by 38.9–76.0% due to the limitations in the O_3 deposition scheme used in CESM and insufficient titration resulted from large underpredictions in NO_2 . Despite these limitations, CESM-NCSU reproduces reasonably well the current atmosphere in terms of radiation, clouds, meteorology, trace gases, aerosols, and aerosol-cloud interactions, making it suitable for future climate simulations

Keywords : CESM/CAM5, Representative concentration pathways, Earth system modeling, Current air quality, Model evaluation, Aerosol indirect effects

Relative impact of on-road vehicular and point-source industrial emissions of air pollutants in a medium-sized Andean city

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Source: Volume 152, March 2017, Pages 279-289

<https://doi.org/10.1016/j.atmosenv.2016.12.048>

Cities in emerging countries are facing a fast growth and urbanization; however, the study of air pollutant emissions and its dynamics is scarce, making their populations vulnerable to potential effects of air pollution. This situation is critical in medium-sized urban areas built along the tropical Andean mountains. This work assesses the contribution of on-road vehicular and point-source industrial activities in the medium-sized Andean city of Manizales, Colombia. Annual fluxes of criteria pollutants, NMVOC, and greenhouse gases were estimated. Emissions were dominated by vehicular activity, with more than 90% of total estimated releases for the majority of air pollutants. On-road vehicular emissions for CO (43.4 Gg/yr) and NMVOC (9.6 Gg/yr) were mainly associated with the use of motorcycles (50% and 81% of total CO and NMVOC emissions respectively). Public transit buses were the main source of PM_{10} (47%) and NO_x (48%). The per-capita emission index was significantly higher in Manizales than in other medium-sized cities, especially for NMVOC, CO, NO_x and CO_2 . The unique mountainous terrain of Andean cities suggest that a methodology based on VSP model could give more realistic emission estimates, with additional model components that include slope and acceleration. Food and beverage facilities were the main contributors of point-source industrial emissions for PM_{10} (63%), SO_x (55%) and NO_x (45%), whereas scrap metal recycling had high emissions of CO (73%) and NMVOC (47%). Results provide the baseline for

ongoing research in atmospheric modeling and urban air quality, in order to improve the understanding of air pollutant fluxes, transport and transformation in the atmosphere. In addition, this emission inventory could be used as a tool to identify areas of public health exposure and provide information for future decision makers.

Keywords : Air pollution, Emission inventories, On-road vehicular sources, Industrial point-sources, Medium-sized cities

A modeling study on the effect of urban land surface forcing to regional meteorology and air quality over South China

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Source: Volume 152, March 2017, Pages 389-404

<https://doi.org/10.1016/j.atmosenv.2016.12.053>

The change of land-use from natural to artificial surface induced by urban expansion can deeply impact the city environment. In this paper, the model WRF/Chem is applied to explore the effect of this change on regional meteorology and air quality over South China, where people have witnessed a rapid rate of urbanization. Two sets of urban maps are adopted to stand for the pre-urbanization and the present urban land-use distributions. Month-long simulations are conducted for January and July, 2014. The results show that urban expansion can obviously change the weather conditions around the big cities of South China. Especially in the Pearl River Delta region (PRD), the urban land-use change can increase the sensible heat flux by 40 W/m² in January and 80 W/m² in July, while decrease the latent heat flux about -50 W/m² in January and -120 W/m² in July. In the consequent, 2-m air temperature (T₂) increases as much as 1 °C and 2 °C (respective to January and July), planetary boundary layer height (PBLH) rises up by 100–150 m and 300 m, 10-m wind speed (WS₁₀) decreases by -1.2 m/s and -0.3 m/s, and 2-m specific humidity is reduced by -0.8 g/kg and -1.5 g/kg. Also, the precipitation in July can be increased as much as 120 mm, with more heavy rains and rainstorms. These variations of meteorological factors can significantly impact the spatial and vertical distribution of air pollutants as well. In PRD, the enhanced updraft can reduce the surface concentrations of PM₁₀ by -40 µg/m³ (30%) in January and -80 µg/m³ (50%) in July, but produce a correlating increase in the concentrations at higher atmospheric layers. However, according to the increase in T₂ and the decrease in surface NO, the surface concentrations of O₃ in PRD can increase by 2–6 ppb in January and 8–12 ppb in July. Meanwhile, there is a significant increase in the O₃ concentrations at upper layers above PRD, which should be attributed to the increase in air temperature and the enhanced upward transport of O₃ and its precursors. As for some relative small cities, such as Haikou, there is very little variation in surface PM₁₀ and O₃ in both months, implying less urbanization in these areas. Moreover, the depletion of O₃ by NO may be the main cause of the reduction of O₃ at upper layers in these small cities.

Keywords : Urbanization, Land-use, South China, Meteorological conditions, Air quality, WRF/Chem

A three-year investigation of daily PM_{2.5} main chemical components in four sites: the routine measurement program of the Supersito Project (Po Valley, Italy)

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Source: Volume 152, March 2017, Pages 418-430
<https://doi.org/10.1016/j.atmosenv.2016.12.052>

The Supersito Project (www.supersito-er.it) has been active in the Emilia-Romagna region, southern part of the Po Valley (Italy), since 2011. Focal aim of the project is to enhance the knowledge on atmospheric aerosol and its impact on human health. In the framework of Supersito, major chemical components of daily PM_{2.5} were investigated over a period of more than three years at four sampling sites, representative of dissimilar territorial conditions: one rural background (SPC) and three urban background sites in the coastal (RN), central (MS) and inner area (PR) of the region.

In all the sites, organic and elemental carbon and water soluble inorganic ions accounted for more than 70% of PM_{2.5} mass, during all seasons. Nitrate and organic carbon (OC) were the main components of winter PM_{2.5}, while summer aerosol was mainly contributed by OC and sulphate. OC was dominated by primary sources, with a potentially important contribution from biomass burning, in winter, while secondary processes dominated OC production in summer.

A substantial homogeneity was observed on a regional scale in terms of spatial distribution of pollutants, with EC only presenting significant differences between urban and rural areas during winters. Nonetheless, differences were observed between the coastal and the inner part of the region, with the former being systematically characterized by higher concentrations of carbonaceous compounds and lower concentrations of ammonium nitrate. The coastal area was likely influenced by the aged OC from the Po Valley outflow in addition to local sources, while the scarcity of local sources of ammonia limited the formation of ammonium nitrate.

In the studied area, local and regional meteorology - mostly governed by geographical collocation and orography - was responsible for PM_{2.5} mass and composition no less than local and regional emission sources.

Keywords : PM_{2.5}, Nitrate, Organic carbon, Elemental carbon, Po Valley

Heterogeneous reaction of SO₂ with soot: The roles of relative humidity and surface composition of soot in surface sulfate formation

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Source: Volume 152, March 2017, Pages 465-476
<https://doi.org/10.1016/j.atmosenv.2017.01.005>

The conversion of SO₂ to sulfates on the surface of soot is still poorly understood. Soot samples with different fractions of unsaturated hydrocarbons and oxygen-containing groups were prepared by combusting n-hexane under well-controlled conditions. The heterogeneous reaction of SO₂ with soot was investigated using in situ attenuated total internal reflection infrared (ATR-IR) spectroscopy, ion chromatography (IC) and a flow tube reactor at the ambient pressure and relative humidity (RH). Water promoted SO₂ adsorption and sulfate formation at the RH range from 6% to 70%, while exceeded water condensed on soot was unfavorable for sulfate formation due to inhibition of SO₂ adsorption when RH was higher than 80%. The surface composition of soot, which was governed by combustion conditions, also played an important role in the heterogeneous reaction of SO₂ with soot. This effect was found to greatly depend on RH. At low RH of 6%, soot with the highest fuel/oxygen ratio of 0.162 exhibited a maximum uptake capacity for SO₂ because it contained a large amount of aromatic C—H groups, which acted as active sites for SO₂ adsorption. At RH of 54%, soot produced with a fuel/oxygen ratio of 0.134 showed the highest reactivity toward SO₂ because it contained appropriate amounts of aromatic C—H groups and oxygen-containing groups, subsequently leading to the optimal surface concentrations of both SO₂ and water. These results suggest that variation in the surface composition of soot from different sources and/or resulting from chemical aging in the atmosphere likely affects the conversion of SO₂ to sulfates.

Keywords : Soot, SO₂, Relative humidity, Surface composition, Sulfate formation

Impact of future climate policy scenarios on air quality and aerosol-cloud interactions using an advanced version of CESM/CAM5: Part II. Future trend analysis and impacts of projected anthropogenic emissions

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Source: Volume 152, March 2017, Pages 531-552

<https://doi.org/10.1016/j.atmosenv.2016.12.034>

Following a comprehensive evaluation of the Community Earth System Model modified at the North Carolina State University (CESM-NCSU), Part II describes the projected changes in the future state of the atmosphere under the representative concentration partway scenarios (RCP4.5 and 8.5) by 2100 for the 2050 time frame and examine the impact of climate change on future air quality under both scenarios, and the impact of projected emission changes under the RCP4.5 scenario on future climate through aerosol direct and indirect effects. Both the RCP4.5 and RCP8.5 simulations predict similar changes in air quality by the 2050 period due to declining emissions under both scenarios. The largest differences occur in O₃, which decreases by global mean of 1.4 ppb under RCP4.5 but increases by global mean of 2.3 ppb under RCP8.5 due to differences in methane levels, and PM₁₀, which decreases by global mean of 1.2 µg m⁻³ under RCP4.5 and increases by global mean of 0.2 µg m⁻³ under RCP8.5 due to differences in dust and sea-salt emissions under both scenarios. Enhancements in cloud formation in the Arctic and Southern Ocean and increases of aerosol optical depth (AOD) in central Africa and South Asia dominate the change in surface radiation in both scenarios, leading to global average dimming of 1.1 W m⁻² and 2.0 W m⁻² in the RCP4.5 and

RCP8.5 scenarios, respectively. Declines in AOD, cloud formation, and cloud optical thickness from reductions of emissions of primary aerosols and aerosol precursors under RCP4.5 result in near surface warming of 0.2 °C from a global average increase of 0.7 W m⁻² in surface downwelling solar radiation. This warming leads to a weakening of the Walker Circulation in the tropics, leading to significant changes in cloud and precipitation that mirror a shift in climate towards the negative phase of the El Nino Southern Oscillation.

Keywords : CESM/CAM5, Representative concentration pathways, Global climate change, Future air quality, Climate change, Emission changes

Large-eddy simulation of dense gas dispersion over a simplified urban area

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Source: Volume 152, March 2017, Pages 605-616

<https://doi.org/10.1016/j.atmosenv.2016.12.039>

Dispersion of neutral and dense gas over a simplified urban area, comprising four cubes, has been investigated by the means of large-eddy simulations (LES). The results have been compared to wind tunnel experiments and both mean and fluctuating quantities of velocity and concentration are in very good agreement.

High-quality inflow profiles are necessary to achieve physically realistic LES results. In this study, profiles matching the atmospheric boundary layer flow in the wind tunnel, are generated by means of a separate precursor simulation.

Emission of dense gas dramatically alters the flow in the near source region and introduces an upstream dispersion. The resulting dispersion patterns of neutral and dense gas differ significantly, where the plume in the latter case is wider and shallower. The dense gas is highly affected by the cube array, which seems to act as a barrier, effectively deflecting the plume. This leads to higher concentrations outside of the array than inside. On the contrary, the neutral gas plume has a Gaussian-type shape, with highest concentrations along the centreline.

It is found that the dense gas reduces the vertical and spanwise turbulent momentum transport and, as a consequence, the turbulence kinetic energy. The reduction coincides with the area where the gradient Richardson number exceeds its critical value, i.e. where the flow may be characterized as stably stratified. Interestingly, this region does not correspond to where the concentration of dense gas is the highest (close to the ground), as this is also where the largest velocity gradients are to be found. Instead there is a layer in the middle of the dense gas cloud where buoyancy is dynamically dominant.

Keywords : LES, Wind tunnel, Dispersion, Dense gas, Urban flow, Atmospheric boundary layer

Environmental studies in two communes of Santiago de Chile by the analysis of magnetic properties of particulate matter deposited on leaves of roadside trees

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Source: Volume 152, March 2017, Pages 617-627

<https://doi.org/10.1016/j.atmosenv.2016.12.047>

Emissions from motor vehicles are considered to be one of the main sources of airborne particulate matter in Santiago. International researchers have shown that particulate matter contains metal oxides and magnetic particles, both of which are emitted mainly from vehicles exhaust pipes. On the other hand, trees are effective in reducing such contamination, so that they act as passive collectors of particulate matter. This work presents the results obtained from the first magnetic study of the particulate matter collected in two areas of the city of Santiago de Chile. Magnetic susceptibility and Saturation Isothermic Remanent Magnetization (SIRM) were determined in leaves from abundant urban trees and from urban dust samples. Results indicate that most of the samples contain ferromagnetic minerals with magnetite (Fe₃O₄) as the main carrier. Values of magnetic susceptibility (SI $\times 10^{-6}$ m³/kg) in the range 0.04–0.24 for leaves and in the range 10–45 for urban dust were determined. In one of the city areas studied, significant correlation between the particulate matter deposited on leaves of *Platanus orientalis* and measured traffic flows was obtained. In addition, it was possible to estimate that the species *Platanus orientalis* and *Acer negundo* have a better ability to capture particulate matter than the species *Robinia pseudoacacia*.

Keywords : Airborne particulate matter capture, Urban trees, Magnetic measurements, Traffic flows, Santiago de Chile

Content, mineral allocation and leaching behavior of heavy metals in urban PM_{2.5}

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Source: Volume 153, March 2017, Pages 47-60

<https://doi.org/10.1016/j.atmosenv.2017.01.009>

To clarify the relationship between airborne particulate exposure and negative impacts on human health, focusing on the heavy metal content alone might not be sufficient. To address this issue, in the present work, mineral allocation and leaching behavior of heavy metals in the PM_{2.5} were investigated. This work, therefore, provides a novel perspective in the field of urban airborne particle investigation that is not currently found in the literature. Four sampling campaigns were

performed in the urban area of Rome (Central Italy) during the winter and summer seasons (February and July 2013 and 2014, respectively). The measured concentrations of the regulated elements of As, Cd, Ni and Pb were consistent with those reported by the local Environmental Agency (ARPA Lazio), but non-regulated heavy metals, including Fe, Cu, Cr and Zn, were also found in PM_{2.5} and analyzed in detail. As a novelty, heavy metals were associated with the host-identified mineral phases, primarily oxides and alloys, and to a lesser extent, other minerals, such as sulfates, carbonates and silicates. Leaching tests of the collected samples were conducted in a buffered solution mimicking the bodily physiological environment. Despite the highest concentration of heavy metals found during the winter sampling period, all of the elements showed a leaching trend leading to major mobility during the summer period. To explain this result, an interesting comparative analysis between the leaching test behavior and innovative mineral allocation was conducted. Both the heavy metal content and mineral allocation in PM_{2.5} might contribute to the bioavailability of toxic elements in the pulmonary environment. Hence, for regulatory purposes, the non-linear dependency of heavy metal bioavailability on the total metal content should be taken into account.

Keywords : PM_{2.5}, Heavy metals, Bioavailability, Mineral allocation, Rome urban area

Numerical air quality forecasting over eastern China: An operational application of WRF-Chem

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Source: Volume 153, March 2017, Pages 94-108

<https://doi.org/10.1016/j.atmosenv.2017.01.020>

The Regional Atmospheric Environmental Modeling System for eastern China (RAEMS) is an operational numerical system to forecast near surface atmospheric pollutants such as PM_{2.5} and O₃ over the eastern China region. This system was based on the fully online coupled weather research and forecasting/chemistry (WRF-Chem) model. Anthropogenic emissions were based on the multi-resolution emission inventory for China (MEIC), and biogenic emissions were online calculated using model of emissions of gases and aerosols from nature (MEGAN2). Authorized by the China Meteorological Administration (CMA), this system started to provide operational forecast in 2013. With a large domain covering eastern China, the system produces daily 72-hr forecast. In this work, a comprehensive evaluation was carried out against measurements for two full years (2014–2015). Evaluation results show that the RAEMS is skillful in forecasting temporal variation and spatial distribution of major air pollutants over the eastern China region. The performance is consistent in different forecast length of 24 h, 48 h, and 72 h. About half of cities have correlation coefficients greater than 0.6 for PM_{2.5} and 0.7 for daily maximum 8-h averaged (DM8H) ozone. The forecasted PM_{2.5} is generally in good agreement with observed concentrations, with most cities having normalized mean biases (NMB) within $\pm 25\%$. Forecasted ozone diurnal variation is very similar to that of observed, and makes small peak time error for DM8H ozone. It also shows good capability in capturing ozone pollution as indicated by high critical success indexes (CSI). The modeling system also exhibits acceptable performance for PM₁₀, NO₂, SO₂, and CO. Meanwhile,

degraded performance for PM_{2.5} is found under heavy polluted conditions, and there is a general over estimation in ozone concentrations.

Keywords : Air quality modeling, Air quality forecasting, WRF-Chem, PM_{2.5}, Ozone

Air pollutants and toxic emissions of various mileage motorcycles for ECE driving cycles

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Source: Volume 153, March 2017, Pages 126-134

<https://doi.org/10.1016/j.atmosenv.2017.01.019>

Motorcycles were selected to determine their fuel consumption and exhaust emissions following ECE driving cycles. Exhaust constituents including CO₂, CO, NO_x, total hydrocarbons (THC) and hydrocarbon species (27 paraffins, 9 olefins, 16 aromatics and 15 carbonyls) were investigated for this work. The age of 10– 90% of the selected motorcycles ranged from 2.5 to 12.4 years, and their mileage ranged from 5400 to 39,300 km. CO emission ranged from 1.4 to 6.4 g/km (median value: 2.98 g/km), THC from 0.41 to 1.54 g/km (median value: 0.98 g/km), NO_x from 0.16 to 0.28 g/km (median value: 0.21 g/km), CO₂ from 58.9 to 62.2 g/km (median value: 60.5 g/km) and fuel consumption from 30.7 to 36.4 km/L (median value: 33.4 km/L), corresponding to the percentage cumulative data from 10 to 90% of the selected motorcycles. Results indicated that the motorcycle exhaust emission and fuel consumption depended on their mileage and ages. An increase in mileage of 1000 km resulted in an increase of 103 mg for CO emission and 14.7 mg for hydrocarbon emission and a reduction of 1.52 mg NO_x emission and 0.11 km per liter fuel consumption. For various VOC groups, a mileage increase of 1000 km corresponding to the increased exhaust emission of paraffins was 6.71 mg, olefins 1.90 mg, aromatics 7.04 mg, carbonyls 0.283 mg and 67 VOC species 15.9 mg. Fuel consumption and emissions of CO and hydrocarbon increased in motorcycles over the guaranteed mileage of 15,000 km.

Keywords : Volatile organic compounds (VOCs), Mileage, Four-stroke motorcycle

Uncertainty and dynamics of natural wetland CH₄ release in China: Research status and priorities

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Source: Volume 154, April 2017, Pages 95-105

<https://doi.org/10.1016/j.atmosenv.2017.01.038>

Natural wetlands represent the largest single source of methane (CH₄), a potent greenhouse gas. China is home to the world's fourth largest wetland area, and it is facing intense climate- and human-related impacts. The scientific community in China has invested considerable effort into investigating wetland CH₄ release and its dynamics. Static chamber and eddy covariance observations have verified the temperature, water regime and air pressure as factors that regulate the diurnal and seasonal variation of CH₄ release. Non-growing seasons, especially freezing-thawing cycles, play a role in CH₄ release. However, a knowledge gap still exists with respect to the inter-annual variability of CH₄ release. Observations also suggest that water and temperature regimes control the micro- and macro-scale spatial pattern of CH₄ release, respectively. Recent bookkeeping surveys, biogeochemical model simulations, and chemical transport model inversions, have narrowed the uncertainty range of national CH₄ release to 2.46–3.20, 2.77–4.95 and 2.38–4.91 Tg CH₄ yr⁻¹, respectively. Wetland loss (especially cropland conversion in Northeast China), despite climate changes, decreased CH₄ release by 45.2%–52.2% from the 1950s–2000s, and by 13.2%–15.4% from the 1980s–2000s. However, future warmer temperatures and rising CO₂ are predicted to strengthen national CH₄ release by 32% (RCP2.6), 55% (RCP4.5) and 91% (RCP8.5) by the 2080s, albeit without the variation in wetland extent having been considered. Furthermore, future research should emphasize the mechanisms involved in CH₄ release during freezing-thawing cycles and interannual variability. Model–data fusion of eddy covariance and manipulative experiments, especially warming and CO₂ enrichment, would benefit estimations and projections of CH₄ release.

Keywords : Methane (CH₄), Natural wetland, Climate change, Land use change, China

Variation of the vertical distribution of Nabro volcano aerosol layers in the stratosphere observed by LIDAR

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Source: Volume 154, April 2017, Pages 1-8

<https://doi.org/10.1016/j.atmosenv.2017.01.033>

We present results of the vertical distribution variation of volcanic aerosol layers in the upper troposphere and lower stratosphere. The data were taken with our multiwavelength aerosol Raman lidar at Gwangju (35.10° N, 126.53° E), Korea. The volcanic ash particles and gases were released around 12 June 2011 during the eruption of the Nabro volcano (13.37° N, 41.7° E) in Eritrea, east Africa. Forward trajectory computations show that the volcanic aerosols were advected from North Africa to East Asia. The first measurement of the aerosol layer over Korea was on 19 June 2011. The aerosol layers appeared between 15 km and 17 km height asl (above sea level). The maximum value of the aerosol layer of the particle backscatter coefficient ($1.5 \pm 0.3 \text{ Mm}^{-1} \text{ sr}^{-1}$) and the linear particle depolarization ratio at 532 nm (2.2%) were observed at 16.4 km height asl. We continuously probed the upper troposphere and lower stratosphere for this volcanic aerosol layer during the following 6 months, until December 2011. The volcanic aerosol layer showed a single-peak of the particle backscatter coefficient and a comparably narrow

vertical thickness at our observation site at the beginning of our observation period (i.e. comparably soon after the initial eruption period). After that initial period the vertical distribution of the plume changed. Multiple peaks and a comparably broad geometrical thickness developed with progressing observation time. The vertical thickness of the volcanic aerosol layer expanded up to 10 km by 3 August 2011. The linear particle depolarization ratios were larger in the lower part of the aerosol layer than the upper part of the aerosol layer. We observed a strong variation of the AOD (aerosol optical depth) in the first two months of our lidar observations. After these two months the AOD gradually decreased with time from September to December 2011 and the maximum particle backscatter coefficients consistently decreased. The corresponding e-folding decay time of the layer AOD was 117 days.

Keywords : Lidar, Volcanic aerosol, Nabro volcano, Depolarization ratio, Stratospheric aerosol

Assessment of emissions of greenhouse gases and air pollutants in Indonesia and impacts of national policy for elimination of kerosene use in cooking

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Source: Volume 154, April 2017, Pages 82-94
<https://doi.org/10.1016/j.atmosenv.2017.01.041>

This study presents an emission inventory (EI) for major anthropogenic sources of Indonesia in 2007 and 2010. The EI was developed using a combination of top-down and bottom-up approaches with comprehensive activity data collected at the provincial/district level to produce spatially and temporally distributed emission of toxic pollutants and greenhouse gases (GHGs). The sources were categorized into: 1) fuel combustion in power plant, 2) industry, 3) transportation, 4) residential and commercial combustion, 5) biomass open burning, and 6) non-combustion agricultural activity and waste disposal. The best estimates of the 2010 national emissions, in Gg, of toxic pollutants were: 1014 SO₂; 3323 NO_x; 24,849 CO; 4077 NMVOC; 1276 NH₃; 2154 PM₁₀; 1728 PM_{2.5}; 246 BC; 718 OC; and GHGs: 540,275 CO₂; 3979 CH₄ and 180 N₂O. During the period from 2007 to 2010, the national emissions increased by 0.7–8.8% (0.23–2.8% per year), varied with species, with the most significant changes obtained for the biomass open burning emissions. For 2010 results, the low and high emission estimates for different species were ranging from –58% to +122% of the corresponding best estimates. The largest range (high uncertainty) was for BC due to the wide range of the limitedly available emission factors. Spatially, higher emission intensity was seen in large urban areas of Java and Sumatra Islands. Temporally, dry months of August–October had higher emissions. During the first 3 years (2007–2010) of implementation, the national policy of elimination of kerosene use in cooking had successfully replaced 4.9 Tg kerosene with 2.6 Tg LPG in 30 designated provinces. The net emission reductions of different species ranged from 48 Mg (SO₂) to 7.6 Tg for CO₂. The global warming potential weighted emissions from the residential cooking alone, collectively for GHGs and short-lived climate pollutants in 20-yr CO₂ eq., would reduce by 2%. More significant reductions in the residential combustion emissions are expected if the solid cooking fuel could be targeted in future fuel conversion programs. The benefits to human health

resulted from the emission reduction of toxic pollutants from residential cooking could be substantial and should be assessed in future studies.

Keywords : Emission inventory, Air pollution, Kerosene fuel switching, Climate co-benefit, Indonesia

Particulate matter air pollution in Europe in a +2 °C warming world

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Source: Volume 154, April 2017, Pages 129-140

<https://doi.org/10.1016/j.atmosenv.2017.01.037>

In the framework of the IMPACT2C project, we have evaluated the future European particulate matter concentrations under the influence of climate change and anthropogenic emission reductions. To do so, 30-year simulations for present and future scenarios were performed with an ensemble of four regional Chemical Transport Models. +2 °C scenarios were issued from different regional climate simulations belonging to the CORDEX experiment (RCP4.5 scenario). Comparing present day simulations to observations shows that these simulations meet the requested quality criteria even if some biases do exist. Also, we showed that using regional climate models instead of meteorological reanalysis was not critical for the quality of our simulations. Present day as well as future scenarios show the large variability between models associated with different meteorology and process parameterizations. Future projections of PM concentrations show a large reduction of PM₁₀ and PM_{2.5} concentrations in a +2 °C climate over the European continent (especially over Benelux), which can be mostly attributed to emission reduction policies. Under a current legislation scenario, annual PM₁₀ could be reduced by between 1.8 and 2.9 µg m⁻³ (14.1–20.4%). If maximum technologically feasible emission reductions were implemented, further reductions of 1.4–1.9 µg m⁻³ (18.6–20.9%) are highlighted. Changes due to a +2 °C warming, in isolation from emission changes, are in general much weaker (–1.1 to +0.4 µg m⁻³, –0.3 to +5.1% for annual PM₁₀ averaged over the European domain). Even if large differences exist between models, we have determined that the decrease of PM over Europe associated with emission reduction is a robust result. The patterns of PM changes resulting from climate change (for example the increase of PM over Spain and southern France and the decrease of PM₁₀ over eastern Europe) are also robustly predicted even if its amplitude remains weak compared to changes associated with emission reductions.

Keywords : Air quality, Chemical transport models, Particulate matter, Climate change, IMPACT2C project

Variations in particulate matter over Indo-Gangetic Plains and Indo-Himalayan Range during four field campaigns in winter monsoon and summer monsoon: Role of pollution pathways

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Source: Volume 154, April 2017, Pages 200-224

<https://doi.org/10.1016/j.atmosenv.2016.12.054>

Both in-situ and space-borne observations reveal an extremely high loading of particulates over the Indo-Gangetic Plains (IGP), all year around. With a burgeoning population and combustion sources (fossil fuels (FFs) and biofuels (BFs)) in close proximity to each other, the IGP is widely regarded as a hotspot for anthropogenic aerosol emission in South Asia. The deteriorating air quality over this region, particularly during winters, is a cause of major concern, since the pollutants undergo long range transport from their source regions to the Indo-Himalayan Range (IHR), Bay of Bengal (BoB) and other remote areas, polluting their pristine atmospheric conditions. Seasonal reversal in winds over the Indian mainland leads to an outflow of continental pollutants into the BoB during winters and a net advection of desert dust aerosols into the IGP from southwest Asia (SW-Asia), northwest India (NW-India) and northern Africa (N-Africa) during summers. Through the course of this study, four observational campaigns were conducted for sampling the ambient PM_{2.5} and PM₁₀ during winter and summer seasons of 2014–2015, at multiple locations (18 sites) in the IGP, IHR, and semi-arid/arid sites towards their south and west, in order to accurately determine the inter-seasonal and inter-annual changes in the aerosol loading at the sites. We have also utilized data from Moderate Resolution Imaging Spectroradiometer (MODIS) on-board Earth Observing System (EOS) Terra satellite for estimating the columnar Aerosol Optical Depth at 550 nm (AOD₅₅₀) and data from EOS Terra and Aqua satellites for discovering openly burning fires in the vicinity of sampling sites. Determination of the major source regions and key transport pathways during both seasons have also been attempted, using back-trajectory cluster analyses, as well as receptor models such as PSCF and CWT.

Keywords : PM_{2.5}, PM₁₀, Indo-Gangetic plains, Indo-Himalayan range, Long-range transport, Cluster analysis, PSCF, CWT

Greenhouse gases concentrations and fluxes from subtropical small reservoirs in relation with watershed urbanization

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Source: Volume 154, April 2017, Pages 225-235
<https://doi.org/10.1016/j.atmosenv.2017.01.047>

Greenhouse gas (GHG) emissions from reservoirs and global urbanization have gained widespread attention, yet the response of GHG emissions to the watershed urbanization is poorly understood. Meanwhile, there are millions of small reservoirs worldwide that receive and accumulate high loads of anthropogenic carbon and nitrogen due to watershed urbanization and can therefore be hotspots of GHG emissions. In this study, we assessed the GHG concentrations and fluxes in sixteen small reservoirs draining urban, agricultural and forested watersheds over a period of one year. The concentrations of $p\text{CO}_2$, CH_4 and N_2O in sampled urban reservoirs that received more sewage input were higher than those in agricultural reservoirs, and were 3, 7 and 10 times higher than those in reservoirs draining in forested areas, respectively. Accordingly, urban reservoirs had the highest estimated GHG flux rate. Regression analysis indicated that dissolved total phosphorus, dissolved organic carbon (DOC) and chlorophyll-*a* (*Chl-a*) had great effect on CO_2 production, while the nitrogen (N) and phosphorus (P) content of surface water were closely related to CH_4 and N_2O production. Therefore, these parameters can act as good predictors of GHG emissions in urban watersheds. Given the rapid progress of global urbanization, small urban reservoirs play a crucial role in accounting for regional GHG emissions and cannot be ignored.

Keywords : GHG, Urbanization, Land use, Small reservoirs, Spatiotemporal variation, Potential controls

Emission factors and characteristics of ammonia, hydrogen sulfide, carbon dioxide, and particulate matter at two high-rise layer hen houses

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Source: Volume 154, April 2017, Pages 260-273
<https://doi.org/10.1016/j.atmosenv.2017.01.050>

Air pollutants emitted from confined animal buildings can cause environmental pollution and ecological damage. Long-term (>6 months) and continuous (or high frequency) monitoring that can reveal seasonal and diurnal variations is needed to obtain emission factors and characteristics about these pollutants. A two-year continuous monitoring of ammonia (NH_3), hydrogen sulfide (H_2S), carbon dioxide (CO_2) and particulate matter (PM_{10}) emissions from two 218,000-hen high-rise layer houses (H-A and H-B) in Indiana, USA was conducted from June 2007 to May 2009. Gaseous pollutant concentrations were measured with two gas analyzers and PM_{10} concentrations were measured with three Tapered Element Oscillating Microbalances. The operation and performance of ventilation fans were continuously monitored with multiple methods. Only the emission rates calculated with valid data days (days with more than 18 h, or 75%, of valid data) are reported in this paper. The two-house and two-year mean \pm standard deviation emissions per day per hen for NH_3 , H_2S , CO_2 , and PM_{10} were 1.08 ± 0.42 g, 1.37 ± 0.83 mg, 76.7 ± 14.6 g, and

20.6 ± 22.5 mg, respectively. Seasonal emission variations were demonstrated for NH₃ and CO₂, but not evident for H₂S and PM₁₀. Ammonia and CO₂ emissions were higher in winter than in summer. Significant daily mean emission variations were observed for all four pollutants between the two houses ($P < 0.05$), and between the two years from the same house ($P < 0.01$) except for CO₂ at one house. Carbon dioxide originated from manure decomposition was >9% of that from bird respiration. Emissions of CO₂ during molting were about 80% of those during normal egg production days. Emissions of H₂S were not a major concern due to their very low quantities. Emissions of PM₁₀ were more variable than other pollutants. However, not all of the emission statistics are explainable.

Keywords : Agricultural air quality, Animal agriculture, Air pollution, Emission baseline, Pollutant emission, Poultry house

Reduction of air pollution levels downwind of a road with an upwind noise barrier

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Source: Volume 155, April 2017, Pages 1-10
<https://doi.org/10.1016/j.atmosenv.2017.02.001>

We propose a dispersion model to estimate the impact of a solid noise barrier upwind of a highway on air pollution concentrations downwind of the road. The model, based on data from wind tunnel experiments conducted by Heist et al. (2009), assumes that the upwind barrier has two main effects: 1) it creates a recirculation zone behind the barrier that sweeps the emissions from the highway back towards the wall, and 2) it enhances vertical dispersion and initial mixing. By combining the upwind barrier model with the mixed wake model for a downwind barrier described in Schulte et al. (2014), we are able to model dispersion of emissions from a highway with noise barriers on both sides. The model provides a good description of measurements made in the wind tunnel. The presence of an upwind barrier causes reductions in concentrations relative to those measured downwind of a road with no barriers. The reduction can be as large as that caused by a downwind barrier if the recirculation zone covers the width of the highway. Barriers on both sides of the highway result in larger reductions downwind of the barriers than those caused by a single barrier either upwind or downwind. As expected, barrier effects are small beyond 10 barrier heights downwind of the highway. We also propose a tentative model to estimate on-road concentrations within the recirculation zone induced by the upwind barrier.

Keywords : Dispersion modeling, Noise barrier, Near-road concentration, Air pollution, Recirculation zone

Characterising an intense PM pollution episode in March 2015 in France from multi-site approach and near real time data: Climatology, variabilities, geographical origins and model evaluation

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Source: Volume 155, April 2017, Pages 68-84
<https://doi.org/10.1016/j.atmosenv.2017.02.012>

During March 2015, a severe and large-scale particulate matter (PM) pollution episode occurred in France. Measurements in near real-time of the major chemical composition at four different urban background sites across the country (Paris, Creil, Metz and Lyon) allowed the investigation of spatiotemporal variabilities during this episode. A climatology approach showed that all sites experienced clear unusual rain shortage, a pattern that is also found on a longer timescale, highlighting the role of synoptic conditions over Western-Europe. This episode is characterized by a strong predominance of secondary pollution, and more particularly of ammonium nitrate, which accounted for more than 50% of submicron aerosols at all sites during the most intense period of the episode. Pollution advection is illustrated by similar variabilities in Paris and Creil (distant of around 100 km), as well as trajectory analyses applied on nitrate and sulphate. Local sources, especially wood burning, are however found to contribute to local/regional sub-episodes, notably in Metz. Finally, simulated concentrations from Chemistry-Transport model CHIMERE were compared to observed ones. Results highlighted different patterns depending on the chemical components and the measuring site, reinforcing the need of such exercises over other pollution episodes and sites.

Keywords : Aerosol, Pollution, France, Aerosol chemical speciation monitor, ACSM, Aethalometer, Model

Composition and mixing states of brown haze particle over the Himalayas along two transboundary south-north transects

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Source: Volume 156, May 2017, Pages 24-35
<https://doi.org/10.1016/j.atmosenv.2017.02.029>

Pollutants that are usually transported from southern Asia to the Tibetan Plateau deposit on the Plateau surface, change snow albedo and thereby surface radiative flux. This results numerous climatic implications like as erratic monsoon, perturbation in hydrological cycle, etc. However, the accurate estimation of these climatic implications is not well understood, because the atmospheric pollution is a heterogeneous mixture of various particle types. Therefore, this part of climate

research requires a detailed investigation of physical and chemical properties of atmospheric pollutants. This study aimed to examine the physical and chemical properties of atmospheric pollutants across the Himalayan regions along two transboundary south-north transects. The information of individual-particles was obtained using microscopy-based techniques that comprises transmission electron microscope (TEM) and Energy-dispersive X-ray spectrometer (EDX). Study capture the signatures of various types of atmospheric species such as black carbon (BC), mineral dust, fly ash, organic matter, sulfate, nitrite, ammonium, and NaCl. Microscopy-based techniques confirm that these particles were generally in mixing state, for example salt-coated particles accounting for 25–56% of the total particles in sampled locations. Our analysis shows that urban and rural locations are characterized with atmospheric particles which sourced from anthropogenic activities, whereas remote locations with those released from natural crustal. However, the relative contributions of anthropogenic particles were higher than that of particles released from natural crustal. The presence of such particles over remote locations of Himalayan region provides an evidence of prevailing atmospheric transport processes, which further need to be well understood. It is expected that this work would be helpful in understanding the regional atmospheric conditions and the transboundary transport process of haze particles. As these informations are of great importance in modeling studies, which further lead to improve understanding of haze particles climate effects.

Keywords : Brown haze, TEM-EDX, Mixing states, Himalayas, Tibetan Plateau

Organic aerosols over Indo-Gangetic Plain: Sources, distributions and climatic implications

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Source: Volume 157, May 2017, Pages 59-74
<https://doi.org/10.1016/j.atmosenv.2017.03.008>

Organic aerosol (OA) constitutes a dominant fraction of airborne particulates over Indo-Gangetic Plain (IGP) especially during post-monsoon and winter. Its exposure has been associated with adverse health effects while there are evidences of its interference with Earth's radiation balance and cloud condensation (CC), resulting possible alteration of hydrological cycle. Therefore, presence and effects of OA directly link it with food security and thereby, sustainability issues. In these contexts, atmospheric chemistry involving formation, volatility and aging of primary OA (POA) and secondary OA (SOA) have been reviewed with specific reference to IGP. Systematic reviews on science of OA sources, evolution and climate perturbations are presented with databases collected from 82 publications available throughout IGP till 2016. Both gaseous and aqueous phase chemical reactions were studied in terms of their potential to form SOA. Efforts were made to recognize the regional variation of OA, its chemical constituents and sources throughout IGP and inferences were made on its possible impacts on regional air quality. Mass fractions of OA to airborne particulate showed spatial variation likewise in Lahore (37 and 44% in fine and coarse fractions, respectively), Patiala (28 and 37%), Delhi (25 and 38%), Kanpur (24 and 30%), Kolkata (11 and 21%) and Dhaka. Source apportionment studies indicate biomass burning, coal combustion and vehicular emissions as predominant OA sources. However, sources represent

considerable seasonal variations with dominance of gasoline and diesel emissions during summer and coal and biomass based emissions during winter and post-monsoon. Crop residue burning over upper-IGP was also frequently held responsible for massive OA emission, mostly characterized by its hygroscopic nature, thus having potential to act as CC nuclei. Conclusively, climatic implication of particulate bound OA has been discussed in terms of its interaction with radiation balance.

Keywords : Organic aerosol, Biomass, Climate change, CCN, PAHs, Indo-Gangetic plain, Trans-boundary

Effectiveness of temporary control measures for lowering PM_{2.5} pollution in Beijing and the implications

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Source: Volume 157, May 2017, Pages 75-83
<https://doi.org/10.1016/j.atmosenv.2017.03.017>

In order to investigate the effects of the temporary strengthening of air quality assurance controlling measures during the Beijing 2015 IAAF World Championships and the Military Parade Assurance Period (MPAP) in China, we collected daily PM_{2.5} aerosol samples at three typical sites (urban downtown, suburban and rural background area, respectively) in Beijing and investigated the variations of concentration of the water-soluble ions, elemental constituents, organic carbon (OC) and elemental carbon (EC) in PM_{2.5} from Aug.15 to Sept.10, 2015. Simultaneously, 1-h high-resolution continuous monitoring results of PM_{2.5} mass concentration as well as the chemical components which were measured at another online monitoring urban site were incorporated. The concentrations of PM_{2.5} and other gaseous pollutants (SO₂, NO₂ and CO) during the parade control period (Aug.20-Sept.3) exhibited a substantially decrease compared with the concentrations during both the non-control (August 15 to August 19 and September 4 to September 10) period and the same period in 2014. According to the CMC results, the major components were identified as secondary inorganic aerosol (SIA, the combination of sulfate, ammonium and nitrate), mineral dust and particular organic matter (POM), which together accounted for more than 80% of PM_{2.5} in urban and suburban sites. POM is found to account for the largest proportion, and the obviously higher proportion of POM in the urban area revealed the significance contribution from vehicles. Compared with the non-control period, the mass concentrations of SIA and secondary organic carbon (SOC) decreased obviously. However, SIA and SOC are observed to play an important role in contributing to the rapid growth process of PM_{2.5} under unfavorable meteorological conditions during the control period. In view of the gradual improvement of air quality in Beijing, as well as the contribution of secondary aerosol formations in total PM_{2.5}, effective control of primary gaseous pollutants and volatile organic compounds (VOCs) will be very significant for further lowering the concentration of PM_{2.5} in Beijing in normal time.

Keywords : Parade blue, Chemical composition, Positive matrix factorization, Chemical mass closure, Secondary aerosols, Enhanced control measures

Exposure to fine particulate, black carbon, and particle number concentration in transportation microenvironments

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Source: Volume 157, May 2017, Pages 135-145

<https://doi.org/10.1016/j.atmosenv.2017.03.006>

This research determined intake dose of fine particulate matter (PM_{2.5}), equivalent black carbon (eBC), and number of sub-micron particles (Np) for commuters in Bogotá, Colombia. Doses were estimated through measurements of exposure concentration, a surrogate of physical activity, as well as travel times and speeds. Impacts of travel mode, traffic load, and street configuration on dose and exposure were explored. Three road segments were selected because of their different traffic loads and composition, and dissimilar street configuration. The transport modes considered include active modes (walking and cycling) and motorized modes (bus, car, taxi, and motorcycle). Measurements were performed simultaneously in the available modes at each road segment. High average eBC concentrations were observed throughout the campaign, ranging from 20 to 120 μgm^{-3} . Commuters in motorized modes experienced significantly higher exposure concentrations than pedestrians and bicyclists. The highest average concentrations of PM_{2.5}, eBC, and Np were measured inside the city's Bus Rapid Transit (BRT) system vehicles. Pedestrians and bicycle users in an open street configuration were exposed to the lowest average concentrations of PM_{2.5} and eBC, six times lower than those experienced by commuters using the BRT in the same street segment. Pedestrians experienced the highest particulate matter intake dose in the road segments studied, despite being exposed to lower concentrations than commuters in motorized modes. Average potential dose of PM_{2.5} and eBC per unit length traveled were nearly three times higher for pedestrians in a street canyon configuration compared to commuters in public transport. Slower travel speed and elevated inhalation rates dominate PM dose for pedestrians. The presence of dedicated bike lanes on sidewalks has a significant impact on reducing the exposure concentration for bicyclists compared to those riding in mixed traffic lanes. This study proposes a simple method to perform loading effect correction for measurements of black carbon using multiple portable aethalometers.

Keywords : Fine particulate, Black carbon, Pollution in urban microenvironments in latin-America, Ultrafine particles, Micro-aethalometer loading correction

Daily ambient air pollution metrics for five cities: Evaluation of data-fusion-based estimates and uncertainties

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Source: Volume 158, June 2017, Pages 36-50

<https://doi.org/10.1016/j.atmosenv.2017.03.022>

Spatiotemporal characterization of ambient air pollutant concentrations is increasingly relying on the combination of observations and air quality models to provide well-constrained, spatially and temporally complete pollutant concentration fields. Air quality models, in particular, are attractive, as they characterize the emissions, meteorological, and physiochemical process linkages explicitly while providing continuous spatial structure. However, such modeling is computationally intensive and has biases. The limitations of spatially sparse and temporally incomplete observations can be overcome by blending the data with estimates from a physically and chemically coherent model, driven by emissions and meteorological inputs. We recently developed a data fusion method that blends ambient ground observations and chemical-transport-modeled (CTM) data to estimate daily, spatially resolved pollutant concentrations and associated correlations. In this study, we assess the ability of the data fusion method to produce daily metrics (i.e., 1-hr max, 8-hr max, and 24-hr average) of ambient air pollution that capture spatiotemporal air pollution trends for 12 pollutants (CO, NO₂, NO_x, O₃, SO₂, PM₁₀, PM_{2.5}, and five PM_{2.5} components) across five metropolitan areas (Atlanta, Birmingham, Dallas, Pittsburgh, and St. Louis), from 2002 to 2008.

Three sets of comparisons are performed: (1) the CTM concentrations are evaluated for each pollutant and metropolitan domain, (2) the data fusion concentrations are compared with the monitor data, (3) a comprehensive cross-validation analysis against observed data evaluates the quality of the data fusion model simulations across multiple metropolitan domains. The resulting daily spatial field estimates of air pollutant concentrations and uncertainties are not only consistent with observations, emissions, and meteorology, but substantially improve CTM-derived results for nearly all pollutants and all cities, with the exception of NO₂ for Birmingham. The greatest improvements occur for O₃ and PM_{2.5}. Squared spatiotemporal correlation coefficients range between simulations and observations determined using cross-validation across all cities for air pollutants of secondary and mixed origins are $R^2 = 0.88\text{--}0.93$ (O₃), $0.81\text{--}0.89$ (SO₄), $0.67\text{--}0.83$ (PM_{2.5}), $0.52\text{--}0.72$ (NO₃), $0.43\text{--}0.80$ (NH₄), $0.32\text{--}0.51$ (OC), and $0.14\text{--}0.71$ (PM₁₀).

Results for relatively homogeneous pollutants of secondary origin, tend to be better than those for more spatially heterogeneous (larger spatial gradients) pollutants of primary origin (NO_x, CO, SO₂ and EC). Generally, background concentrations and spatial concentration gradients reflect interurban airshed complexity and the effects of regional transport, whereas daily spatial pattern variability shows intra-urban consistency in the fused data. With sufficiently high CTM spatial resolution, traffic-related pollutants exhibit gradual concentration gradients that peak toward the urban centers. Ambient pollutant concentration uncertainty estimates for the fused data are both more accurate and smaller than those for either the observations or the model simulations alone.

Keywords : Air quality, Spatiotemporal, Fusion, Blending, CMAQ

Role of organic aerosols in CCN activation and closure over a rural background site in Western Ghats, India

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Source: Volume 158, June 2017, Pages 148-159

<https://doi.org/10.1016/j.atmosenv.2017.03.037>

The cloud condensation nuclei (CCN) closure study was performed to exemplify the effect of aerosol chemical composition on the CCN activity of aerosols at Mahabaleshwar, a high altitude background site in the Western Ghats, India. For this, collocated aerosol, CCN, Elemental Carbon (EC), Organic Carbon (OC), sub-micron aerosol chemical speciation for the period from 3rd June to 19th June 2015 was used. The chemical composition of non-refractory particulate matter ($<1\ \mu\text{m}$) as measured by Time of Flight – Aerosol Chemical Speciation Monitor (ToF-ACSM) was dominated by organics with average concentration of 3.81 ± 1.6 , 0.32 ± 0.06 , 0.15 ± 0.02 , 0.13 ± 0.03 and $0.95 \pm 0.12\ \mu\text{g m}^{-3}$ for organics, ammonium, chloride, nitrate and sulphate, respectively. The PM1 number concentration as obtained by Wide Range Aerosol Spectrometer (WRAS) varied from 750 to 6480 cm^{-3} . The average mass concentration of elemental carbon (EC) as measured by OC-EC analyzer was $1.16 \pm 0.4\ \mu\text{g m}^{-3}$. The average CCN concentrations obtained from CCN counter (CCNC) at five super-saturations (SS's) was $118 \pm 58\ \text{cm}^{-3}$ (0.1% SS), $873 \pm 448\ \text{cm}^{-3}$ (0.31% SS), $1308 \pm 603\ \text{cm}^{-3}$ (0.52% SS), $1610 \pm 838\ \text{cm}^{-3}$ (0.73% SS) and $1826 \pm 985\ \text{cm}^{-3}$ (0.94% SS). The CCN concentrations were predicted using Köhler theory on the basis of measured aerosol particle number size distribution, size independent NR-PM1 chemical composition and calculated hygroscopicity. The CCN closure study was evaluated for 3 scenarios, B-I (all soluble inorganics), B-IO (all soluble organics and inorganics) and B-IOOA (all soluble inorganic and soluble oxygenated organic aerosol, OOA). OOA component was derived from the positive matrix factorization (PMF) analysis of organic aerosol mass spectra. Considering the bulk composition as internal mixture, CCN closure study was underestimated by 16–39% for B-I and overestimated by 47–62% for B-IO. The CCN closure result was appreciably improved for B-IOOA where the knowledge of OOA fraction was introduced and uncertainty reduced to within 8–10%.

Keywords : ACSM, NR-PM1, PMF, OOA, Köhler theory, CCN closure

The contribution of residential coal combustion to PM_{2.5} pollution over China's Beijing-Tianjin-Hebei region in winter

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Source: Volume 159, June 2017, Pages 147-161

<https://doi.org/10.1016/j.atmosenv.2017.03.054>

The Beijing-Tianjin-Hebei (BTH) region experiences severe haze episodes throughout the year, and especially during the winter heating season. Residential combustion of coal has increasing been cited as a possible source for the PM_{2.5} pollution that causes the haze episodes. To investigate these claims, a WRF-CMAQ system is used to reproduce the regional haze episodes observed during December 2015. The contribution of residential coal combustion to PM_{2.5} concentrations in the BTH region is quantified using the Brute Force approach. Across the region, residential coal combustion contributed 46% of the monthly averaged PM_{2.5} concentration (3% each from Beijing and Tianjin and 40% from Hebei Province). During the haze episodes, the contribution varied between 30 and 57%. At the city scale, the contribution ranged from 22 to 58% averaged across the month and 15–65% during the haze episodes. Langfang was the city that was the most affected by residential coal combustion in the BTH region. The large contribution to air pollution in Tianjin and Beijing from households in Hebei Province suggests that regional control measures are required.

Keywords : Beijing-Tianjin-Hebei region, PM2.5, Residential coal combustion, Urban air pollution, WRF-CMAQ, Air quality modeling

Human health risk due to variations in PM10-PM2.5 and associated PAHs levels

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Source: Volume 160, July 2017, Pages 27-35
<https://doi.org/10.1016/j.atmosenv.2017.04.004>

WHO (2012) reports that chronic exposure to air pollutants, including particulate matter (PM), causes the death of 7 million people, constituting the most important environmental risk for health in the world. IARC classifies contaminated outdoor air as carcinogenic, Group 1 category. However, in our countries there are few studies regarding air pollution levels and possible associated effects on public health.

The current study determined PM and associated polycyclic aromatic hydrocarbons (PAHs) levels in outdoor air, identified their possible emission sources and analysed health risks in the city of Tandil (Argentina). PM10 and PM2.5 samples were collected using a low volume sampler (MiniVol TAS) in three areas: city centre, industrial and residential. Concentrations were determined by gravimetric methods and the content of the US EPA 16 priority PAHs was found by high performance liquid chromatography (HPLC). Description of the main emission sources and selection of monitoring sites resulted from spatial analysis and the IVE (International Vehicle Emissions) model was used in the characterisation of the traffic flow. Median values of 35.7 μgm^{-3} and 9.6 μgm^{-3} in PM10 and PM2.5 respectively and characteristic profiles were found for each area. Local values PAHs associated to PM10 and PM2.5, in general, were lower than 10 ngm^{-3} . The estimated Unit Risk for the three areas exceeds US EPA standards (9×10^{-5}). The number of deaths attributable to short term exposure to outdoor PM10 was 4 cases in children under 5 years of age, and 21 cases in total population, for a relative risk of 1.037.

Keywords : Air quality, PM, PM-PAHs, Environmental burden of disease, Unit Risk, Tandil Argentina

Atmospheric oxidation of selected chlorinated alkenes by O3, OH, NO3 and Cl

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Source: Volume 170, December 2017, Pages 12-21
<https://doi.org/10.1016/j.atmosenv.2017.09.031>

An experimental study on the 3-chloro-2-methyl-1-propene (CMP), 2,3-dichloropropene (DCP) and 3,4-dichlorobutene (DCB) reactions with atmospheric oxidants at (298 ± 1) K and atmospheric pressure is reported. Rate constants for the gas phase reactions of the three chlorinated alkenes with O₃, OH and NO₃ radicals and Cl atom were determined in a 100 L Teflon reactor by gas chromatography with flame ionization detector (GC-FID). The obtained rate constants are $(3.03 \pm 0.15) \times 10^{-18}$, $(3.83 \pm 1.30) \times 10^{-11}$, $(1.99 \pm 0.19) \times 10^{-14}$, and $(2.40 \pm 0.41) \times 10^{-10}$ cm³ molecule⁻¹ s⁻¹ for CMP reactions with O₃, OH, NO₃, and Cl, respectively, $(4.62 \pm 1.41) \times 10^{-20}$, $(1.37 \pm 1.02) \times 10^{-11}$, $(1.45 \pm 0.15) \times 10^{-15}$ and $(1.30 \pm 0.99) \times 10^{-11}$ cm³ molecule⁻¹ s⁻¹ for DCP reactions and $(2.09 \pm 0.24) \times 10^{-19}$, $(1.45 \pm 0.59) \times 10^{-11}$, $(3.00 \pm 0.82) \times 10^{-16}$ and $(1.91 \pm 0.19) \times 10^{-10}$ cm³ molecule⁻¹ s⁻¹ for DCB reactions. The CMP reaction products were detected and possible reaction mechanisms of their formation were proposed. Chloroacetone was found to be the major product in all four oxidation reactions. The loss process of CMP in the atmosphere is mostly controlled by its reaction with the OH radical during daytime and with NO₃ during nighttime, with lifetimes of 3.6 h and 27.9 h respectively. Atmospheric implications of both these reactions and their potential products are discussed.

Keywords :Chlorinated alkene, Rate constant , Reaction mechanism , Kinetics, Gas chromatography

Environmental Science & Pollution Research

Variations of polycyclic aromatic hydrocarbons in ambient air during haze and non-haze episodes in warm seasons in Hangzhou, China

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Source: January 2017, Volume 24, Issue 1, pp 135–145

To investigate the characteristics of polycyclic aromatic hydrocarbons (PAHs) during haze episodes in warm seasons, daily PM_{2.5} and gaseous samples were collected from March to September 2015 in Hangzhou, China. Daily samples were further divided into four groups by the definition of haze according to visibility and relative humidity (RH), including non-haze (visibility, >10 km), light haze (visibility, 8–10 km, RH <90 %), medium haze (visibility, 5–8 km, RH <90 %), and heavy haze (visibility, <5 km, RH <90 %). Significantly higher concentrations of PM_{2.5}-bound PAHs were found in haze days, but the mean PM_{2.5}-bound PAH concentrations obviously decreased with the aggravation of haze pollution from light to heavy. The gas/particle partitioning coefficients of PAHs decreased from light-haze to heavy-haze episodes, which indicated that PM_{2.5}-bound PAHs were restricted to adhere to the particulate phase with the aggravation of haze pollution. Absorption was considered the main mechanism of gas/particle partitioning of PAHs from gaseous to particulate phase. Analysis of air mass transport indicated that the PM_{2.5}-bound PAH pollution in haze days was largely from regional sources but also significantly affected by long-range air mass transport. The inhalation cancer risk associated with PAHs exceeded the acceptable risk level markedly in both haze and non-haze days.

Keywords : Haze Polycyclic aromatic hydrocarbons Gas/particle partitioning Air mass transport Inhalation cancer risk

15N-labeled ammonium nitrogen uptake and physiological responses of poplar exposed to PM2.5 particles

Huihong Guo, Hui Wang, Qingqian Liu, Hailong An, Chao Liu, Xinli Xia, Weilun Yin

Source: January 2017, Volume 24, Issue 1, pp 500–508

Air pollution caused by particulate matter with aerodynamic diameters less than 2.5 µm (PM_{2.5}) is a serious environmental problem. Plants can improve air quality by removing PM_{2.5} from the atmosphere. However, direct evidence of PM_{2.5} absorption and assimilation into plants has not yet been found. In this study, we demonstrate that ¹⁵NH₄⁺ in PM_{2.5} was absorbed by poplar leaves in low and high PM_{2.5} treatment groups (namely, LPT and HPT). Then, ¹⁵N was subsequently transferred to other parts of the treated seedlings as shown by ¹⁵N tracing and simulated PM_{2.5} generation. ¹⁵N and total N contents

were the highest in high pollution treatment (HPT), followed by that in low pollution treatment (LPT) and the control. Glutamate dehydrogenase (GDH) contributed more to NH_4^+ assimilation than glutamine synthetase and glutamate synthase in the leaves of treated seedlings. GDH aminating activity was induced upon NH_4^+ exposure whereas GDH deaminating activity was repressed in both LPT and HPT, suggesting that poplar seedlings can alleviate NH_4^+ toxicity by enhancing NH_4^+ assimilation. At the end of $\text{PM}_{2.5}$ treatment period, the decreased amino acid content in the treated seedlings was attributed to the probably altered balance of amino acid metabolism. The decline in the net photosynthetic rate (Pn) was accompanied by the decrease in the stomatal conductance in poplar leaves with the extension of $\text{PM}_{2.5}$ treatment time, indicating that stomatal limitation is a major reason for Pn reduction. This study may provide novel insights into the relationship between $\text{PM}_{2.5}$ pollution and plants.

Keywords : PM_{2.5} exposure Poplar seedlings NH₄⁺ assimilation 15N Enzymes Physiological responses

Chemical characterization of PM_{1.0} aerosol in Delhi and source apportionment using positive matrix factorization

Jaiprakash, Amrita Singhai, Gazala Habib, Ramya Sunder Raman, Tarun Gupta

Source: January 2017, Volume 24, Issue 1, pp 445–462

Fine aerosol fraction (particulate matter with aerodynamic diameter $\leq 1.0 \mu\text{m}$ (PM)_{1.0}) over the Indian Institute of Technology Delhi campus was monitored day and night (10 h each) at 30 m height from November 2009 to March 2010. The samples were analyzed for 5 ions (NH_4^+ , NO_3^- , SO_4^{2-} , F^- , and Cl^-) and 12 trace elements (Na, K, Mg, Ca, Pb, Zn, Fe, Mn, Cu, Cd, Cr, and Ni). Importantly, secondary aerosol (sulfate and nitrate) formation was observed during dense foggy events, supporting the fog-smog-fog cycle. A total of 76 samples were used for source apportionment of PM mass. Six factors were resolved by PMF analyses and were identified as secondary aerosol, secondary chloride, biomass burning, soil dust, iron-rich source, and vehicular emission. The geographical location of the sources and/or preferred transport pathways was identified by conditional probability function (for local sources) and potential source contribution function (for regional sources) analyses. Medium- and small-scale metal processing (e.g. steel sheet rolling) industries in Haryana and National Capital Region (NCR) Delhi, coke and petroleum refining in Punjab, and thermal power plants in Pakistan, Punjab, and NCR Delhi were likely contributors to secondary sulfate, nitrate, and secondary chloride at the receptor site. The agricultural residue burning after harvesting season (Sept–Dec and Feb–Apr) in Punjab, and Haryana contributed to potassium at receptor site during November–December and March 2010. The soil dust from North and East Pakistan, and Rajasthan, North-East Punjab, and Haryana along with the local dust contributed to soil dust at the receptor site, during February and March 2010. A combination of temporal behavior and air parcel trajectory ensemble analyses indicated that the iron-rich source was most likely a local source attributed to emissions from metal processing facilities. Further, as expected, the vehicular emissions source did not show any seasonality and was local in origin.

Keywords : Source apportionment Positive matrix factorization (PMF) Biomass burning Secondary aerosol

Surface properties of PM2.5 calcite fine particulate matter in the presence of same size bacterial cells and exocellular polymeric substances (EPS) of *Bacillus mucitagnosus*

Qiongfang Li, Faqin Dong, Qunwei Dai, Cunkai Zhang, Lujia Yu

Source: <https://doi.org/10.1007/s11356-017-0829-x>

Microorganism cells and spores are the main components of PM2.5 (fine particulate matter) as well as fine mineral particles. In the microscopic system, the microorganisms will affect the minerals through attachment, charge neutralization, and dissolution related to the cell surface structure and metabolite. To explore the process and the results of microbial cells and their extracellular polymeric substances (EPS) acting on the surface properties of minerals of PM2.5 through the metabolism, a common native soil bacterium *Bacillus mucitagnosus* with abundant extracellular polymers was chosen as the tested strain. Meanwhile, as one of the PM2.5 common minerals, calcite fine particles were taken as the research object to explore the influence of microbial cells and extracellular polymers on its surface properties. High performance liquid chromatography (HPLC), inductively coupled plasma spectrometry (ICP), Zeta potential analysis, Fourier-transform infrared spectroscopy (FTIR), X-ray diffraction spectroscopy (XRD), and scanning electron microscopy (SEM) were used to characterize the composition of EPS, the soluble ions, surface charge, surface groups, crystal form, and surface morphology of calcite residual solid after being treated by the bacterial cells and EPS. The results revealed the EPS of *B. mucitagnosus* mainly consisted of protein and polysaccharides. Both the whole cell and its EPS could promote the dissolution of calcite particles into calcium ions. Due to the adhesion of organic groups on the calcite surface, the surface potential shifted significantly in the negative direction and the solution pH was clearly increased. The morphology of calcite surface was significantly changed after dissolution and re-crystallization. Experimental results also showed that the existence of the bacteria cells and EPS significantly affected the surface properties of calcite and provide a theoretical basis for the mechanism of PM fine particulate matter on human health impact for further study.

Keywords : Bacteria EPS Calcite Fine particulate matter Surface properties *Bacillus mucitagnosus*

Seasonal variations in size distribution, water-soluble ions, and carbon content of size-segregated aerosols over New Delhi

Pawan Kumar, Sushil Kumar, Sudesh Yadav

Source: <https://doi.org/10.1007/s11356-017-0954-6>

Size distribution, water-soluble inorganic ions (WSII), and organic carbon (OC) and elemental carbon (EC) in size-segregated aerosols were investigated during a year-long sampling in 2010 over New Delhi. Among different size fractions of PM10, PM0.95 was the dominant fraction (45%) followed by PM3–7.2 (20%), PM7.2–10 (15%), PM0.95–1.5 (10%), and PM1.5–3 (10%). All size fractions exceeded the

ambient air quality standards of India for PM_{2.5}. Annual average mass size distributions of ions were specific to size and ion(s); Ca²⁺, Mg²⁺, K⁺, NO₃⁻, and Cl⁻ followed bimodal distribution while SO₄²⁻ and NH₄⁺ ions showed one mode in PM_{0.95}. The concentrations of secondary WSII (NO₃⁻, SO₄²⁻, and NH₄⁺) increased in winters due to closed and moist atmosphere whereas open atmospheric conditions in summers lead to dispersal of pollutants. NH₄⁺ and Ca²⁺ were dominant neutralization ions but in different size fractions. The summer-time dust transport from upwind region by S SW winds resulted in significantly high concentrations of PM_{0.95} and PM_{3–7.2} and PM_{7.2–10}. This indicated influence of dust generation in Thar Desert and its transport is size selective in nature in downwind direction. The mixing of different sources (geogenic, coal combustions, biomass burning, plastic burning, incinerators, and vehicular emissions sources) for soluble ions in different size fractions was noticed in principle component analysis. Total carbon (TC = EC + OC) constituted 8–31% of the total PM_{0.95} mass, and OC dominated over EC. Among EC, char (EC₁) dominated over soot (EC₂ + EC₃). High SOC contribution (82%) to OC and OC/EC ratio of 2.7 suggested possible role of mineral dust and high photochemical activity in SOC production. Mass concentrations of aerosols and WSII and their contributions to each size fraction of PM₁₀ are governed by nature of sources, emission strength of source(s), and seasonality in meteorological parameters.

Keywords : Size-segregated aerosols Carbonaceous aerosols Sources Soluble ions Biomass burning

Modeling the impacts of ambient temperatures on cardiovascular mortality in Yinchuan: evidence from a northwestern city of China

Huiling Zhang, Qingan Wang, Yajuan Zhang, Yi Yang, Yi Zhao, Jianren Sang, Yulong Zhang, Yine Zhang, Fan Xie, Shanshan Li, Yuhong Zhang, Yuming Guo

Source: <https://doi.org/10.1007/s11356-017-0920-3>

No evidence is available on whether cardiovascular mortality is affected by the ambient temperatures in Yinchuan, which is located in the northwestern region of China, with a typical continental semi-humid semi-arid climate. Daily data on cardiovascular mortality and meteorological factors was collected from Yinchuan city for the period of 2010–2015. A distributed lag non-linear model with quasi-Poisson link was used to assess the association between daily temperatures and cardiovascular deaths, after controlling for seasonality, day of the week, atmospheric pressure, humidity, sunshine duration, and wind speed. The relationship between ambient temperature and cardiovascular mortality was non-linear, with a U-shaped exposure-response curve. For all cardiovascular mortality, the effects of high temperatures appeared at lag 2–5 days, with the largest hot effect at lag 3 day (RR 1.082, 95% CI 1.021–1.146), while the effects of cold temperatures were insignificant. Both cold and high temperatures have more serious influence on the elderly (age ≥ 65) and males than the youth and females, respectively. The study has shown that both cold and high temperatures affect cardiovascular mortality. The findings may be helpful to identify the susceptible subgroups of cardiovascular mortality induced by temperatures, and to provide useful information for establishing public health programs that would better protect local population health from ambient temperatures.

Keywords : Ambient temperatures Cardiovascular mortality Distributed lag non-linear model Time-series Northwestern China

Impacts of integrated nutrient management on methane emission, global warming potential and carbon storage capacity in rice grown in a northeast India soil

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Source: <https://doi.org/10.1007/s11356-017-0879-0>

Rice soil is a source of emission of two major greenhouse gases (methane (CH₄) and nitrous oxide (N₂O)) and a sink of carbon dioxide (CO₂). The effect of inorganic fertilizers in combination with various organics (cow dung, green manure (*Sesbania aculeata*) *Azolla*compost, rice husk) on CH₄ emission, global warming potential, and soil carbon storage along with crop productivity were studied at university farm under field conditions. The experiment was conducted in a randomized block design for 2 years in a monsoon rice (cv. *Ranjit*) ecosystem (June–November, 2014 and 2015). Combined application of inorganic (NPK) with *Sesbania aculeata* resulted in high global warming potential (GWP) of 887.4 kg CO₂ ha⁻¹ and low GWP of 540.6 kg CO₂ ha⁻¹ was recorded from inorganic fertilizer applied field. Irrespective of the type of organic amendments, flag leaf photosynthesis of the rice crop increased over NPK application (control). There was an increase in CH₄ emission from the organic amended fields compared to NPK alone. The combined application of NPK and *Azolla* compost was effective in the buildup of soil carbon (16.93 g kg⁻¹) and capacity of soil carbon storage (28.1 Mg C ha⁻¹) with high carbon efficiency ratio (16.9). *Azolla* compost application along with NPK recorded 15.66% higher CH₄ emission with 27.43% yield increment over control. *Azolla* compost application significantly enhanced carbon storage of soil and improved the yielding ability of grain (6.55 Mg ha⁻¹) over other treatments.

Keywords : Carbon storage Organic amendments Methane Rice soil Soil carbon fractions

Acute effects of air pollution on spontaneous abortion, premature delivery, and stillbirth in Ahvaz, Iran: a time-series study

aryam Dastoorpoor, Esmaeil Idani, Gholamreza Goudarzi, Narges Khanjani

Source: <https://doi.org/10.1007/s11356-017-0692-9>

Living in areas with high air pollution may have adverse effects on human health. There are few studies about the association between breathing polluted air and adverse pregnancy outcomes in the Middle East. The aim of this study was to determine the relationship between air pollution and spontaneous abortion, premature birth, and stillbirth in Ahvaz. A time-series study was conducted. Data about

spontaneous abortion, premature deliveries, and stillbirth was collected from Ahvaz Imam Khomeini Hospital. Air pollution data including NO, CO, NO₂, PM₁₀, SO₂, O₃, and climate data were, respectively, collected from the Environmental Protection Agency and the Khuzestan Province Meteorology Office from March 2008 until March 2015. The relationship between air pollutants with the number of abortions, premature births, and stillbirths was found using a quasi-Poisson distributed lag model, adjusted by trend, seasonality, temperature, relative humidity, weekdays, and holidays. The average daily dust in Ahvaz on 7.2% days of the year was higher than 500 µg/m³ (very dangerous). Findings from this study indicate a significant association between each 10-unit increase in SO₂ and spontaneous abortion in lag 0 and 9 days. There was a significant relation between each 10-unit increase in NO₂ and CO, and premature birth in lag 0. Also, we found a significant association between each 10-unit increase in CO and premature delivery in lag 1; PM₁₀ and premature delivery in lags 10, 11, and 12; and NO and premature delivery in lags 3, 4, 10, 11, 12, and 13 (*p*-value < 0.05). Contact with polluted air during pregnancy may increase adverse pregnancy outcomes and stillbirth. Pregnant women should avoid polluted air.

Keywords : Air pollution Abortion Premature delivery Stillbirth Pregnancy

Elemental composition of PM_{2.5} in the urban environment of San Juan, Argentina

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Source: <https://doi.org/10.1007/s11356-017-0793-5>

This study contributes to the current knowledge about air pollution in the province of San Juan, Argentina. Sampling was carried out to measure the fine particulate matter in the atmosphere (PM_{2.5}) of the city of San Juan. PM_{2.5} was collected continuously during the winter and spring seasons of 2014 and 2015, and the concentrations of 14 elements (Pb, Ca, K, Cd, Ni, Cr, Mn, V, Cu, Ti, Ba, Co, Sr, and Fe) were determined in PM_{2.5} filters using the technique of X-ray fluorescence by synchrotron radiation (SR-XRF). The results revealed that PM_{2.5} presented annual and seasonal variations, showing a higher concentration during the winter seasons. In addition, for the elements quantified in the filters, a multivariate analysis (Positive Matrix Factorization) was performed to identify the main sources of emission of these elements in the study area, with a series of components being obtained that corresponded to their compositions, which were assigned physical meanings. The first factor, which was the most important in contribution of the sum of the measured elements (45%), was determined mainly by the elements K, Ti, V, Mn, and Fe, which came predominantly from soil particles. The second factor contributed 30% to the measured species in PM_{2.5}, with higher Ba and Zn content perhaps being related to emissions from vehicular traffic. Finally, the third factor, in which Pb, Cr, and Ca predominated, may be an indicator of industrial activity and contributed 25% of the sum of the measured elements of PM_{2.5}. The results of this study provide the first PM composition database in the province, and this can now be used in the development of mitigation and prevention programs.

Keywords : Fine particulate matter Trace elements Air pollution Source of emission San Juan Argentina

Air quality study in the coastal city of Crotona (Southern Italy) hosting a small-size harbor

Paola Romagnoli, Francesca Vichi, Catia Balducci, Andrea Imperiali, Mattia Perilli, Lucia Paciucci, Francesco Petracchini, Angelo Cecinato

Source: November 2017, Volume 24, Issue 32, pp 25260–25275

Particulate polycyclic aromatic hydrocarbons (PAHs), n-alkanes, and gaseous pollutants were collected from the harbor and the urban area of Crotona (Southern Italy) in October 2015. The atmospheric concentrations of organic substances associated to PM₁₀ were determined daily, while gaseous pollutants (BTX, O₃, SO₂, NO_x, NO₂, and NH₃) were monitored on monthly basis by means of diffusive sampling. Total PAHs reached, on the average, 1.56 ± 0.72 , 0.33 ± 0.14 , and 0.59 ± 0.37 ng/m³ at the urban monitoring stations (Fiore, Fermi) and at the harbor, respectively. The percent distribution and diagnostic concentration ratios of PAHs were similar at Fermi and harbor, whereas differences were found through comparison with Fiore site. Biogenic n-alkanes (n-C₂₉, and n-C₃₁) were the most abundant components, indicating the important impact of terrestrial higher plants in all sites. On the other hand, n-C₂₃-n-C₂₅ homologs originated from incomplete combustion of fossil fuel were not negligible (CPI_{2.5} = 2.4) in harbor, confirming the role of anthropogenic sources there. Inside the harbor, SO₂ concentrations ranged from 5.6 to 14.8 µg/m³ showing the maximum value within the old part of the harbor (touristic port). A statistical significant difference between the harbor and the surroundings was indeed observed for this pollutant, which is a specific marker of ship emissions. The other gaseous species monitored did not exhibit the same distribution, with exception of NH₃ and benzene, whose concentration values ranged from 2.8 to 6.9 µg/m³ and 0.3 to 1.4 µg/m³, respectively, and peaked at the same harbor site. Similarities were found in NO_x, NO₂, and O₃ concentration distributions, showing high values in the New Port area.

Keywords : Harbor PAHs Airborne particulates n-alkanes Inorganic pollutants

Spatial variation of air quality index and urban driving factors linkages: evidence from Chinese cities

Haixia Pu, Kunli Luo, Pin Wang, Shaobin Wang, Shun Kang

Source: February 2017, Volume 24, Issue 5, pp 4457–4468

Daily air quality index (AQI) of 161 Chinese cities obtained from the Ministry of Environmental Protection of China in 2015 is conducted. In this study, to better explore spatial distribution and regional

characteristic of AQI, global and local spatial autocorrelation is utilized. Pearson's correlation is introduced to determine the influence of single urban indicator on AQI value. Meanwhile, multiple linear stepwise regression is chosen to estimate quantitatively the most influential urban indicators on AQI. The spatial autocorrelation analysis indicates that the AQI value of Chinese 161 cities shows a spatial dependency. Higher AQI is mainly located in north and northwest regions, whereas low AQI is concentrated in the south and the Qinghai-Tibet regions. The low AQI and high AQI values in China both exhibit relative immobility through seasonal variation. The influence degree of three adverse urban driving factors on AQI value is ranked from high to low: coal consumption of manufacturing > building area > coal consumption of the power industry. It is worth noting that the risk of exposed population to poor quality is greater in the northern region than in other regions. The results of the study provide a reference for the formulation of urban policy and improvement of air quality in China.

Keywords : Air quality index Urban indicators Spatial dependency Regional difference China

Global warming potential and greenhouse gas emission under different soil nutrient management practices in soybean–wheat system of central India

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Jyothi Raghuwanshi

Source: February 2017, Volume 24, Issue 5, pp 4603–4612

Soil nutrient management is a key component contributing to the greenhouse gas (GHG) flux and mitigation potential of agricultural production systems. However, the effect of soil nutrient management practices on GHG flux and global warming potential (GWP) is less understood in agricultural soils of India. The present study was conducted to compare three nutrient management systems practiced for nine consecutive years in a soybean–wheat cropping system in the Vertisols of India, in terms of GHG flux and GWP. The treatments were composed of 100% organic (ONM), 100% inorganic (NPK), and integrated nutrient management (INM) with 50% organic + 50% inorganic inputs. The gas samples for GHGs (CO₂, CH₄, and N₂O) were collected by static chamber method at about 15-day interval during 2012–13 growing season. The change in soil organic carbon (SOC) content was estimated in terms of the changes in SOC stock in the 0–15 cm soil over the 9-year period covering 2004 to 2013. There was a net uptake of CH₄ in all the treatments in both soybean and wheat crop seasons. The cumulative N₂O and CO₂ emissions were in the order of INM > ONM > NPK with significant difference between treatments ($p < 0.05$) in both the crop seasons. The annual GWP, expressed in terms of CH₄ and N₂O emission, also followed the same trend and was estimated to be 1126, 1002, and 896 kg CO₂ eq ha⁻¹ year⁻¹ under INM, ONM, and NPK treatments, respectively. However, the change in SOC stock was significantly higher under ONM (1250 kg ha⁻¹ year⁻¹) followed by INM (417 kg ha⁻¹ year⁻¹) and least under NPK (198 kg ha⁻¹ year⁻¹) treatment. The wheat equivalent yield was similar under ONM and INM treatments and was significantly lower under NPK treatment. Thus, the GWP per unit grain yield was lower under ONM followed by NPK and INM treatments and varied from 250, 261, and 307 kg CO₂ eq Mg⁻¹ grain yield under ONM, NPK, and INM treatments, respectively.

Keywords : Climate change mitigation GWP per unit yield Organic farming Trace gas emission

Characterization of indoor diesel exhaust emissions from the parking garage of a school

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Source: February 2017, Volume 24, Issue 5, pp 4655–4665

Diesel exhaust (DE) emissions from a parking garage located in the basement of a school were characterized during spring and winter using direct reading devices and integrated sampling methods. Concentrations of CO and NO₂ were evaluated using electrochemical sensors and passive colorimetric tubes, respectively. Elemental and total carbon concentrations were measured using the NIOSH 5040 method. Particle number concentrations (PNCs), respirable particulate matter (PM_{resp}) mass concentrations, and size distributions were evaluated using direct reading devices. Indoor concentrations of elemental carbon, PNC, CO, and NO₂ showed significant seasonal variation; concentrations were much higher during winter ($p < 0.01$). Concentrations of the PM_{resp} and total carbon did not show significant seasonal variation. Pearson correlation coefficients were 0.9 ($p < 0.01$) and 0.94 ($p < 0.01$) between the parking garage and ground floor average daily PNCs, and between the parking garage and first floor average daily PNCs, respectively. Since DE is the main identified source of fine and ultrafine particles in the school, these results suggest that DE emissions migrate from the parking garage into the school. Our results highlight the relevance of direct reading instruments in identifying migration of contaminants and suggest that monitoring PNC is a more specific way of assessing exposure to DE than monitoring the common PM_{resp} fraction.

Keywords : Diesel Exposure Ultrafine particles Elemental carbon Number concentration

Influence of transport from urban sources and domestic biomass combustion on the air quality of a mountain area

Francesco Petracchini, Paola Romagnoli, Lucia Paciucci, Francesca Vichi, Andrea Imperiali, Valerio Paolini, Flavia Liotta, Angelo Cecinato

Source: February 2017, Volume 24, Issue 5, pp 4741–4754

The environmental influence of biomass burning for civil uses was investigated through the determination of several air toxicants in the town of Leonessa and its surroundings, in the mountain region of central Italy. Attention was focussed on PM₁₀, polycyclic aromatic hydrocarbons (PAHs) and regulated gaseous pollutants (nitrogen dioxide, ozone and benzene). Two in-field campaigns were carried out during the summer 2012 and the winter 2013. Contemporarily, air quality was monitored in

Rome and other localities of Lazio region. In the summer, all pollutants, with the exception of ozone, were more abundant in Rome. On the other hand, in the winter, PAH concentration was higher in Leonessa (15.8 vs. 7.0 ng/m³), while PM₁₀ was less concentrated (22 vs. 34 µg/m³). Due to lack of other important sources and to limited impact of vehicle traffic, biomass burning was identified as the major PAH source in Leonessa during the winter. This hypothesis was confirmed by PAH molecular signature of PM₁₀ (i.e. concentration diagnostic ratios and 206 ion mass trace in the chromatograms). A similar phenomenon (i.e. airborne particulate levels similar to those of the capital city but higher PAH loads) was observed in other locations of the province, suggesting that uncontrolled biomass burning contributed to pollution across the Rome metropolitan area.

Keywords : Polycyclic aromatic hydrocarbons (PAHs) Biomass burning Air pollution Molecular signatures Source apportionment Diffusive sampling

Airing ‘clean air’ in Clean India Mission

T. Banerjee, M. Kumar, R.K. Mall, R.S. Singh

Source: March 2017, Volume 24, Issue 7, pp 6399–6413

The submission explores the possibility of a policy revision for considering clean air quality in recently launched nationwide campaign, Clean India Mission (CIM). Despite of several efforts for improving availability of clean household energy and sanitation facilities, situation remain still depressing as almost half of global population lacks access to clean energy and proper sanitation. Globally, at least 2.5 billion people do not have access to basic sanitation facilities. There are also evidences of 7 million premature deaths by air pollution in year 2012. The situation is even more disastrous for India especially in rural areas. Although, India has reasonably progressed in developing sanitary facilities and disseminating clean fuel to its urban households, the situation in rural areas is still miserable and needs to be reviewed. Several policy interventions and campaigns were made to improve the scenario but outcomes were remarkably poor. Indian census revealed a mere 31% sanitation coverage (in 2011) compared to 22% in 2001 while 60% of population (700 million) still use solid biofuels and traditional cook stoves for household cooking. Further, last decade (2001–2011) witnessed the progress decelerating down with rural households without sanitation facilities increased by 8.3 million while minimum progress has been made in conversion of conventional to modern fuels. To revamp the sanitation coverage, an overambitious nationwide campaign CIM was initiated in 2014 and present submission explores the possibility of including ‘clean air’ considerations within it. The article draws evidence from literatures on scenarios of rural sanitation, energy practises, pollution induced mortality and climatic impacts of air pollution. This subsequently hypothesised with possible modification in available technologies, dissemination modes, financing and implementation for integration of CIM with ‘clean air’ so that access to both sanitation and clean household energy may be effectively addressed.

Keywords : Aerosol Climate change Clean energy Health Sanitation Swachh Bharat

Personal exposure to PM_{2.5} associated with heavy metals in four travel modes of Tianjin during the summer season

Bao Qing Wang, Jian Feng Liu, Bo Wei Liu, Hong Hong Niu, Rong Hui Chen, Ze Bei Wang, Jia Jia Zhao, Zi Hui Ren

Source: March 2017, Volume 24, Issue 7, pp 6667–6678

Personal exposure to PM_{2.5} associated with heavy metals were investigated at and around the same road by cycling, walking, taxi and bus in Tianjin, China. One trip on each mode was undertaken during 4 h of both morning and evening peak hours. Results of one-way analysis of variance (ANOVA) to compare mean concentrations of PM_{2.5} and each metal measured by four modes, the enrichment level of heavy metals in four modes and the carcinogenic, non-carcinogenic risk and probabilistic estimation of health risks of metals (Cr, Ni, Cu, Zn and Pb). Arithmetic means of PM_{2.5} personal exposure were 323.66, 313.37, 214.84 and 160.71 µg/m³ for cycling, walking, bus and taxi, which resulted from the difference of source (vehicle exhaust and road dust) of exposure to PM_{2.5}. Na has the highest concentration, followed by Al, Ca, K, Fe, Mg, Zn, Ni, Pb, Cu and Cr. The higher Na concentrations were observed in Tianjin in light of its major sea salt influence. The concentrations of Ca, Mg, Fe and Zn in four modes followed different orders, while other metals have no significant difference between four modes. Enrichment factors of metals in PM_{2.5} showed that some metals are enriched, ranging from contaminated to extremely contaminated, for example, Ni, Cu, Zn, Pb, Na and Cr. Others are barely enriched such as Ca, K, Mg and Fe. It illustrated the former is mainly effected by anthropogenic activities and the source of latter comes from crust. From the results of non-carcinogenic and carcinogenic risks of metals, the intake of metals with inhalation for 4 h by four modes did not pose a significant potential chronic-toxic risk and was an acceptable or tolerable risk at present. But uncertainty analysis of health risks showed there were 4.05 and 6.87% probability that make carcinogenic risk values to exceed 10⁻⁴ when male choose walking/cycling to work. Commuters' rush hour exposures were significantly influenced by mode of transport. We suggest that future work should focus on further research between heavy metals in PM_{2.5} exposure and its specific epidemiology effects.

Keywords : Personal exposure PM_{2.5} Heavy metal Travel mode

Modeling of air pollutant concentrations in an industrial region of Turkey

Gizem Tuna Tuygun, Hicran Altuğ, Tolga Elbir, Eftade E. Gaga

Source: March 2017, Volume 24, Issue 9, pp 8230–8241

The hourly SO₂ and PM₁₀ concentrations in ambient air of the Kutahya city located at the western part of Turkey have exceeded the air quality limits in winter months since several years. The region has major industrial plants including lignite-fired power plants and open-cast mining activities, residential areas, and traffic sources. To obtain and quantify the sector-wise anthropogenic emissions and spatial

distribution of the major pollutants including SO₂, NO_x, PM₁₀, and CO, a comprehensive emission inventory with 1-km spatial resolution was prepared for the year of 2014, and the AERMOD dispersion model was used to predict ambient air concentrations in a domain of 140 km by 110 km. Validation of the model results was also done referring to in situ routine measurements at two monitoring stations located in the study area. Total emissions of SO₂, PM₁₀, NO_x, and CO in the study area were calculated as 64,399, 9770, 24,627, and 29,198 tons/year, respectively. The results showed that industrial plants were the largest sources of SO₂, NO_x, and PM₁₀ emissions, while residential heating and road traffic were the most contributing sectors for CO emissions. Three major power plants in the region with total annual lignite consumption of 10 million tons per year were main sources of high SO₂ concentrations, while high PM₁₀ concentrations mainly originated from two major open-cast lignite mines. Major contributors of high NO_x and CO concentrations were traffic including highways and urban streets, and residential heating with high lignite consumption in urban areas. Results of the dispersion model run with the emission inventory resulted in partially high index of agreement (0.75) with SO₂ measured in the urban station within the modeled area.

Keywords : Emission inventory Dispersion modeling AERMOD Power plants Open-cast mining

Biomonitoring of atmospheric pollution: possibilities and future challenges

Susana Marta Almeida, Humbert Wolterbeek, Bernd Markert, Stefano Loppi

Source: May 2017, Volume 24, Issue 13, pp 11865–11866

This special issue of Environmental Science and Pollution Research highlights selected papers presented at the Seventh International Workshop of BioMAP (BioMAP7), which is focused on biomonitoring of atmospheric pollution, and which was held on June 14–19, 2015, in Lisbon, Portugal.

The series of BioMAP workshops was initiated in 1997, emerged as an effective 3-annual platform for (academic) exchange, and was now held for the third time in Portugal, organized by the Centro de Ciências e Tecnologias Nucleares (Instituto Superior Técnico, Universidade de Lisboa). The workshop brings together both scientists, policy makers and other practitioners in environmental sciences from all over the world, to share answers and ideas and discuss the challenges that should be faced within the realm of atmospheric pollution.

There is an ever growing need for information within the context of possible health hazards due to environmental pollution. This information is necessary to improve air quality management. Biomonitoring is a sensitive, selective and user-friendly method of air quality monitoring, to be used in both ambient, indoor and working place conditions, and the relevant information may be deduced from either the abundance, the behaviour of the organisms, or from the presence of specific substances in the monitor tissues. Biomonitors may be applied both in in situ situations, as in surveys in which monitors are exposed that are transplanted from background level sites.

BioMAP7, in addition to the specific issues related to biomonitoring as a technique, specifically addressed the potential of biomonitoring in assessing human exposure to and effects of exposure to toxic substances: as it is, biomonitoring comprises interdisciplinary approaches, which need input from environmental, biological, chemico-analytical, data-analytical and medical-epidemiological domains.

This special issue cannot fully reflect the diversity and creativity of the ideas and new insights that were shared at BioMAP7. However, as editors, we hope that this issue may prompt scientists from the diverse fields to participate in BioMAP workshops to come: the collected papers show and justify the strong position of the biomonitoring technique in worldwide studies on atmospheric pollution.

Assessment of dioxin-like activity in PM₁₀ air samples from an industrial location in Algeria, using the DRE-CALUX bioassay

Sidali Khedidji, Kim Croes, Noureddine Yassaa, Riad Ladj, Michael S. Denison,
Willy Baeyens, Marc Elskens

Source: May 2017, Volume 24, Issue 13, pp 11868–11877

When compared to the European guidelines, PM₁₀ (particulate matter up to 10-μm size) concentrations in Algeria are often exceeding the maximum limits, and in general, no information exists on the compounds bound on its surface. The objective of this study was to measure the dioxin-like activity of polychlorinated dibenzodioxines and dibenzofurans (PCDD/Fs) and dioxin-like polychlorinated biphenyls (PCBs) in the PM₁₀ fraction at the Sour El Ghoulane cement plant in Algeria. PM₁₀ samples ($n = 23$) were taken between 24 March and 15 April 2013, using a medium volume sampler and 47-mm PTFE filters. The 24-h samples were dried to determine the PM₁₀ content and afterward extracted, cleaned up, and analyzed with the dioxin-responsive element–chemical-activated luciferase gene expression (DRE-CALUX) bioassay. Our results showed that the measured bioanalytical equivalents (BEQs) were similar to those in other international industrial sites worldwide. The PCDD/Fs and dioxin-like PCBs (dl-PCBs) were positively correlated ($\rho = 0.6, p = 0.002$), indicating that they have similar sources. Furthermore, samples from March showed higher PCDD/F and dl-PCB BEQs and humidity but lower temperatures compared to samples from April, while there was no difference in the PM₁₀ concentrations between the two months. These results reveal that PM₁₀ alone is not a good proxy and that meteorological conditions are an important factor in assessing dioxin-like pollution in the atmosphere. It seems that, at present, there is no health hazard through direct airborne human exposure to dioxin-like pollutants in PM₁₀ from this site. However, it is important to monitor these POPs for a longer period of time and also to gain more insight in their distribution between the particulate and gas phase in relation to meteorological conditions.

Keywords: PCDD/Fs PCBs CALUX PM₁₀ Algeria

Exposure assessment of a cyclist to particles and chemical elements

C. A. Ramos, J. R. Silva, T. Faria, T. H. Wolterbeek, S. M. Almeida

Source: May 2017, Volume 24, Issue 13, pp 11879–11889

Cycle paths can be used as a route for active transportation or simply to cycle for physical activity and leisure. However, exposure to air pollutants can be boosted while cycling, in urban environments, due to the proximity to vehicular emissions and elevated breathing rates. The objective of this work was to assess the exposure of a cyclist to particles and to chemical elements by combining real-time aerosol mass concentration reading equipment and biomonitoring techniques. PM₁₀ and PM_{2.5} were measured on three cycle paths located in Lisbon, during weekdays and weekends and during rush hours and off-peak hours resulting in a total of 60 campaigns. Lichens were exposed along cycle paths for 3 months, and their element contents were measured by instrumental neutron activation analysis using the k_0 methodology (k_0 -INAA). Using a bicycle commute route of lower traffic intensity and avoiding rush hours or other times with elevated vehicular congestion facilitate a reduction in exposure to pollutants. The implementation of cycle paths in cities is important to stimulate physical activity and active transportation; however, it is essential to consider ambient air and pollutant sources to create safer infrastructures.

Keywords : Bicycle Particles Biomonitoring Chemical elements INAA

Monitoring PAHs in the petrochemical area of Tarragona County, Spain: comparing passive air samplers with lichen transplants

Noelia Domínguez-Morueco, Sofia Augusto, Laura Trabalón, Eva Pocurull, Francesc Borrull, Marta Schuhmacher, José L. Domingo, Martí Nadal

Source: May 2017, Volume 24, Issue 13, pp 11890–11900

The levels of 16 polycyclic aromatic hydrocarbons (PAHs) were determined in 8 passive air samples (PAS) and 6 lichen transplants (*Ramalina fastigiata*) deployed for a period of 2 months in different zones of Tarragona County (Catalonia, Spain), an area with an important number of chemical and petrochemical industries. The accumulated amount of the sum of the 16 PAHs ranged between 1363 to 7866 ng/sample in air samples. The highest concentration was found in the neighborhood of Puigdelfí (village of Perafort), in the vicinity of a big oil refinery and well under the potential influence of the petrochemical emissions. In lichen samples, the sum of the 16 PAHs ranged between 247 and 841 ng/g (dry weight), being the greatest value also observed in Puigdelfí. Data on the levels and profiles of PAHs in both passive monitoring methods were compared. A significant positive linear correlation was found between the concentrations of low molecular weight PAHs in lichens and the amounts accumulated in passive air samples ($R = 0.827$, $P < 0.05$), being especially significant the correlation of 4-ring PAHs ($R = 0.941$, $P < 0.05$). These results strongly suggest that lichens can be used to monitor gas-phase PAHs, providing data that can be quantitatively translated into equivalents for air.

Keywords : Polycyclic aromatic hydrocarbons (PAHs) Passive air sampling (PAS) Polyurethane foams (PUF) Lichens samples Biomonitoring Tarragona, Spain

Magnetic properties and element concentrations in lichens exposed to airborne pollutants released during cement production

Luca Paoli, Aldo Winkler, Anna Guttová, Leonardo Sagnotti, Alice Grassi, Anna Lackovičová, Dušan Senko, Stefano Loppi

Source: May 2017, Volume 24, Issue 13, pp 12063–12080

The content of selected elements (Al, As, Ca, Cd, Cr, Cu, Fe, Hg, Mn, Ni, Pb, S, Ti, V and Zn) was measured in samples of the lichen *Evernia prunastri* exposed for 30, 90 and 180 days around a cement mill, limestone and basalt quarries and urban and agricultural areas in SW Slovakia. Lichens transplanted around the investigated quarries and the cement mill rapidly (30 days) reflected the deposition of dust-associated elements, namely Ca (at the cement mill and the limestone quarry) and Fe, Ti and V (around the cement mill and the basalt quarry), and their content remained significantly higher throughout the whole period (30–180 days) with respect to the surrounding environment. Airborne pollutants (such as S) progressively increased in the study area from 30 to 180 days. The magnetic properties of lichen transplants exposed for 180 days have been characterized and compared with those of native lichens (*Xanthoria parietina*) and neighbouring bark, soil and rock samples, in order to test the suitability of native and transplanted samples as air pollution magnetic biomonitors. The magnetic mineralogy was homogeneous in all samples, with the exception of the samples from the basalt quarry. The transplants showed excellent correlations between the saturation remanent magnetization (Mrs) and the content of Fe. Native samples had a similar magnetic signature, but the values of the concentration-dependent magnetic parameters were up to two orders of magnitude higher, reflecting higher concentrations of magnetic particles. The concentrations of As, Ca and Cr in lichens correlated with Mrs values after neglecting the samples from the basalt quarry, which showed distinct magnetic properties, suggesting the cement mill as a likely source. Conversely, Ti and Mn were mostly (but not exclusively) associated with dust from the basalt quarry. It is suggested that the natural geological characteristics of the substrate may strongly affect the magnetic properties of lichen thalli. Taking this into account, the results of this study point out the suitability of lichens as air pollution magnetic biomonitors.

Keywords : Air pollution Bioindicators Calcium Dust Environmental magnetism Transplants

The dynamic relationship between structural change and CO₂ emissions in Malaysia: a cointegrating approach

Wajahat Ali, Azrai Abdullah, Muhammad Azam

Source: May 2017, Volume 24, Issue 14, pp 12723–12739

The current study investigates the dynamic relationship between structural changes, real GDP per capita, energy consumption, trade openness, population density, and carbon dioxide (CO₂) emissions within the EKC framework over a period 1971–2013. The study used the autoregressive distributed lagged (ARDL) approach to investigate the long-run relationship between the selected variables. The study also employed the dynamic ordinary least squared (DOLS) technique to obtain the robust long-run estimates. Moreover, the causal relationship between the variables is explored using the VECM Granger causality test. Empirical results reveal a negative relationship between structural change and CO₂ emissions in the long run. The results indicate a positive relationship between energy consumption, trade openness, and CO₂ emissions. The study applied the turning point formula of Itkonen (2012) rather than the conventional formula of the turning point. The empirical estimates of the study do not support the presence of the EKC relationship between income and CO₂ emissions. The Granger causality test indicates the presence of long-run bidirectional causality between energy consumption, structural change, and CO₂ emissions in the long run. Economic growth, openness to trade, and population density unidirectionally cause CO₂ emissions. These results suggest that the government should focus more on information-based services rather than energy-intensive manufacturing activities. The feedback relationship between energy consumption and CO₂ emissions suggests that there is an ominous need to refurbish the energy-related policy reforms to ensure the installations of some energy-efficient modern technologies.

Keywords : Structural change CO₂ EKC ARDL Malaysia

Optimal air quality policies and health: a multi-objective nonlinear approach

Helder Relvas, Ana Isabel Miranda, Claudio Carnevale, Giuseppe Maffei, Enrico Turrini,
Marialuisa Volta

Source: May 2017, Volume 24, Issue 15, pp 13687–13699|

The use of modelling tools to support decision-makers to plan air quality policies is now quite widespread in Europe. In this paper, the Regional Integrated Assessment Tool (RIAT+), which was designed to support policy-maker decision on optimal emission reduction measures to improve air quality at minimum costs, is applied to the Porto Urban Area (Portugal). In addition to technological measures, some local measures were included in the optimization process. Case study results are presented for a multi-objective approach focused on both NO₂ and PM₁₀ control measures, assuming equivalent importance in the optimization process. The optimal set of air quality measures is capable to reduce simultaneously the annual average concentrations values of PM₁₀ and NO₂ in 1.7 and 1.0 µg/m³, respectively. This paper illustrates how the tool could be used to prioritize policy objectives and help making informed decisions about reducing air pollution and improving public health.

Keywords : Urban air quality planning Integrated assessment modelling Emission reduction scenarios Surrogate model Cost-benefit Multi-objective approach

Traffic aerosol lobar doses deposited in the human respiratory system

Maurizio Manigrasso, Claudio Vernale, Pasquale Avino

Source: June 2017, Volume 24, Issue 16, pp 13866–13873

Aerosol pollution in urban environments has been recognized to be responsible for important pathologies of the cardiovascular and respiratory systems. In this perspective, great attention has been addressed to Ultra Fine Particles (UFPs < 100 nm), because they efficiently penetrate into the respiratory system and are capable of translocating from the airways into the blood circulation. This paper describes the aerosol regional doses deposited in the human respiratory system in a high-traffic urban area. The aerosol measurements were carried out on a curbside in downtown Rome, on a street characterized by a high density of autovehicular traffic. Aerosol number-size distributions were measured by means of a Fast Mobility Particle Sizer in the range from 5.6 to 560 nm with a 1 s time resolution. Dosimetry estimates were performed with the Multiple-Path Particle Dosimetry model by means of the stochastic lung model. The exposure scenario close to traffic is represented by a sequence of short-term peak exposures: about 6.6×10^{10} particles are deposited hourly into the respiratory system. After 1 h of exposure in proximity of traffic, 1.29×10^{10} , 1.88×10^{10} , and 3.45×10^{10} particles are deposited in the head, tracheobronchial, and alveolar regions. More than 95 % of such doses are represented by UFPs. Finally, according to the greater dose estimated, the right lung lobes are expected to be more susceptible to respiratory pathologies than the left lobes.

Keywords : Urban air Ultrafine particles Deposition doses Lung lobes FMPS MPPD

Assessment of indoor climate of Mogiła Abbey in Kraków (Poland) and the application of the analogues method to predict microclimate indoor conditions

F. Frasca, A. M. Siani, G. R. Casale, M. Pedone, Ł. Bratasz, M. Strojceki, A. Mleczkowska

Source: June 2017, Volume 24, Issue 16, pp 13895–13907

The microclimatic monitoring of the historic church of Mogiła Abbey (Kraków, Poland) was carried out to study the impact of the environmental parameters on the organic and hygroscopic artworks. Specific indexes were proposed to objectively assess the quality of time series of temperature (T), relative humidity (RH), and carbon dioxide (CO₂) before applying the exploratory data analysis. The series were used to define the historic environmental conditions as stated in the European Standard EN 15757:2010 and with the use of the climate evaluation chart (CEC). It was found that the percentage of time in which T and RH values are within the allowable limits of the ASHRAE (2011) Class B is more than 85 %. This means that, for about 15 % of the time, there is a high risk of mechanical damage to highly vulnerable objects mainly due to the RH variability. The environment at the chancel resulted moister than that at the cornice, and the fungal growth is possible. In addition, the time-weighted preservation index (TWPI) is computed to evaluate the life expectancy of the objects, taking into account the environmental conditions of the site under study. The method of analogues, developed to predict the evolution of a

system given observations of the past and without the knowledge of any equation among variables, was proposed and applied to the time series of temperature, relative humidity, and carbon dioxide with a 1-h sampling time to avoid the influence of the autocorrelation.

Keywords : Temperature Relative humidity Carbon dioxide concentration Historic climate Organic hygroscopic objects Analogues method

Dynamic of submicrometer particles in urban environment

Pasquale Avino, Maurizio Manigrasso

Source: June 2017, Volume 24, Issue 16, pp 13908–13920

Many studies show that particle toxicity increases with decreasing their size, emphasizing the role of submicrometric particles, in particular of ultrafine particles (<100 nm). In fact, particles greater than 2.5 μm are quickly removed through dry and wet deposition on the timescale of hours whereas submicrometer particles may reside in atmosphere for weeks, penetrate in indoor environment, and be long-range transported. High aerosol size resolution measurements are important for a correct assessment of the deposition efficiency in the human respiratory system, and time resolution is another important requisite. Starting from such considerations, time-resolved aerosol particle number size distributions have been measured in downtown Rome. Fast Mobility Particle Sizer (FMPS) and Scanning Mobility Particle Sizer (SMPS) measurements have been carried out at the INAIL's Pilot Station, located in downtown Rome, in an area characterized by high density of autovehicular traffic. The two instruments have allowed to investigate deeply the urban aerosol in the range of 5.6–560 and 3.5–117 nm, respectively. In particular, the FMPS measurements have confirmed the interpretation about the transition phenomena in the time interval of few seconds, timescale typically associated with the emission of gasoline and diesel engines. In downtown Rome, the hourly average size distribution is bimodal or trimodal with maxima at about 5–15, 20–30, and 70–100 nm. Particle formation in the nucleation mode was associated to freshly emitted autovehicular exhaust.

Keywords : Submicrometer particles Ultrafine particles Size distribution FMPS Radon NOx Urban Atmosphere

Short-term effects of air pollution on daily hospital admissions for cardiovascular diseases in western China

Yuxia Ma, Haipeng Zhang, Yuxin Zhao, Jianding Zhou, Sixu Yang, Xiaodong Zheng, Shigong Wang

Source: June 2017, Volume 24, Issue 16, pp 14071–14079

Controlling the confounding factors on cardiovascular diseases, such as long-time trend, calendar effect, and meteorological factors, a generalized additive model (GAM) was used to investigate the short-term effects of air pollutants (PM₁₀, SO₂, and NO₂) on daily cardiovascular admissions from March 1st to May 31st during 2007 to 2011 in Lanzhou, a heavily polluted city in western China. The influences of air pollutants were examined with different lag structures, and the potential effect modification by dust storm in spring was also investigated. Significant associations were found between air pollutants and hospital admissions for cardiovascular diseases both on dust event days and non-dust event days in spring. Air pollutants had lag effects on different age and gender groups. Relative risks (RRs) and their 95% confidence intervals (CIs) associated with a 10 µg/m³ increase were 1.14 (1.04~1.26) on lag1 for PM₁₀, 1.31 (1.21~1.51) on lag01 for SO₂, and 1.96 (1.49~2.57) on lag02 for NO₂ on dust days. Stronger effects of air pollutants were observed for females and the elderly (≥60 years). Our analysis concluded that the effects of air pollutants on cardiovascular admissions on dust days were significantly stronger than non-dust days. The current study strengthens the evidence of effects of air pollution on health and dust-exacerbated cardiovascular admissions in Lanzhou.

Keywords : Air pollution Hospital admissions Cardiovascular diseases Time-series Dust storm

Quantitative cancer risk assessment and local mortality burden for ambient air pollution in an eastern Mediterranean City

Hassan R. Dhaini, Thérèse Salameh, Antoine Waked, Stéphane Sauvage, Agnès Borbon, Paola Formenti, Jean-François Doussin, Nadine Locoge, Charbel Afif

Source: June 2017, Volume 24, Issue 16, pp 14151–14162

Health risks posed by ambient air pollutants to the urban Lebanese population have not been well characterized. The aim of this study is to assess cancer risk and mortality burden of non-methane hydrocarbons (NMHCs) and particulates (PM) based on two field-sampling campaigns conducted during summer and winter seasons in Beirut. Seventy NMHCs were analyzed by TD-GC-FID. PM_{2.5} elemental carbon (EC) components were examined using a Lab OC-EC aerosol Analyzer, and polycyclic aromatic hydrocarbons were analyzed by GC-MS. The US EPA fraction-based approach was used to assess non-cancer hazard and cancer risk for the hydrocarbon mixture, and the UK Committee on Medical Effects of Air Pollutants (COMEAP) guidelines were followed to determine the PM_{2.5} attributable mortality burden. The average cumulative cancer risk exceeded the US EPA acceptable level (10⁻⁶) by 40-fold in the summer and 30-fold in the winter. Benzene was found to be the highest contributor to cancer risk (39–43%), followed by 1,3-butadiene (25–29%), both originating from traffic gasoline evaporation and combustion. The EC attributable average mortality fraction was 7.8–10%, while the average attributable number of deaths (AD) and years of life lost (YLL) were found to be 257–327 and 3086–3923, respectively. Our findings provide a baseline for future air monitoring programs, and for interventions aiming at reducing cancer risk in this population.

Keywords : Air pollution NMHC Particulate matter Lebanon Cancer risk Mortality burden

Carbon dioxide emissions, output, and energy consumption categories in Algeria

Fethi Amri

Source: June 2017, Volume 24, Issue 17, pp 14567–14578

This study examines the relation between CO₂ emissions, income, non-renewable, and renewable energy consumption in Algeria during the period extending from 1980 to 2011. Our work gives particular attention to the validity of environmental Kuznets curve (EKC) hypothesis. The autoregressive distributed lag (ARDL) with break point method outcome demonstrates the positive effect of non-renewable type of energy on CO₂ emissions consumption. On the contrary, the results reveal an insignificant effect of renewable energy on environment improvement. Moreover, the results accept the existence of EKC hypothesis but the highest gross domestic product value in logarithm scale of our data is inferior to the estimated turning point. Consequently, policy-makers in Algeria should expand the ratio of renewable energy and should decrease the quota of non-renewable energy consumption.

Keywords : CO₂ emissions Renewable energy consumption Non-renewable energy consumption Economic growth Algeria Environmental Kuznets Curve ARDL model with breakpoint

Chemical characteristics and source apportionment of PM_{2.5} using PCA/APCS, UNMIX, and PMF at an urban site of Delhi, India

Srishti Jain, Sudhir Kumar Sharma, Nikki Choudhary, Renu Masiwal, Mohit Saxena, Ashima Sharma, Tuhin Kumar Mandal, Anshu Gupta, Naresh Chandra Gupta, Chhemendra Sharma

Source: June 2017, Volume 24, Issue 17, pp 14637–14656

The present study investigated the comprehensive chemical composition [organic carbon (OC), elemental carbon (EC), water-soluble inorganic ionic components (WSICs), and major & trace elements] of particulate matter (PM_{2.5}) and scrutinized their emission sources for urban region of Delhi. The 135 PM_{2.5} samples were collected from January 2013 to December 2014 and analyzed for chemical constituents for source apportionment study. The average concentration of PM_{2.5} was recorded as $121.9 \pm 93.2 \mu\text{g m}^{-3}$ (range 25.1–429.8 $\mu\text{g m}^{-3}$), whereas the total concentration of trace elements (Na, Ca, Mg, Al, S, Cl, K, Cr, Si, Ti, As, Br, Pb, Fe, Zn, and Mn) was accounted for ~17% of PM_{2.5}. Strong seasonal variation was observed in PM_{2.5} mass concentration and its chemical composition with maxima during winter and minima during monsoon seasons. The chemical composition of the PM_{2.5} was reconstructed using IMPROVE equation, which was observed to be in good agreement with the gravimetric mass. Source apportionment of PM_{2.5} was carried out using the following three different receptor models: principal component analysis with absolute principal component scores (PCA/APCS), which identified five major sources; UNMIX which identified four major sources; and positive matrix factorization (PMF), which explored seven major sources. The applied models were able

to identify the major sources contributing to the PM_{2.5} and re-confirmed that secondary aerosols (SAs), soil/road dust (SD), vehicular emissions (VEs), biomass burning (BB), fossil fuel combustion (FFC), and industrial emission (IE) were dominant contributors to PM_{2.5} in Delhi. The influences of local and regional sources were also explored using 5-day backward air mass trajectory analysis, cluster analysis, and potential source contribution function (PSCF). Cluster and PSCF results indicated that local as well as long-transported PM_{2.5} from the north-west India and Pakistan were mostly pertinent.

Keywords : Receptor model PCA/APCS UNMIX PMF PSCF Source apportionment

Integrated management of ash from industrial and domestic combustion: a new sustainable approach for reducing greenhouse gas emissions from energy conversion

Laura Benassi, Rogerta Dalipi, Veronica Consigli, Michela Pasquali, Laura Borgese,
Laura E. Depero, Francis Clegg, Paul A. Bingham, Elza Bontempi

Source: June 2017, Volume 24, Issue 17, pp 14834–14846

This work supports, for the first time, the integrated management of waste materials arising from industrial processes (fly ash from municipal solid waste incineration and coal fly ash), agriculture (rice husk ash), and domestic activities (ash from wood biomass burning in domestic stoves). The main novelty of the paper is the reuse of wood pellet ash, an underestimated environmental problem, by the application of a new technology (COSMOS-RICE) that already involves the reuse of fly ashes from industrial and agricultural origins. The reaction mechanism involves carbonation: this occurs at room temperature and promotes permanent carbon dioxide sequestration. The obtained samples were characterized using XRD and TGA (coupled with mass spectroscopy). This allowed quantification of the mass loss attributed to different calcium carbonate phases. In particular, samples stabilized using wood pellet ash show a weight loss, attributed to the decomposition of carbonates greater than 20%. In view of these results, it is possible to conclude that there are several environmental benefits from wood pellet ash reuse in this way. In particular, using this technology, it is shown that for wood pellet biomass the carbon dioxide conversion can be considered negative.

Keywords : Waste Pellet ash reuse Rice husk ash COSMOS-RICE, CO₂ sequestration, sustainability

Impact of shipping emissions on ozone levels over Europe: assessing the relative importance of the Standard Nomenclature for Air Pollution (SNAP) categories

Efthimios Tagaris, Ioannis Stergiou, Rafaella–Eleni P. Sotiropoulou

Source: June 2017, Volume 24, Issue 17, pp 14903–14909

The impact of shipping emissions on ozone mixing ratio over Europe is assessed for July 2006 using the Community Multiscale Air Quality modeling system and the Netherlands Organization for Applied Scientific Research anthropogenic emission inventory. Results suggest that ship-induced ozone contribution to the total surface ozone exceeds 5% over the sea and near the coastline, while an increase up to 5% is simulated over a large portion of the European land. The largest impact (i.e., an increase up to 30%) is simulated over the Mediterranean Sea. In addition, shipping emissions are simulated to increase NO₂ mixing ratio more than 90%, locally, and to modify the oxidizing capacity of the atmosphere through hydroxyl radical formation (increase by 20–60% over the sea along the European coasts and near the coastal zone). Therefore, emissions from ships may counteract the benefits derived from the anthropogenic emissions reduction strategies over the continent. Simulations suggest regions where shipping emissions have a major impact on ozone mixing ratio compared to individual anthropogenic emission sector categories. Shipping emissions are estimated to play an important role on ozone levels compared to road transport sector near the coastal zone. The impact of shipping emissions on ozone formation is also profound over a great part of the European land compared to the rest of anthropogenic emission categories.

Keywords : Shipping emissions Ozone Europe CMAQ SNAP categories

Effects of fine particulate matter and its constituents on emergency room visits for asthma in southern Taiwan during 2008–2010: a population-based study

Su-Lun Hwang, Yu-Ching Lin, Chieh-Mo Lin, Kuang-Yu Hsiao

Source: June 2017, Volume 24, Issue 17, pp 15012–15021

This population-based study evaluated the short-term association between fine particulate matter (PM_{2.5}) concentrations and its constituents and hospital emergency room visits (ERVs) for asthma in southern Taiwan during the period 2008–2010. Data on hospital ERVs for asthma and ambient PM_{2.5} levels and its constituents were obtained from the National Health Insurance Research database and the Environmental Protection Administration, respectively. The quasi-Poisson generalized additive model was used to explore the associations between PM_{2.5} and hospital ERVs for asthma. During the study period, the average daily number of ERVs for asthma and mean 24-h average level of PM_{2.5} was 20.0 and 39.4 $\mu\text{g m}^{-3}$, respectively. The estimated effects of PM_{2.5} on asthma ERVs fluctuated with increasing tendencies after adjusting for O₃ and attenuating tendencies after adjusting for NO₂, SO₂, and CO. Children were more susceptible than other age groups to the effects of PM_{2.5} exposure on asthma ERVs, with the relative risks (RRs) for every 10 $\mu\text{g m}^{-3}$ increase in PM_{2.5} being 1.016 [95% confidence interval (CI) = 1.002–1.030] and 1.018 (95% CI = 1.002–1.034), respectively, at a lag 0 day (i.e., no lag days) and lag 0–1 days. The effect of PM_{2.5} concentrations on asthma ERVs was similar in male and female. Furthermore, asthma ERVs was significantly associated with concentrations of nitrate (NO₃[–]), with the RR for each 1 $\mu\text{g m}^{-3}$ increase in NO₃[–] concentrations being 1.004 (95% CI = 1.001–1.007) at lag 0 day. In conclusion, both PM_{2.5} concentrations and its chemical constituents are associated with ERVs for asthma; moreover, children were more susceptible to the effects of PM_{2.5} in southern Taiwan. PM_{2.5} constituent, nitrate, is more closely related to ERVs for asthma.

Difference of performance in response to disease admissions between daily time air quality indices and those derived from average and entropy functions

Li-Wei Lai, Wan-Li Cheng

Source: June 2017, Volume 24, Issue 17, pp 14924–14933

Daily time air quality indices, which can reflect air quality in 1 day, are suitable for identifying daily exposure during conditions of poor air quality. The aim of this study is to compare the main effectiveness of four daily time indices in representing variation in the number of disease admissions. These indices include pollution standard index (PSI), air quality index (AQI) and their respective indices derived from mean and entropy functions: MEPSI and MEAQI. The hourly concentrations of fine particulate matter less than 10 μm in diameter (PM₁₀), PM_{2.5}, O₃, CO, NO₂ and SO₂ from 1 January 2006 to 31 December 2010 were obtained from 14 air quality monitoring stations owned by the Environmental Protection Administration (EPA) in the Kaoping region, Taiwan.

Instead of circulatory system disease admissions, the indices were correlative with the number of respiratory disease admissions with correlative coefficients of 0.49 to 0.56 ($P < 0.05$). The daily time air quality indices derived from mean and entropy functions improved their performance of reactive range and air pollution identification. The reactive range of MEPSI and MEAQI was 1.4–3 times that of the original indices. The MEPSI and MEAQI increased identification from 40 to 180 in index scale and revealed one to two additional categories of public health effect information. In comparison with other indices, MEAQI is more effective for application to pollution events with multiple air pollutants.

Keywords : Pollutant standard index (PSI) Air quality index (AQI) PSI derived from mean and entropy functions (MEPSI) AQI derived from mean and entropy functions (MEAQI) Circulatory system disease admissions Respiratory disease admissions Public health

The short-term associations of weather and air pollution with emergency ambulance calls for paroxysmal atrial fibrillation

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Kristina Lopatiene, Nijole Ragaisyte

Source: June 2017, Volume 24, Issue 17, pp 15031–15043

A circadian variation in the cardiovascular parameters has been detected. It is plausible that the influence of the environment varies during different periods of the day. We investigated the association between daily emergency ambulance calls (EC) for paroxysmal atrial fibrillation (AF) that occurred during the time intervals of 8:00–13:59, 14:00–21:59, and 22:00–7:59, and weather conditions and exposure to CO and PM10. We used Poisson regression to explore the association between the risk of EC for AF and environmental variables, adjusting for seasonal variation. Before noon, the risk was associated with an IQR (0.333 mg/m³) increase in CO at lag 2–6 days above the median (RR = 1.15, P = 0.002); a protective impact of CO on previous day was observed (RR = 0.91, P = 0.018). During 14:00–21:59, a negative effect of air temperature below 1.9 °C (lag 2–3 days) was detected (per 10 °C decrease: RR = 1.17, P = 0.044). At night, the elevated risk was associated with wind speed above the median (lag 2–4 days) (per 1-kt increase: RR = 1.07, P = 0.001) and with PM10 at lag 2–5 days below the median (per IQR (7.31 µg/m³) increase: RR = 1.21, P = 0.002). Individuals over 65 years of age were more sensitive to air pollution, especially at night (CO lag 2–3 days < median, per IQR (0.12 mg/m³) increase: RR = 1.14, P = 0.045; PM10 lag 2–5 days < median, per IQR increase: RR = 1.32, P = 0.001). The associations of air pollution and other environmental variables with acute events may be analyzed depending on the time of the event.

Keywords : Weather Carbon monoxide Particulate matter Emergency ambulance calls Paroxysmal atrial fibrillation Risk

Air pollution and respiratory health among diabetic and non-diabetic subjects in Pune, India—results from the Wellcome Trust Genetic Study

Morteza Abdullatif Khafaie, Sundeep Santosh Salvi, Chittaranjan Sakerlal Yajnik, Ajay Ojha, Behzad Khafaie, Sharad Damodar Gore

Diabetics may be more vulnerable to the harmful effects of ambient air pollutants than healthy individuals. But, the risk factors that lead to susceptibility to air pollution in diabetics have not yet been identified. We examined the effect of exposure to ambient PM10 on chronic symptoms and the pulmonary function tests (PFT) in diabetic and non-diabetic subjects. Also, to investigate possible determinants of susceptibility, we recruited 400 type 2 diabetic and 465 healthy subjects who were investigated for chronic respiratory symptoms (CRSs) and then underwent measurement of forced vital capacity (FVC) and forced expiratory volume 1 (FEV1) according to standard protocol. Percent predicted FEV1 and FVC (FEV1% and FVC%, respectively) for each subject were calculated. Particulate matter (PM10) concentrations at residence place of subjects were estimated using AERMOD dispersion model. The association between PM10 and CRSs was explored using logistic regression. We also used linear regression models controlling for potential confounders to study the association between chronic exposure to PM10 and FEV1% and FVC%. Prevalence of current wheezing, allergy symptom, chest tightness, FEV1/FVC <70%, and physician-diagnosed asthma and COPD was significantly higher among diabetic subjects than non-diabetics. There was no significant difference between percent predicted value of PFT among diabetic and non-diabetic subjects (P < 0.05). We estimated that 1 SD increase in PM10 concentration was associated with a greater risk of having dyspnea by 1.50-fold (95% CI, 1.12–

2.01). Higher exposure to PM10 concentration was also significantly associated with lower FVC%. The size of effect for 1 SD $\mu\text{g}/\text{m}^3$ (=98.38) increase in PM10 concentration was 3.71% (95% CI, 0.48–4.99) decrease in FVC%. In addition, we indicated that strength of these associations was higher in overweight, smoker, and aged persons. We demonstrated a possible contribution of air pollution to reduced lung function independent of diabetes status. This study suggests that decline in exposure may significantly reduce disease manifestation as dyspnea and impaired lung function. We conduct that higher BMI, smoking, and older age were associated with higher levels of air pollution effects.

Keywords : Air pollution PM10 Chronic respiratory symptom Lung function Type 2 diabetes mellitus

Portuguese agriculture and the evolution of greenhouse gas emissions— can vegetables control livestock emissions?

Paulo Reis Mourao, Vítor Domingues Martinho

Source: July 2017, Volume 24, Issue 19, pp 16107–16119

One of the most serious externalities of agricultural activity relates to greenhouse gas emissions. This work tests this relationship for the Portuguese case by examining data compiled since 1961. Employing cointegration techniques and vector error correction models (VECMs), we conclude that the evolution of the most representative vegetables and fruits in Portuguese production are associated with higher controls on the evolution of greenhouse gas emissions. Reversely, the evolution of the output levels of livestock and the most representative animal production have significantly increased the level of CO₂ (carbon dioxide) reported in Portugal. We also analyze the cycle length of the long-term relationship between agricultural activity and greenhouse gas emissions. In particular, we highlight the case of synthetic fertilizers, whose values of CO₂ have quickly risen due to changes in Portuguese vegetables, fruit, and animal production levels.

Keywords : Agricultural externalities Greenhouse gas emission Portugal

Impact of socioeconomic and meteorological factors on reservoirs' air quality: a case in the Three Gorges Reservoir of Chongqing (TGRC), China over a 10-year period

Ying Peng, Fengwu Zhou, Jian Cui, Ke Du, Qiangmei Leng, Fumo Yang, Andy Chan,
Hongting Zhao

Source: July 2017, Volume 24, Issue 19, pp 16206–16219

The Three Gorges Dam's construction and industrial transfer have resulted in a new air pollution pattern with the potential to threaten the reservoir eco-environment. To assess the impact of socioeconomic

factors on the pattern of air quality variation and economical risks, concentrations of SO₂, NO₂, and PM₁₀, industry genres, and meteorological conditions were selected in the Three Gorges Reservoir of Chongqing (TGRC) during 2006–2015. Results showed that air quality had improved to some extent, but atmospheric NO₂ showed an increased trend during 2011–2015. Spatially, higher atmospheric NO₂ extended to the surrounding area. The primary industry, especially for agriculture, had shown to be responsible for the remarkable increase of atmospheric NO₂ ($p < 0.01$) due to the direct burning of agricultural straws and the emission of livestock breeding. The improvement of regional industrial structure and industrialization benefited air pollutant reductions, but construction industries had inhibited the improvement of regional air quality. In the tertiary industry, the cargo industry at ports had significantly decreased atmospheric NO₂ as a result of eliminating the obsolete small ships. Contrarily, the highway transportation had brought more air pollutants. The relative humidity was shown to be the main meteorological factor, which had an extremely remarkable relation with atmospheric SO₂ ($p < 0.01$) and a significant correlation with atmospheric NO₂ ($p < 0.05$), respectively. In the future, the development of agriculture and livestock breeding would make regional air quality improvement difficult, and atmospheric SO₂, NO₂, and PM₁₀ deposition would aggravate regional soil and water acidification and reactivate heavy metal in soil and sediment, further to pose a high level of ecological risk in the TGRC and other countries with reservoirs in the world.

Keywords : Air quality Temporal-spatial variation Socioeconomic factor Meteorology Three Gorges Reservoir

Air protection programmes in Poland in the context of the low emission

Janusz Adamczyk, Arkadiusz Piwowar, Maciej Dzikuć

Source: July 2017, Volume 24, Issue 19, pp 16316–16327

The protection of the air against pollutants from individual boiler plants is a big challenge in Poland. It results mainly from the preference for coal, the national energy carrier, the use of old low-efficiency boilers and the location of Poland in a temperate climate where the heating period lasts at least 5 months. This article presents a wide range of activities aimed at the reduction of the environmental impact of the emissions of pollutants from individual heat sources—the so-called low emission. The article presents the extent of the national legislation resulting from the European Union regulations. It discusses the assumptions of the air protection programmes (APPs) and the low emission reduction programmes (LERPs). The assumptions mentioned above are analysed as part of a life cycle assessment (LCA) analysis and a multi-criterion analysis. An important result of these analyses (in the Polish conditions) is the conclusion that a boiler fired with large pieces of wood is an optimal solution from the economic and ecological points of view. The article proposes systemic, organisational and legislative solutions whose implementation could contribute to raising the effectiveness of the protection of the atmosphere.

Keywords : “Low emission” Air protection programmes LCA analysis Choice of heat

Human health impact assessment of exposure to particulate matter: an AirQ software modeling

Mohammad Miri, Hamideh Ebrahimi Aval, Mohammad Hassan Ehrampoush, Amir Mohammadi, Ali Toolabi, Ali Nikonahad, Zahra Derakhshan, Ali Abdollahnejad

Source: July 2017, Volume 24, Issue 19, pp 16513–16519

The aim of this study was to assess the health impacts related to particulate matter less than 10 μm (PM10) exposure in the city of Yazd, Iran. For this aim, AirQ 2.2.3 software was used to model relationship between short-term exposure to PM10 and disease cases proposed by the World Health Organization (WHO). The annual mean concentration of PM10 was 97 $\mu\text{g}/\text{m}^3$. The maximum concentration value of PM10 was measured during the summer (731 $\mu\text{g}/\text{m}^3$). 4.988% (95%CI: 3.381–6.542%) of the total mortality, 7.3% (95%CI: 4.19–10.21%) of cardiovascular mortality, and 10.21% (95%CI: 4.19–14.89%) of respiratory mortality were related to the PM10 concentrations. Consequently, the AirQ software can provide valuable information about the importance of air pollution and the substantial impacts of PM10 on the society for policymakers.

Keywords : AirQ model Health impact assessment Mortality Air pollution

Temporal comparison of global inventories of CO₂ emissions from biomass burning during 2002–2011 derived from remotely sensed data

Yusheng Shi, Tsuneo Matsunaga

Source: July 2017, Volume 24, Issue 20, pp 16905–16916

Biomass burning is a large important source of greenhouse gases and atmospheric aerosols, and can contribute greatly to the temporal variations of CO₂ emissions at regional and global scales. In this study, we compared four globally gridded CO₂ emission inventories from biomass burning during the period of 2002–2011, highlighting the similarities and differences in seasonality and interannual variability of the CO₂ emissions both at regional and global scales. The four datasets included Global Fire Emissions Database 4s with small fires (GFED4s), Global Fire Assimilation System 1.0 (GFAS1.0), Fire INventory from NCAR 1.0 (FINN1.0), and Global Inventory for Chemistry-Climate studies-GFED4s (G-G). The results showed that in general, the four inventories presented consistent temporal trend but with large differences as well. Globally, CO₂ emissions of GFED4s, GFAS1.0, and G-G all peaked in August with the exception in FINN1.0, which recorded another peak in annual March. The interannual trend of all datasets displayed an overall decrease in CO₂ emissions during 2002–2011, except for the inconsistent FINN1.0, which showed a tendency to increase during the considered period. Meanwhile, GFED4s and GFAS1.0 noted consistent agreement from 2002 to 2011 at both global ($R^2 > 0.8$) and continental levels ($R^2 > 0.7$). FINN1.0 was found to have the poorest temporal correlations with the other three inventories globally ($R^2 < 0.6$). The lower estimation in savanna CO₂ emissions and higher calculation in cropland CO₂ emissions by FINN1.0 from 2002 to 2011 was the primary reason for the temporal

differences of the four inventories. Besides, the contributions of the three land covers (forest, savanna, and cropland) on CO₂ emissions in each region varied greatly within the year (>80%) but showed small variations through the years (<40%).

Keywords : Biomass burning CO₂ emissions Remote sensing Temporal variation Fires

Historical and future emission of hazardous air pollutants (HAPs) from gas-fired combustion in Beijing, China

Yifeng Xue, Lei Nie, Zhen Zhou, Hezhong Tian, Jing Yan, Xiaoqing Wu, Linglong Cheng

Source: July 2017, Volume 24, Issue 20, pp 16946–16957

The consumption of natural gas in Beijing has increased in the past decade due to energy structure adjustments and air pollution abatement. In this study, an integrated emission inventory of hazardous air pollutants (HAPs) emitted from gas-fired combustion in Beijing was developed for the period from 2000 to 2014 using a technology-based approach. Future emission trends were projected through 2030 based on current energy-related and emission control policies. We found that emissions of primary HAPs exhibited an increasing trend with the rapid increase in natural gas consumption. Our estimates indicated that the total emissions of NO_x, particulate matter (PM)₁₀, PM_{2.5}, CO, VOCs, SO₂, black carbon, Pb, Cd, Hg, As, Cr, Cu, Ni, Zn, polychlorinated dibenzo-*p*-dioxins and dibenzofurans, and benzo[a]pyrene from gas-fired combustion in Beijing were approximately 22,422 t, 1042 t, 781 t, 19,097 t, 653 t, 82 t, 19 t, 0.6 kg, 0.1 kg, 43 kg, 52 kg, 0.3 kg, 0.03 kg, 4.3 kg, 0.6 kg, 216 µg, and 242 g, respectively, in 2014. To mitigate the associated air pollution and health risks caused by gas-fired combustion, stricter emission standards must be established. Additionally, combustion optimization and flue gas purification system could be used for lowering NO_x emissions from gas-fired combustion, and gas-fired facilities should be continuously monitored based on emission limits.

Keywords : Gas-fired combustion Hazardous air pollutants (HAPs) Emission inventory Temporal trends Spatial variation

Does trade openness affect CO₂ emissions: evidence from ten newly industrialized countries?

Shun Zhang, Xuyi Liu, Junghan Bae

Source: July 2017, Volume 24, Issue 21, pp 17616–17625

This paper examines whether the hypothetical environmental Kuznet curve (EKC) exists or not and investigates how trade openness affects CO₂ emissions, together with real GDP and total primary energy consumption. The study sample comprises ten newly industrialized countries (NICs-10) from

1971 to 2013. The results support the existence of hypothetical EKC and indicate that trade openness negatively and significantly affects emissions, while real GDP and energy do positive effects of emissions. Moreover, the empirical results of short-run causalities indicate feedback hypothetical linkage of real GDP and trade, unidirectional linkages from energy to emissions, and from trade to energy. The error correction terms (ECTs) reveal in the long run, feedback linkages of emissions, real GDP, and trade openness, while energy Granger causes emissions, real GDP, and trade, respectively. The study recommendations are that our policymakers should encourage and expand the trade openness in these countries, not only to restrain CO₂ emissions but also to boost their growth.

Keywords : Trade openness Newly industrialized countries Carbon dioxide emissions Environmental Kuznet curve Total energy Real GDP

Geographic variations in female breast cancer incidence in relation to ambient air emissions of polycyclic aromatic hydrocarbons

Courtney Large, Yudan Wei

Source: July 2017, Volume 24, Issue 21, pp 17874–17880

A significant geographic variation of breast cancer incidence exists, with incidence rates being much higher in industrialized regions. The objective of the current study was to assess the role of environmental factors such as exposure to ambient air pollution, specifically carcinogenic polycyclic aromatic hydrocarbons (PAHs) that may be playing in the geographic variations in breast cancer incidence. Female breast cancer incidence and ambient air emissions of PAHs were examined in the northeastern and southeastern regions of the USA by analyzing data from the Surveillance, Epidemiology, and End Results (SEER) Program and the State Cancer Profiles of the National Cancer Institute and from the Environmental Protection Agency. Linear regression analysis was conducted to evaluate the association between PAH emissions and breast cancer incidence in unadjusted and adjusted models. Significantly higher age-adjusted incidence rates of female breast cancer were seen in northeastern SEER regions, when compared to southeastern regions, during the years of 2000–2012. After adjusting for potential confounders, emission densities of total PAHs and four carcinogenic individual PAHs (benzo[*a*]pyrene, dibenz[*a,h*]anthracene, naphthalene, and benzo[*b*]fluoranthene) showed a significantly positive association with annual incidence rates of breast cancer, with a β of 0.85 ($p = 0.004$), 58.37 ($p = 0.010$), 628.56 ($p = 0.002$), 0.44 ($p = 0.041$), and 77.68 ($p = 0.002$), respectively, among the northeastern and southeastern states. This study suggests a potential relationship between ambient air emissions of carcinogenic PAHs and geographic variations of female breast cancer incidence in the northeastern and southeastern US. Further investigations are needed to explore these interactions and elucidate the role of PAHs in regional variations of breast cancer incidence.

Keywords : Air emission Breast cancer incidence Geographic variations Polycyclic aromatic hydrocarbons

Different exposure levels of fine particulate matter and preterm birth: a meta-analysis based on cohort studies

Chenchen Liu, Jiantao Sun, Yuewei Liu, Hui Liang, Minsheng Wang, Chunhong Wang, Tingming Shi

Source: August 2017, Volume 24, Issue 22, pp 17976–17984

The previous studies estimated the association between PM_{2.5} (particulate matter with aerodynamic diameter less than or equal to 2.5 µm) exposure during pregnancy and preterm birth, only considered and highlighted the hazard effects of high levels of air pollutant exposure, and underestimated that low levels of pollutant exposure might also affect pregnancy outcome. We conducted a meta-analysis of 11 cohort studies, a total of more than 1,500,000 subjects. The results of these studies were pooled by exposure levels and study periods. PM_{2.5} exposure during pregnancy was positively associated with preterm birth (OR = 1.15, 95% CI = 1.07–1.23), and during the first trimester of pregnancy, low levels of PM_{2.5} exposure were also positively associated with preterm birth (OR = 1.17, 95% CI = 1.04–1.30). It is important to protect pregnant women from PM_{2.5} exposures, especially during their first trimester of pregnancy even when the ambient PM_{2.5} concentration is relatively low. More relevant health policy should be carried out to prevent hazard effect of air pollutants.

Keywords : Exposure levels Fine particulate matter Preterm birth Cohort study Meta-analysis

A comparative study of hospital admissions for respiratory diseases during normal and dusty days in Iran

Sahar Geravandi, Pierre Sicard, Yusef Omidi Khaniabadi, Alessandra De Marco, Ali Ghomeishi, Gholamreza Goudarzi, Mohammad Mahboubi, Ahmad Reza Yari, Sina Dobaradaran, Ghasem Hassani, Mohammad Javad Mohammadi, Shahram Sadeghi

Source: August 2017, Volume 24, Issue 22, pp 18152–18159

During the last century, most of people around the world moved from communicable to non-communicable diseases, mainly due to air pollution. Air pollutants and dust storm increase risk of morbidity, for cardiovascular and respiratory diseases, and increase the number of deaths. The city of Ahvaz is considered as the focal point of air pollution and dust storm in Iran. The aim of this study was to determine the number of Hospital Admission Respiratory Disease (HARD) including asthma attacks, acute bronchitis and chronic obstructive pulmonary disease attributed to PM₁₀ by a descriptive study during normal and dust event days in Ahvaz during the time period 2010–2012. The hourly PM₁₀ data was collected from the Iranian Environmental Protection Agency and Razi hospital. The annual PM₁₀ mean concentrations reached 282, 288 and 278 µg/m³ in 2010, 2011 and 2012, respectively. The number of HARD attributed to PM₁₀ was 1438, 1945 and 1393 people, respectively, and the highest number of daily admissions was attributed to the highest daily PM₁₀ concentration in Ahvaz. The average number of daily HARD during dusty days was higher than normal days, and a significant positive

correlation, between the number of hospital admissions and dusty days, was found. Dust had significant impact on HARD in Ahvaz.

Keywords : PM10 Dusty days Hospital admission Respiratory disease Iran

Influence of polymethyl acrylate additive on the formation of particulate matter and NOX emission of a biodiesel–diesel-fueled engine

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Nurin Wahidah Mohd Zulkifli, Islam Shancita

Source: August 2017, Volume 24, Issue 22, pp 18479–18493

The aim of this study is to investigate the effect of the polymethyl acrylate (PMA) additive on the formation of particulate matter (PM) and nitrogen oxide (NOX) emission from a diesel coconut and/or *Calophyllum inophyllum* biodiesel-fueled engine. The physicochemical properties of 20% of coconut and/or *C. inophyllum* biodiesel–diesel blend (B20), 0.03 wt% of PMA with B20 (B20P), and diesel fuel were measured and compared to ASTM D6751, D7467, and EN 14214 standard. The test results showed that the addition of PMA additive with B20 significantly improves the cold-flow properties such as pour point (PP), cloud point (CP), and cold filter plugging point (CFPP). The addition of PMA additives reduced the engine's brake-specific energy consumption of all tested fuels. Engine emission results showed that the additive-added fuel reduce PM concentration than B20 and diesel, whereas the PM size and NOX emission both increased than B20 fuel and baseline diesel fuel. Also, the effect of adding PMA into B20 reduced Carbon (C), Aluminum (Al), Potassium (K), and volatile materials in the soot, whereas it increased Oxygen (O), Fluorine (F), Zinc (Zn), Barium (Ba), Chlorine (Cl), Sodium (Na), and fixed carbon. The scanning electron microscope (SEM) results for B20P showed the lower agglomeration than B20 and diesel fuel. Therefore, B20P fuel can be used as an alternative to diesel fuel in diesel engines to lower the harmful emissions without compromising the fuel quality.

Keywords : Cold-flow property PMA BSEC Particulate matter NOX emission

Air pollution and hospital visits for acute upper and lower respiratory infections among children in Ningbo, China: A time-series analysis

Pei-wen Zheng, Jian-bing Wang, Zhen-yu Zhang, Peng Shen, Peng-fei Chai, Die Li, Ming-juan Jin, Meng-Ling Tang, Huai-chu Lu, Hong-bo Lin, Kun Chen

Source: August 2017, Volume 24, Issue 23, pp 18860–18869

Acute upper and lower respiratory infections are main causes of mortality and morbidity in children. Air pollution has been recognized as an important contributor to development and exacerbation of

respiratory infections. However, few studies are available in China. In this study, we investigated the short-term effect of air pollution on hospital visits for acute upper and lower respiratory infections among children under 15 years in Ningbo, China. Poisson generalized models were used to estimate the associations between air pollution and hospital visits for acute upper and lower respiratory infections adjusted for temporal, seasonal, and meteorological effects. We found that four pollutants (PM_{2.5}, PM₁₀, NO₂, and SO₂) were significantly associated with hospital visits for acute upper and lower respiratory infections. The effect estimates for acute upper respiratory infections tended to be higher (PM_{2.5} ER = 3.46, 95% CI 2.18, 4.76; PM₁₀ ER = 2.81, 95% CI 1.93, 3.69; NO₂ ER = 11.27, 95% CI 8.70, 13.89; SO₂ ER = 15.17, 95% CI 11.29, 19.19). Significant associations for gaseous pollutants (NO₂ and SO₂) were observed after adjustment for particulate matter. Stronger associations were observed among older children and in the cold period. Our study suggested that short-term exposure to outdoor air pollution was associated with hospital visits for acute upper and lower respiratory infections in Ningbo.

Keywords : Air pollution Hospital visits Acute upper respiratory infection Acute lower respiratory infection

Evaluation of air pollution tolerance index and anticipated performance index of plants and their application in development of green space along the urban areas

Mandeep Kaur, Avinash Kaur Nagpal

Source: August 2017, Volume 24, Issue 23, pp 18881–18895

Air pollution due to vehicular emissions has become one of the most serious problems in the whole world and has resulted in huge threat to both the environment and the health of living organisms (plants, humans, animals, microorganisms). Plants growing along the roadsides get affected at the maximum as they are the primary recipients to different air pollutants and show varied levels of tolerance and sensitivity. Taking this into account, the present work was based on assessment of seasonal variation in air pollution tolerance index (APTI) and anticipated performance index (API) of four roadside plants, namely, *Alstonia scholaris*, *Nerium oleander*, *Tabernaemontana coronaria*, and *Thevetia peruviana* belonging to family Apocynaceae. APTI was calculated by the determination of four important biochemical parameters, viz., pH, relative water content (RWC), total chlorophyll (TChl), and ascorbic acid (AsA) content of leaves. The leaf samples were collected from plants growing at seven different sites of Amritsar (Punjab), India, for pre-monsoon and post-monsoon seasons. Highest APTI (82.14) was reported in *N. oleander* during the pre-monsoon season while the lowest was recorded in *T. coronaria* (18.59) in the post-monsoon season. On the basis of API score, *A. scholaris* was anticipated to be an excellent performer during the pre-monsoon and post-monsoon seasons followed by *N. oleander*, *T. coronaria*, and *T. peruviana*. Linear regression analysis and Pearson's correlation coefficient depicted significant positive correlation between APTI and ascorbic acid content during the pre-monsoon and post-monsoon seasons.

Keywords : Vehicular pollution Roadside plants Total chlorophyll Ascorbic acid Air pollution tolerance index (APTI) Anticipated performance index (API)

Potential lung carcinogenicity induced by chronic exposure to PM2.5 in the rat

Xiaoli Hu, Qingzhao Li, Shifeng Shao, Qiang Zeng, Shoufang Jiang , Qi Wu, Chunyang Jiang

Source: August 2017, Volume 24, Issue 23, pp 18991–19000

Exposure to fine particulate matter (PM2.5) may increase lung cancer risk, but the underlying mechanisms are poorly understood. This study explored the potential carcinogenicity in rat lung induced by chronic exposure to PM2.5. Adult male rats (200–220 g) were treated with PM2.5 (10 mg/kg body weight) by tracheal perfusion once per week for 1 year; the rats were killed, and expression of tumor markers (carcinoembryonic antigen (CEA), neuron-specific enolase (NSE), squamous cell carcinoma antigen (SCCA)), cancer-related genes, and pathological changes were detected. Chronic treatment with PM2.5 significantly increased SCCA and NSE expression in rat lung tissue and serum. Damaged lung tissue structure was observed by hematoxylin and eosin staining. Although no evidence of tumors was detected, the Wnt/ β -catenin signaling, epithelial–mesenchymal transition, vascular endothelial growth factor, and epidermal growth factor receptor pathways were all activated or overexpressed and likely involved in the potential carcinogenicity in the rat model. Additionally, abnormal expression of the proto-oncogenes c-Myc and K-Ras and tumor suppressor p53 can be seen in lung tissue induced by PM2.5 exposure. Chronic exposure to PM2.5 has the potential to be carcinogenic in rat lung.

Keywords : Fine particulate matter Carcinogenicity Lung cancer Wnt Epithelial–mesenchymal transition

Physicochemical properties, in vitro cytotoxic and genotoxic effects of PM1.0 and PM2.5 from Shanghai, China

Yajuan Zou, Yizhao Wu, Yali Wang, Yinsheng Li, Chengyu Jin

Source: August 2017, Volume 24, Issue 24, pp 19508–19516

Exposure to ambient particulate matter (PM) links with a variety of respiratory diseases. However, compared with coarse particles (PM10) and fine particles (PM2.5), submicrometer particles (PM1.0) may be a more important indicator of human health risks. In this study, the cytotoxic and genotoxic effects of PM1.0 samples from Shanghai were examined using A549 cells, and compared with the effects of PM2.5, to better understand the health effects of PM1.0 in this area. The PM1.0 and PM2.5 samples were characterized for morphology, water-soluble inorganic ions, organic and elemental carbon, and metal elements. The cytotoxicity of PMs was measured using cell viability and cell membrane damage assays. The genotoxic effects of PMs were determined using the comet assay, and DNA damage was

quantified using olive tail moment (OTM) values. The physicochemical characterization indicated that PM_{1.0} was enriched in carbonaceous elements and hazardous metals (Al, Zn, Pb, Mn, Cu, and V), whereas PM_{2.5} was more abundant in large, irregular mineral particles. The biological results revealed that both PM_{1.0} and PM_{2.5} could induce significant cytotoxicity and genotoxicity in A549 cells, and that exposure to PM_{1.0} caused more extensive toxic effects than exposure to PM_{2.5}. The greater cytotoxic effects of PM_{1.0} can be attributed to the combined effects of size and chemical composition, whereas the genotoxic effects of PM_{1.0} may be mainly associated with chemical species.

Keywords : PM_{1.0} Physicochemical properties A549 cells Cytotoxic effects Genotoxic effects DNA damage

Chemical characterization of size-segregated PM from different public transport modes and implications of source specific contribution to public exposure

Sabrina Yanan Jiang, Nirmal Kumar Gali, Fenhuan Yang, Junke Zhang, Zhi Ning

Source: August 2017, Volume 24, Issue 24, pp 20029–20040

To investigate the chemical properties of particulate matter (PM) in different public transport microenvironments in Hong Kong, the coarse (2.5–10 µm) and fine (<2.5 µm) PM samples were collected in three different types of transport modes including Mass Transit Railway (MTR)-Aboveground (AG), MTR Underground (UG), and Bus routes from October 2013 to April 2014. Average PM_{2.5} concentrations through UG, AG, and Bus routes were 47.9, 86.8, and 43.8 µg m⁻³, respectively, whereas the coarse PM concentrations were 4–5 folds less. The PM_{2.5}total metal concentrations of AG route were 2.3 and 3.7 times of UG and BUS routes, respectively, compared to those in the other two routes. The most abundant metals at three stations in PM_{2.5} and coarse PM were quite similar and mainly generated by frictional processes of wheels, rails, and brakes of the system as well as by the mechanical wearing of these parts. The most abundant PAH in three routes in PM_{2.5} was ATRQN, followed by 2-MNA, and the sum of them contributed to 35 and 42% of total PAHs in coarse PM and PM_{2.5}, respectively. Crude oils, lubricant oil, diesel emissions would be the major sources of PAHs from MTR aboveground stations. The relative abundance of the *n*-alkanes among different samples was similar to the PAHs and the carbon preference index (CPI) values of the whole *n*-alkanes range were consistently from 0.99 to 1.04 among all samples indicating the significant contribution from the vehicle exhaust and fossil fuel burning. The concentrations of hopanes and steranes were higher in PM_{2.5} than in coarse PM due to diesel and coal burning. These results may provide a unique opportunity to investigate source specific contribution of the PM pollutants to the commuter exposure in public transport.

Keywords : Source specific exposure Urban commute Trace metals Organic species

Chemical characterization of size-segregated PM from different public transport modes and implications of source specific contribution to public exposure

Sabrina Yanan Jiang, Nirmal Kumar Gali, Fenhuan Yang, Junke Zhang, Zhi Ning,

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Keywords : Source specific exposure Urban commute Trace metals Organic species

Effects of air/fuel ratio on gas emissions in a small spark-ignited non-road engine operating with different gasoline/ethanol blends

Waldir Nagel Schirmer, Luciano Zart Olanyk, Carmen Luisa Barbosa Guedes,
Talita Pedroso Quessada, Camilo Bastos Ribeiro, Marlon André Capanema

Source: September 2017, Volume 24, Issue 25, pp 20354–20359

This study investigates the effects of several blends of gasoline and anhydrous ethanol on exhaust emission concentrations of carbon monoxide (CO), total hydrocarbons (HCs), and nitrogen oxides (NO_x) from a small spark-ignited non-road engine (SSINRE). Tests were carried out for different air/fuel equivalence ratios as measured by lambda (λ). A 196 cm³single-cylinder four-stroke engine-generator operating at a constant load of 2.0 kW was used; pollutant gas concentrations were measured with an

automatic analyzer similar to those typically used in vehicle inspections. The results showed that as the ethanol content of the mixture increased the concentrations of CO, HCs, and NO_x reduced by 15, 53, and 34%, respectively, for values of $\lambda < 1$ (rich mixture) and by 52, 31, and 16% for values of $\lambda > 1$ (lean mixture). Overall, addition of anhydrous ethanol to the gasoline helped to reduce emissions of the pollutant gases investigated, what contributes to photochemical smog reduction and quality of life in urban areas.

Keywords : Atmospheric pollution Biofuels Combustion Ethanol Gasoline Otto-cycle engine

Microplastic pollution in deposited urban dust, Tehran metropolis, Iran

Sharareh Dehghani , Farid Moore, Razegheh Akhbarizadeh

Source: September 2017, Volume 24, Issue 25, pp 20360–20371

Environmental pollutants such as microplastics have become a major concern over the last few decades. We investigated the presence, characteristics, and potential health risks of microplastic dust ingestion. The plastic load of 88 to 605 microplastics per 30 g dry dust with a dominance of black and yellow granule microplastics ranging in size from 250 to 500 μm was determined in 10 street dust samples using a binocular microscope. Fluorescence microscopy was found to be ineffective for detecting and counting plastic debris. Scanning electron microscopy, however, was useful for accurate detection of microplastic particles of different sizes, colors, and shapes (e.g., fiber, spherule, hexagonal, irregular polyhedron). Trace amounts of Al, Na, Ca, Mg, and Si, detected using energy dispersive X-ray spectroscopy, revealed additives of plastic polymers or adsorbed debris on microplastic surfaces. As a first step to estimate the adverse health effects of microplastics in street dust, the frequency of microplastic ingestion per day/year via ingestion of street dust was calculated. Considering exposure during outdoor activities and workspaces with high abundant microplastics as acute exposure, a mean of 3223 and 1063 microplastic particles per year is ingested by children and adults, respectively. Consequently, street dust is a potentially important source of microplastic contamination in the urban environment and control measures are required.

Keywords : Microplastic Potential health risk Street dust Ingestion Tehran

Elemental and isotopic determination of lead (Pb) in particulate matter in the Brazilian city of Goiânia (GO) using ICP-MS technique

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Juliana Aparecida Galhardi, Ana Carla Fernandes Gomes, Eduardo de Almeida,
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Source: September 2017, Volume 24, Issue 25, pp 20616–20625

The toxic metal lead (Pb) can be harmful to human health in various manners, but is also considered as a distinguished tracer of environmental pollution since the relative abundance of its four stable isotopes with the atomic masses of 204, 206, 207, and 208 varies with the emission source. This study is focused on the Pb concentrations and isotope ratios in the particulate matter of the Brazilian city of Goiânia in order to determine the main Pb emission sources. Particulate matter samples were collected on clean Teflon filters during rainy and dry season in 2014 in the center of Goiânia city near main roads with a high traffic volume. Pb concentrations as well as stable Pb isotope ratios of the particulate matter samples were analyzed by inductively coupled plasma-mass spectrometry. To apply this analytical technique successfully, it was necessary to optimize parameters in case of acquisition time, detector dead time, and mass discrimination, which affect the measurement accuracy and precision. Results showed that Pb concentrations in Goiânia were different between rainy and dry season. Pb concentrations showed higher values and less variation in dry season than in rainy season. Pb isotope ratios demonstrated significant variations between dry and rainy season. An enrichment of ^{206}Pb isotopes related to ^{207}Pb and ^{208}Pb isotopes was observed in dry season. However, the comparison of the obtained isotopic Pb signature with data of potential Pb sources from previous studies indicated that traffic-related sources should be considered as main Pb source in the particulate matter of Goiânia. These assumptions were incorporated by the calculation of the contribution factor of Pb coming from traffic-related sources by applying binary mixing equations.

Keywords : Lead isotope ratios Particulate matter Industrial emissions Air pollution ICP-MS

Pilot study investigating ambient air toxics emissions near a Canadian kraft pulp and paper facility in Pictou County, Nova Scotia

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Pantelis Andreou

Source: September 2017, Volume 24, Issue 25, pp 20685–20698

Air toxics are airborne pollutants known or suspected to cause cancer or other serious health effects, including certain volatile organic compounds (VOCs), prioritized by the US Environmental Protection Agency (EPA). While several EPA-designated air toxics are monitored at a subset of Canadian National Air Pollution Surveillance (NAPS) sites, Canada has no specific “air toxics” control priorities. Although pulp and paper (P&P) mills are major industrial emitters of air pollutants, few studies quantified the spectrum of air quality exposures. Moreover, most NAPS monitoring sites are in urban centers; in contrast, rural NAPS sites are sparse with few exposure risk records. The objective of this pilot study was to investigate prioritized air toxic ambient VOC concentrations using NAPS hourly emissions data from a rural Pictou, Nova Scotia Kraft P&P town to document concentration levels, and to determine whether these concentrations correlated with wind direction at the NAPS site (located southwest of the mill). Publicly accessible Environment and Climate Change Canada data (VOC concentrations [Granton NAPS ID: 31201] and local meteorological conditions [Caribou Point]) were examined using temporal (2006–2013) and spatial analytic methods. Results revealed several VOCs (1,3-butadiene, benzene, and carbon tetrachloride) routinely exceeded EPA air toxics-associated cancer risk thresholds. 1,3-Butadiene and tetrachloroethylene were significantly higher ($p < 0.05$) when prevailing wind direction blew from the

northeast and the mill towards the NAPS site. Conversely, when prevailing winds originated from the southwest towards the mill, higher median VOC air toxics concentrations at the NAPS site, except carbon tetrachloride, were not observed. Despite study limitations, this is one of few investigations documenting elevated concentrations of certain VOCs air toxics to be associated with P&P emissions in a community. Findings support the need for more research on the extent to which air toxics emissions exist in P&P towns and contribute to poor health in nearby communities.

Keywords : Air toxics Air quality Volatile organic compounds (VOCs) Community health Pulp and paper Cancer risk

Loss of crop yields in India due to surface ozone: an estimation based on a network of observations

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Sachchida Nand Tripathi, Ujjaini Sarkar, Trupti Das, Yerramsetti Venkata Swamy,
Kotalo Rama Gopal, Harish Gadhavi, Modathi Kottungal Satheesh Kumar

Source: September 2017, Volume 24, Issue 26, pp 20972–20981

Surface ozone is mainly produced by photochemical reactions involving various anthropogenic pollutants, whose emissions are increasing rapidly in India due to fast-growing anthropogenic activities. This study estimates the losses of wheat and rice crop yields using surface ozone observations from a group of 17 sites, for the first time, covering different parts of India. We used the mean ozone for 7 h during the day (M7) and accumulated ozone over a threshold of 40 ppbv (AOT40) metrics for the calculation of crop losses for the northern, eastern, western and southern regions of India. Our estimates show the highest annual loss of wheat (about 9 million ton) in the northern India, one of the most polluted regions in India, and that of rice (about 2.6 million ton) in the eastern region. The total all India annual loss of 4.0–14.2 million ton (4.2–15.0%) for wheat and 0.3–6.7 million ton (0.3–6.3%) for rice are estimated. The results show lower crop loss for rice than that of wheat mainly due to lower surface ozone levels during the cropping season after the Indian summer monsoon. These estimates based on a network of observation sites show lower losses than earlier estimates based on limited observations and much lower losses compared to global model estimates. However, these losses are slightly higher compared to a regional model estimate. Further, the results show large differences in the loss rates of both the two crops using the M7 and AOT40 metrics. This study also confirms that AOT40 cannot be fit with a linear relation over the Indian region and suggests for the need of new metrics that are based on factors suitable for this region.

Keywords : Crop yield loss Wheat and rice Surface ozone Crop loss Loss metrics India

Seasonal variation of chemical composition and source apportionment of PM_{2.5} in Pune, India

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Source: September 2017, Volume 24, Issue 26, pp 21065–21072

Particulate matter with size less than or equal to 2.5 μm (PM_{2.5}) samples were collected from an urban site Pune, India, during April 2015 to April 2016. The samples were analyzed for various chemical constituents, including water soluble inorganic ions, organic carbon (OC), and elemental carbon (EC). The yearly mean total mass concentration of PM_{2.5} at Pune was 37.3 $\mu\text{g}/\text{m}^3$, which is almost four times higher than the annual WHO standard (10 $\mu\text{g}/\text{m}^3$), and almost equal to that recommended by the Central Pollution Control Board, India (40 $\mu\text{g}/\text{m}^3$). Measured (OC, EC) and estimated organic matter (OM) were the dominant component ($56 \pm 11\%$) in the total particulate matter which play major role in the regional atmospheric chemistry. Total measured inorganic components formed about 35% of PM_{2.5}. Major chemical contributors to PM_{2.5} mass were OC (30%), SO₄²⁻ (13%), and Cl⁻ and EC (9% each). The high ratios of OC/EC demonstrated the existence of secondary organic carbon. The air mass origin and correlations between the various components indicate that long range transport of pollutants from Indo-Gangetic Plain (IGP) and Southern part of the Arabian Peninsula might have contributed to the high aerosol mass during the dry and winter seasons. To our knowledge, this is the first systematic study that comprehensively explores the chemical characterization and source apportionment of PM_{2.5} aerosol speciation in Pune by applying multiple approaches based on a seasonal perspective. This study is broadly applicable to understanding the differences in anthropogenic and natural sources in the urban environment of particle air pollution over this region.

Keywords : Air pollution PM_{2.5} Chemical composition Biomass burning Long range transport

Geographic variation in Chinese children's forced vital capacity and its association with long-term exposure to local PM₁₀: a national cross-sectional study

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Source: October 2017, Volume 24, Issue 28, pp 22442–22449

The purpose of this study was to estimate the association between Chinese children's forced vital capacity (FVC) and particulate matter with aerodynamic diameter $\leq 10 \mu\text{m}$ (PM₁₀). The FVC data of 71,763 children aged 7 to 18 was collected from 2010 Chinese National Survey on Students' Construction and Health (CNSSCH). The local annual average concentration of PM₁₀, relative humidity, ambient temperature, and other air pollutant data of 30 cities was collected from China Meteorological Administration and Ministry of Environment Protection of China. Then, we used generalized additive model (GAM) to estimate the association between children's FVC and PM₁₀. The obvious geographic

variation in FVC was found in children of 30 Chinese cities ranging from 1647 ml in Xining to 2571 ml in Beijing. The annual average concentration of PM₁₀ was also different, ranging from 40 µg/m³ in Haikou to 155 µg/m³ in Lanzhou. After adjusted individual characteristics, socioeconomic conditions, ambient temperature, relative humidity, and other air pollutants (e.g., NO₂ and SO₂) in the generalized additive model, we found that the increase of PM₁₀ was associated with decrease of FVC in Chinese children. A 10-µg/m³ increase of PM₁₀ was associated with 1.33-ml decrease in FVC (95% confidence interval: -2.18 to -0.47). We also found a larger effect estimate of PM₁₀ on FVC in boys than that in girls. Consistent associations were found in both physically inactive and active children. The increase of PM₁₀ was associated with decrease of children's FVC. We should develop proper public health policy to protect children's respiratory health during growth and development in polluted areas.

Keywords : PM₁₀ Forced vital capacity Long-term exposure Children

Ozone generated by air purifier in low concentrations: friend or foe?

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Source: October 2017, Volume 24, Issue 28, pp 22673–22678

Ozone helps decontamination environments due to its oxidative power, however present toxicity when it is in high concentrations, by long periods of exposition. This study aimed to assess the safety of ozone generator air purifier at concentrations of 0.05 ppm in rats exposed to 3 and 24 h/day for 14 and 28 days. No significant differences are observed between groups in clinical signs, feed and water intake, relative body weight gain and relative weight of organs, macroscopy and microscopy of lungs, and oxidative plasma assay. In this exposure regime, ozone does not cause genotoxicity and no significant changes in pulmonary histology indicative of toxicity. Ozone generated in low concentrations, even in exposure regimes above the recommended is safe, both acute and sub-acute exposition.

Keywords : Acute toxicity Sub-acute toxicity Rats Ozone Oxidative stress Comet assay Micronucleus

The effect of sources and air mass transport on the variability of trace element deposition in central Poland: a cluster-based approach

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Source: October 2017, Volume 24, Issue 29, pp 23026–23038

Measurements of trace element (As, Cu, Cd, Cr, Ni, Pb, Zn) deposition fluxes were conducted simultaneously in two contrasted environments, i.e., urban and forest, between April 2013 and October

2014. This was the first such project in central Poland, aimed at long-term observations of trace elements in the atmosphere and their distribution, transport, and deposition pattern. The receptor sites were different in terms of local meteorological conditions, emission potential, and distance to major anthropogenic sources. The deposition fluxes of all trace elements showed clear seasonal variations, with relatively higher values in winter than in summer. The main factors affecting interannual differences in concentrations and deposition of trace elements in central Poland were local emission from industrial and commercial sources, and changes in atmospheric conditions (wind speed and direction, boundary layer, precipitation amount, air mass origin). In this study, the impact of regional and long-range transport on trace element deposition was determined using the air back-trajectory cluster analysis. During the summertime of 2013 and 2014, the predominant SW and E advections from regional and remote anthropogenic sources in Europe were responsible for high deposition of Cd, Cr, Pb, Cu, and Zn, whereas during the wintertime of 2013/2014, we observed a significant influence of polluted air masses from southeastern regions. Based on the Pb/Zn ratio, it was found that regional sources significantly influenced the aerosol composition and rainwater chemistry within the study domain. However, the role of a long-range transport of anthropogenic pollutants was also important. In addition, a relatively small difference in the Pb/Zn ratio between both sites (urban 0.26 ± 0.18 , forest 0.23 ± 0.17) may suggest (1) very similar contribution of anthropogenic sources and (2) minor importance of atmospheric transformation processes of these metals in the aqueous phase.

Keywords : Urban area Anthropogenic sources Trajectory Trace elements Bulk deposition Rainwater Cluster analysis

Emission analysis on the effect of nanoparticles on neat biodiesel in unmodified diesel engine

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Source: October 2017, Volume 24, Issue 29, pp 23273–23278

Biodiesels derived from the mahua seeds are established as a promising alternative for the diesel fuel owing to its non-edible nature and improved properties. TiO₂ nanoparticle in powder form is added to neat mahua oil biodiesel (BD100) to examine its effect on emission characteristics. TiO₂ nanoparticle is chosen as an additive owing to its catalytic effect, higher surface energy, and larger surface to volume ratio. TiO₂ nanoparticle with an average size of 60 nm was synthesized by sol-gel route. TiO₂ nanoparticles are added with mahua biodiesel (BD100) at 100 and 200 ppm. Mahua oil biodiesel doped with 100 and 200 ppm of TiO₂ nanoparticles are referred as BD100T100 and BD100T200. A constant speed diesel engine is employed for the experimental trail. Engine is fueled with diesel, BD100, BD100T100, and BD100T200, respectively. Experimental result confirmed that the modified fuels (BD100T200 and BD100T100) showed a significant reduction in all the emissions. Further, the addition of TiO₂ nanoparticle (200 ppm) to mahua biodiesel gave respective reduction of 9.3, 5.8, 6.6, and 2.7% in carbon monoxide, hydrocarbon, nitrogen oxide, and smoke emissions when compared to neat mahua biodiesel.

Real-world vehicle emissions as measured by in situ analysis of exhaust plumes

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Source: October 2017, Volume 24, Issue 29, pp 23279–23289

We conducted a 60-day roadside measurement campaign on a busy street in Münster, Germany, during summer 2016. We used gas and particle concentration measurements with high temporal resolution (10 Hz) to quantify both the emission ratios of nitrogen oxides per carbon dioxide (NO_x/CO_2) for over 70,000 individual exhaust plumes as well as the emission ratios for size-resolved particle numbers per carbon dioxide ($d(\text{PN CO}_2^{-1})/d\log D$) for about 10,000 plumes. The real-world fleet passing by the measurement station consisted of passenger cars (85%), buses (5.9%), light duty commercial vehicles (5.7%), trucks (1.7%), and motorcycles (1.6%). The median measured NO_x/CO_2 ratio was 3.33 g kg^{-1} . The median measured PN/CO_2 emission ratio for particles with diameters between 0.03 and $10 \mu\text{m}$ was $5.6 \times 10^{14} \text{ kg}^{-1}$. We compared our results with the Handbook Emission Factors for Road Transport (HBEFA) and the Euro 5 and Euro 6 emission standards by employing traffic counts, assuming the diesel-to-gasoline ratios of vehicles according to registration statistics, and estimating that stop-and-go traffic occurred 65% of the time. Using a conservative estimate, our median ratios exceeded the HBEFA data by more than 65% for NO_x/CO and by a factor of about 100 for PN/CO_2 . Furthermore, our median NO_x emission per kilometer travelled ($\text{NO}_x \text{ km}^{-1}$) exceeded the Euro 5 emission limit for diesel cars by a factor of 3 and exceeded the Euro 6 limit by almost a factor of 7. Additionally, our median particle number emission (PN km^{-1}) exceeded the Euro 5 and Euro 6 limits of diesel cars by a factor of almost 150. These results confirm the presumption that the emissions of a real-world traffic fleet comprehensively exceed the legal limits. Very likely, the widespread presence of defeat devices in vehicle emission control systems plays a major role in this discrepancy. This has a strong impact on the apparent inability of authorities to comply with the legal limits of the NO_2 concentrations in urban air.

Keywords : Real-world vehicle emissions Exhaust plume analysis HBEFA EU emission limits Air quality in Europe Diesel car emissions Nitrogen oxides (NO_x) emissions

Performance, emission, and combustion characteristics of twin-cylinder common rail diesel engine fuelled with butanol-diesel blends

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Source: October 2017, Volume 24, Issue 29, pp 23351–23362

Nitrogen oxides and smoke are the substantial emissions for the diesel engines. Fuels comprising high-level oxygen content can have low smoke emission due to better oxidation of soot. The objective of the paper is to assess the potential to employ oxygenated fuel, i.e., n-butanol and its blends with the neat diesel from 0 to 30% by volume. The experimental and computational fluid dynamic (CFD) simulation is carried out to estimate the performance, combustion, and exhaust emission characteristics of n-butanol-diesel blends for various injection timings (9°, 12°, 15°, and 18°) using modern twin-cylinder, four-stroke, common rail direct injection (CRDI) engine. Experimental results reveal the increase in brake thermal efficiency (BTE) by ~ 4.5, 6, and 8% for butanol-diesel blends of 10% (Bu10), 20% (Bu20), and 30% (Bu30), respectively, compared to neat diesel (Bu0). Maximum BTE for Bu0 is 38.4%, which is obtained at 12° BTDC; however, for Bu10, Bu20 and Bu30 are 40.19, 40.9, and 41.7%, which are obtained at 15° BTDC, respectively. Higher flame speed of n-butanol-diesel blends burn a large amount of fuel in the premixed phase, which improves the combustion as well as emission characteristics. CFD and experimental results are compared and validated for all fuel blends for in-cylinder pressure and nitrogen oxides (NOx), and found to be in good agreement. Both experimental and simulation results witnessed in reduction of smoke opacity, NOx, and carbon monoxide emissions with the increasing n-butanol percentage in diesel fuel.

Keywords : CRDI Combustion analysis Biofuel Emission Butanol CFD

Simulations of the impacts of building height layout on air quality in natural-ventilated rooms around street canyons

Fang Yang, Ke Zhong, Yonghang Chen, Yanming Kang

Source: October 2017, Volume 24, Issue 30, pp 23620–23635

Numerical simulations were conducted to investigate the effects of building height ratio (i.e., HR, the height ratio of the upstream building to the downstream building) on the air quality in buildings beside street canyons, and both regular and staggered canyons were considered for the simulations. The results show that the building height ratio affects not only the ventilation fluxes of the rooms in the downstream building but also the pollutant concentrations around the building. The parameter, outdoor effective source intensity of a room, is then proposed to calculate the amount of vehicular pollutants that enters into building rooms. Smaller value of this parameter indicates less pollutant enters the room. The numerical results reveal that HRs from 2/7 to 7/2 are the favorable height ratios for the regular canyons, as they obtain smaller values than the other cases. While HR values of 5/7, 7/7, and 7/5 are appropriate for staggered canyons. In addition, in terms of improving indoor air quality by natural ventilation, the staggered canyons with favorable HR are better than those of the regular canyons.

Keywords : Asymmetrical street canyon Building height ratio Buildings around streets Ventilation flux Outdoor effective source intensity Natural ventilation

Science Of Total Environment

Variability, drivers, and effects of atmospheric nitrogen inputs across an urban area: Emerging patterns among human activities, the atmosphere, and soils

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Source: Volume 609, 31 December 2017, Pages 1524-1534

<https://doi.org/10.1016/j.scitotenv.2017.07.166>

Atmospheric deposition of nitrogen (N) is a major input of N to the biosphere and is elevated beyond preindustrial levels throughout many ecosystems. Deposition monitoring networks in the United States generally avoid urban areas in order to capture regional patterns of N deposition, and studies measuring N deposition in cities usually include only one or two urban sites in an urban-rural comparison or as an anchor along an urban-to-rural gradient. Describing patterns and drivers of atmospheric N inputs is crucial for understanding the effects of N deposition; however, little is known about the variability and drivers of atmospheric N inputs or their effects on soil biogeochemistry within urban ecosystems. We measured rates of canopy throughfall N as a measure of atmospheric N inputs, as well as soil net N mineralization and nitrification, soil solution N, and soil respiration at 15 sites across the greater Boston, Massachusetts area. Rates of throughfall N are $8.70 \pm 0.68 \text{ kg N ha}^{-1} \text{ yr}^{-1}$, vary 3.5-fold across sites, and are positively correlated with rates of local vehicle N emissions. Ammonium (NH_4^+) composes $69.9 \pm 2.2\%$ of inorganic throughfall N inputs and is highest in late spring, suggesting a contribution from local fertilizer inputs. Soil solution NO_3^- is positively correlated with throughfall NO_3^- inputs. In contrast, soil solution NH_4^+ , net N mineralization, nitrification, and soil respiration are not correlated with rates of throughfall N inputs. Rather, these processes are correlated with soil properties such as soil organic matter. Our results demonstrate high variability in rates of urban throughfall N inputs, correlation of throughfall N inputs with local vehicle N emissions, and a decoupling of urban soil biogeochemistry and throughfall N inputs.

Keywords : Atmospheric deposition, Soil nutrient cycling, Vehicle NO_x and NH_3 emissions, Soil respiration, Nitrogen cycle, Urban biogeochemistry

Co-digestion of rice straw and cow dung to supply cooking fuel and fertilizers in rural India: Impact on human health, resource flows and climate change

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Source: Volume 609, 31 December 2017, Pages 1600-1615

<https://doi.org/10.1016/j.scitotenv.2017.07.150>

Anaerobic digestion of cow dung with new feedstock such as crop residues to increase the biogas potential is an option to help overcoming several issues faced by India. Anaerobic digestion provides biogas that can replace biomass cooking fuels and reduce indoor air pollution. It also provides digestate, a fertilizer that can contribute to compensate nutrient shortage on agricultural land. Moreover, it avoids the burning of rice straw in the fields which contributes to air pollution in India and climate change globally. Not only the technical and economical feasibility but also the environmental sustainability of such systems needs to be assessed. The potential effects of implementing community digesters co-digesting cow dung and rice straw on carbon and nutrients flows, human health, resource efficiency and climate change are analyzed by conducting a Substance Flow Analysis and a Life Cycle Assessment. The implementation of the technology is considered at the level of the state of Chhattisgarh. Implementing this scenario reduces the dependency of the rural community to nitrogen and phosphorus from synthetic fertilizers only by 0.1 and 1.6%, respectively, but the dependency of farmers to potassium from synthetic fertilizers by 31%. The prospective scenario returns more organic carbon to agricultural land and thus has a potential positive effect on soil quality. The implementation of the prospective scenario can reduce the health impact of the local population by 48%, increase the resource efficiency of the system by 60% and lower the impact on climate change by 13%. This study highlights the large potential of anaerobic digestion to overcome the aforementioned issues faced by India. It demonstrates the need to couple local and global assessments and to conduct analyses at the substance level to assess the sustainability of such systems.

Keywords : Agricultural waste, Biogas, Nutrients, Substance Flow Analysis, Life Cycle Assessment

Characteristics of black carbon in snow from Laohugou No. 12 glacier on the northern Tibetan Plateau

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Source: *Volumes 607–608, 31 December 2017, Pages 1237-1249*

<https://doi.org/10.1016/j.scitotenv.2017.07.100>

Black carbon (BC) emitted from the incomplete combustion of biomass and fossil fuel impacts the climate system, cryospheric change, and human health. This study documents black carbon deposition in snow from a benchmark glacier on the northern Tibetan Plateau. Significant seasonality of BC concentrations indicates different input or post-depositional processes. BC particles deposited in snow had a mass volume median diameter slightly larger than that of black carbon particles typically found in the atmosphere. Also, unlike black carbon particles in the atmosphere, the particles deposited in snow did not exhibit highly fractal morphology by Scanning Transmission Electron Microscope. Footprint analysis indicated BC deposited on the glacier in summer originated mainly from Central Asia; in winter, the depositing air masses generally originated from Central Asia and Pakistan. Anthropogenic emissions play an important role on black carbon deposition in glacial snow, especially in winter. The mass absorption efficiency of BC in snow at 632 nm exhibited significantly seasonality, with higher values in summer and lower

values in winter. The information on black carbon deposition in glacial snow provided in this study could be used to help mitigate the impacts of BC on glacier melting on the northern Tibetan Plateau.

Keywords : Black carbon, BC size, BC morphology, Glacier, Tibetan Plateau

Temporal trends of surface urban heat islands and associated determinants in major Chinese cities

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Source: Volume 609, 31 December 2017, Pages 742-754

<https://doi.org/10.1016/j.scitotenv.2017.07.217>

There are many studies focusing on spatial variations of surface urban heat islands (SUHIs) in literature. In this study, MODIS land surface temperature (LST) data and China's Land Use/Cover Datasets (CLUDs) were used to examine the temporal trends of SUHIs in 31 major Chinese cities during 2001–2015 using three indicators: SUHI intensity (SUHII), area of the SUHI (AreaSUHI) and percentage of area with increasing SUHII (PAISUHII). Correlation analyses between SUHII and background (rural) LST (extracted from MODIS LST), vegetation coverage (reflected by MODIS EVI data) and anthropogenic heat release (reflected by nighttime light data) were performed from temporal rather than spatial perspectives. Our findings showed that the SUHII and AreaSUHI in urbanized areas increased significantly in most cities in summer days, whereas they increased significantly in approximately half and more than half of the cities in summer and winter nights, respectively. In summer days, summer nights and winter nights, the PAISUHII was approximately 80% and over 50% in urban areas and the 20 km buffer, respectively. Correlation analyses indicated that the SUHII in stable urban areas was negatively correlated with the background LST in summer and winter days for most cities, especially in northern China. A reduction in vegetation contributed to the increasing SUHII in urbanized areas in summer days and nights. The increasing anthropogenic heat release was an important factor for increases in the SUHII in urbanized areas.

Keywords : Surface urban heat island, Urbanization, Temporal trend, China

Projections of NH₃ emissions from manure generated by livestock production in China to 2030 under six mitigation scenarios

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Source: Volumes 607–608, 31 December 2017, Pages 78-86

<https://doi.org/10.1016/j.scitotenv.2017.06.258>

China's rapid urbanization, large population, and increasing consumption of calorie- and meat-intensive diets, have resulted in China becoming the world's largest source of ammonia (NH₃) emissions from livestock production. This is the first study to use provincial, condition-specific

emission factors based on most recently available studies on Chinese manure management and environmental conditions. The estimated NH₃ emission temporal trends and spatial patterns are interpreted in relation to government policies affecting livestock production. Scenario analysis is used to project emissions and estimate mitigation potential of NH₃ emissions, to year 2030. We produce a 1 km × 1 km gridded NH₃ emission inventory for 2008 based on county-level activity data, which can help identify locations of highest NH₃ emissions. The total NH₃ emissions from manure generated by livestock production in 2008 were 7.3 Tg NH₃·yr⁻¹ (interquartile range from 6.1 to 8.6 Tg NH₃·yr⁻¹), and the major sources were poultry (29.9%), pigs (28.4%), other cattle (27.9%), and dairy cattle (7.0%), while sheep and goats (3.6%), donkeys (1.3%), horses (1.2%), and mules (0.7%) had smaller contributions. From 1978 to 2008, annual NH₃ emissions fluctuated with two peaks (1996 and 2006), and total emissions increased from 2.2 to 7.3 Tg·yr⁻¹ increasing on average 4.4%·yr⁻¹. Under a business-as-usual (BAU) scenario, NH₃ emissions in 2030 are expected to be 13.9 Tg NH₃·yr⁻¹ (11.5–16.3 Tg NH₃·yr⁻¹). Under mitigation scenarios, the projected emissions could be reduced by 18.9–37.3% compared to 2030 BAU emissions. This study improves our understanding of NH₃ emissions from livestock production, which is needed to guide stakeholders and policymakers to make well informed mitigation decisions for NH₃ emissions from livestock production at the country and regional levels.

Keywords : Livestock manure, Ammonia emissions, Policies analysis, Uncertainty, Mitigation scenarios

Temporal and spatial distribution of PM_{2.5} chemical composition in a coastal city of Southeast China

Author links open overlay

panelMengrenLiaMinHuacBohanDubQingfengGuoaTianyiTanaJingZhengaXiaofengHuangbL
ingyanHebZhijunWuaSongGuoa

Source: *Volumes 605–606, 15 December 2017, Pages 337–346*

<https://doi.org/10.1016/j.scitotenv.2017.03.260>

Rapid economic development and urbanization in China has been concentrated in coastal cities, resulting in haze and photochemical smog issues, especially in the densely-populated Yangtze River Delta. In this study, we explore particulate matter (specifically PM_{2.5}) pollution in a city in Zhejiang Province (Ningbo), chosen to represent a typical, densely-populated urban city with residential and industrial sections. PM_{2.5} samples were collected at five sites in four seasons from Dec. 2012 to Nov. 2013. The annual average PM_{2.5} mass concentration was $53.2 \pm 30.4 \mu\text{g}/\text{m}^3$, with the highest concentration in winter and lowest in summer. Among the five sites, PM_{2.5} concentration was highest in an urban residential site and lowest in a suburban site, due to effects of urbanization and the anthropogenic influences. The chemical components of PM_{2.5} show significant seasonal variation. In addition, secondary transformation was high in Ningbo, with the highest proportion of secondary components found at a suburban site and the lowest at the industrial sites. Ningbo is controlled by five major air masses originating from inland China, from the Bohai Sea, offshore from the southeast, the Yellow Sea, and off the east coast of Korea. The relative contributions of these air masses differ, by season, with the Bohai Sea air mass dominating in winter and spring, the maritime southeast air mass in summer, and the Yellow Sea and coastal Korean air masses dominating in autumn. The continental air mass is associated with a high PM_{2.5} concentration, indicating that it is

primarily transports primary emissions. In contrast, the concentration ratios among secondary formed pollutants were higher in the maritime air masses, which suggests that sea breezes control temporal and spatial variations of air pollution over coastal cities.

Keywords : Chemical components, Temporal and spatial distribution, Secondary formation, Coastal city

Source apportionment of PM₁₀ and PM_{2.5} in major urban Greek agglomerations using a hybrid source-receptor modeling process

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Source: Volumes 601–602, 1 December 2017, Pages 906-917

<https://doi.org/10.1016/j.scitotenv.2017.05.088>

A hybrid source-receptor modeling process was assembled, to apportion and infer source locations of PM₁₀ and PM_{2.5} in three heavily-impacted urban areas of Greece, during the warm period of 2011, and the cold period of 2012. The assembled process involved application of an advanced computational procedure, the so-called Robotic Chemical Mass Balance (RCMB) model. Source locations were inferred using two well-established probability functions: (a) the Conditional Probability Function (CPF), to correlate the output of RCMB with local wind directional data, and (b) the Potential Source Contribution Function (PSCF), to correlate the output of RCMB with 72 h air-mass back-trajectories, arriving at the receptor sites, during sampling. Regarding CPF, a higher-level conditional probability function was defined as well, from the common locus of CPF sectors derived for neighboring receptor sites. With respect to PSCF, a non-parametric bootstrapping method was applied to discriminate the statistically significant values. RCMB modeling showed that resuspended dust is actually one of the main barriers for attaining the European Union (EU) limit values in Mediterranean urban agglomerations, where the drier climate favors build-up. The shift in the energy mix of Greece (caused by the economic recession) was also evidenced, since biomass burning was found to contribute more significantly to the sampling sites belonging to the coldest climatic zone, particularly during the cold period. The CPF analysis showed that short-range transport of anthropogenic emissions from urban traffic to urban background sites was very likely to have occurred, within all the examined urban agglomerations. The PSCF analysis confirmed that long-range transport of primary and/or secondary aerosols may indeed be possible, even from distances over 1000 km away from study areas.

Keywords : Particulate Matter, Southeastern Europe, Robotic Chemical Mass Balance (RCMB), Conditional Probability Field (CPF), Potential Source Contribution Function (PSCF), Non-parametric bootstrapping

Chemical characteristics and source apportionment of PM_{2.5} in Lanzhou, China

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Source: Volumes 601–602, 1 December 2017, Pages 1743–1752

<https://doi.org/10.1016/j.scitotenv.2017.06.050>

Daily PM_{2.5} samples were collected during winter 2012 and summer 2013 at an urban site in Lanzhou and were analyzed for chemical compounds including water soluble inorganic ions (WSIN), trace elements, water soluble organic carbon (WSOC), carbonaceous species (OC/EC), polycyclic aromatic hydrocarbons (PAHs), and humic-like substances (HULIS). The seasonal-average reconstructed PM_{2.5} mass was 120.5 $\mu\text{g m}^{-3}$ in winter and 34.1 $\mu\text{g m}^{-3}$ in summer. The top three groups of species in PM_{2.5} were OC ($35.4 \pm 13.9 \mu\text{g m}^{-3}$), WSIN ($34.89 \pm 14.21 \mu\text{g m}^{-3}$), and EC ($13.80 \pm 5.41 \mu\text{g m}^{-3}$) in winter and WSIN ($11.25 \pm 3.25 \mu\text{g m}^{-3}$), OC ($9.74 \pm 3.30 \mu\text{g m}^{-3}$), and EC ($4.44 \pm 2.00 \mu\text{g m}^{-3}$) in summer. EC exceeded SO₄²⁻ – on most of the days. Several anthropogenic produced primary pollutants such as PAHs, Cl⁻, Pb, Cd and OC_{pri} were 4–22 times higher in winter than summer. Carcinogenic substances such as Arsenic, BaP, Pb, and Cd in PM_{2.5} exceeded the WHO guideline limits by 274%, 153%, 23% and 7%, respectively. Positive Matrix Factorization analysis identified seven source factors including steel industry, secondary aerosols, coal combustion, power plants, vehicle emissions, crustal dust, and smelting industry, which contributed 7.1%, 33.0%, 28.7%, 3.12%, 8.8%, 13.3%, and 6.0%, respectively, to PM_{2.5} in winter, and 6.7%, 14.8%, 3.1%, 3.4%, 25.2%, 11.6% and 35.2% in summer. Smelting industry and steel industry were identified for the first time as sources of PM_{2.5} in this city, and power plant was distinguished from industrial boiler and residential coal burning.

Keywords : Atmospheric aerosols, Fine particulate matter, Chemical composition, Source apportionment analysis

Spatio-temporal analysis of particulate matter intake fractions for vehicular emissions: Hourly variation by micro-environments in the Greater Toronto and Hamilton Area, Canada

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Source: Volumes 599–600, 1 December 2017, Pages 1813–1822

<https://doi.org/10.1016/j.scitotenv.2017.05.134>

Previous investigations have reported intake fraction (iF) for different environments, which include ambient concentrations (outdoor exposure) and microenvironments (indoor exposure). However, little is known about iF variations due to space-time factors, especially in microenvironments. In this paper, we performed a spatio-temporal analysis of particulate matter (PM_{2.5}) intake fractions for vehicular emissions. Specifically, we investigated hourly variation (12:00 am–11:00 pm) by

micro-environments (residences and workplaces) in the Greater Toronto and Hamilton Area (GTHA), Canada. We used GIS modeling to estimate air pollution data (ambient concentration, and traffic emission) and population data in each microenvironment. Our estimates showed that the total iF at residences and workplaces accounts for 85% and 15%, respectively. Workplaces presented the highest 24 h average iF (1.06 ppm), which accounted for 25% higher than residences. Observing the iF by hour at residences, our estimates showed the highest average iF at 2:00 am (iF = 3.72 ppm). These estimates indicate that approximately 4 g of PM_{2.5} emitted from motor vehicles are inhaled for every million grams of PM_{2.5} emitted. For the workplaces, the highest exposure was observed at 10:00 am, with average iF equal to 2.04 ppm. The period of the day with the lower average iF for residences was at 8:00 am (average iF = 0.11 ppm), while for the workplaces was at 4:00 am (average iF = 0.47 ppm). Our approach provides a new perspective on human exposure to air pollution. Our results showed significant hourly variation in iF across the GTHA. Our findings can be incorporated in future investigations to advance environmental health effects research and human health risk assessment.

Keywords : Particulate matter, Vehicular emissions, Intake fractions, Spatio-temporal analysis

Meteorological factors had more impact on airborne bacterial communities than air pollutants

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Source: Volumes 601–602, 1 December 2017, Pages 703-712

<https://doi.org/10.1016/j.scitotenv.2017.05.049>

Airborne bacteria have gained increasing attention because they affect ecological balance and pose potential risks on human health. Recently, some studies have focused on the abundance and composition of airborne bacteria under heavy, hazy polluted weather in China, but they reached different conclusions about the comparisons with non-polluted days. In this study, we tested the hypothesis that meteorological factors could have a higher impact on shaping airborne bacterial communities than air pollutants by systematically monitoring the communities for 1 year. Total suspended particles in Beijing were sampled for 20 consecutive days in each season of 2015. Bacterial abundance varied from 8.71×10^3 to 2.14×10^7 ribosomal operons per cubic meter according to the quantitative PCR analysis. There were relatively higher bacterial counts in spring and in autumn than in winter and summer. Airborne bacterial communities displayed a strong seasonality, according to the hierarchical cluster analysis. Only two exceptions overtook the seasonal trend, and both occurred in or after violent meteorological changes (sandstorm or rain). Aggregated boosted tree analysis performed on bacterial abundance showed that the dominant factors shaping bacterial communities were meteorological. They were air pressure in winter, air temperature and relative humidity in spring, RH in summer, and vapor pressure in autumn. Variation partition analysis on community structure showed that meteorological factors explained more variations than air pollutants. Therefore, both of the two models verified our hypothesis that the differences in airborne bacterial communities in polluted days or non-polluted days were mainly driven by the discrepancies of meteorological factors rather than by the presence of air pollutants.

Keywords : Abundance, Air pollutants, Airborne bacterial community, Meteorological factors

A one-year, on-line, multi-site observational study on water-soluble inorganic ions in PM_{2.5} over the Pearl River Delta region, China

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Source: Volumes 601–602, 1 December 2017, Pages 1720-1732

<https://doi.org/10.1016/j.scitotenv.2017.06.039>

As the significant components of PM_{2.5}, almost all of previous studies on water-soluble inorganic ions (WSIIs) have been limited by the use of single sampling station, short sampling times or low temporal resolution. This paper focuses on analysing one-year (2012) observations of WSIs at a regional central (RCEN) site, a coastal urban (CURB) site and a coastal rural (CRUR) site in the Pearl River Delta region. On average, secondary inorganic aerosols (SIA) were the most abundant component and accounted for over 80% of the total WSIs. The ratio among sulfate, nitrate and ammonium mass concentrations was close to 2:1:1 (5:2:1) at the RCEN and CURB sites (CRUR site). Most components (except Na⁺) showed higher concentrations in the dry season. The diurnal variations of different ions showed obvious differences, which were partially controlled by photochemical reactions and diffusion conditions in the boundary layer. Ionic formation patterns were different among the three sites. Secondary inorganic pollution was much more serious in the northwestern PRD, and it had a significant effect on pollution in the coastal areas. High SO₄²⁻ concentrations at the CRUR site may be associated with local emissions, such as dimethylsulfide (DMS). Long-range transport along the southeastern coastline also played an important role in SO₄²⁻ pollution over the PRD region. Sea salt aerosols were an important source in coastal regions; they contributed large amounts of Cl⁻, Na⁺, Mg²⁺ at the CRUR site and large amounts of Na⁺, Mg²⁺ at the CURB site. The case studies found that sea salt aerosols concentrations increased obviously during the heavy precipitation period of typhoon. The presence of warm-wet air masses before continuous moist weather (CMW) was favourable for the formation of SIA. On the other hand, during CMW periods, SIA concentrations decreased rapidly.

Keywords : Temporal variations, Potential pollution sources, Special weather processes, MARGA

Refined 2013-based vehicle emission inventory and its spatial and temporal characteristics in Zhengzhou, China

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Source: Volumes 599–600, 1 December 2017, Pages 1149-1159

<https://doi.org/10.1016/j.scitotenv.2017.03.299>

Vehicle emission is becoming one of the most important pollution sources because of the increase in vehicle population and activity in China. A more reasonable and complete vehicle emission

inventory in Zhengzhou for the year 2013 was developed in this study. This inventory is suitable for local emission factors and vehicle kilometers of travel. Estimates show that the total carbon monoxide (CO), hydrocarbon (HC), nitrogen oxide (NOX), particulate matter (PM2.5 and PM10) and sulfur dioxide (SO2) emissions were 291 Gg, 35 Gg, 106 Gg, 6 Gg, 7 Gg, and 3 Gg, respectively. Approximately 55% of CO and HC emissions were from light duty gasoline vehicles and normal gasoline motorcycles, whereas approximately 60% of NOX, PM2.5, PM10 and SO2 were from heavy duty diesel vehicles, heavy duty diesel trucks, and medium duty diesel trucks. The spatial distribution of emissions was allocated in grid cells based on a road network and traffic flows with a resolution of 1 km × 1 km at different road types and locations, which shows that the six aforementioned air pollutants have similar characteristics in administrative districts. Emissions are mainly concentrated on the central grid cells of each part and in good agreement with line sources. The spatial characteristics were compared at a resolution of 3 km × 3 km and in a population-based approach. The network approach yields better level estimates in this study. Meanwhile, the preliminary temporal profiles were also established for on-road mobile source.

Keywords : Diagnosis, Traffic emission, Spatial distribution, Grid-based, Air quality

Impact of dust and smoke mixing on column-integrated aerosol properties from observations during a severe wildfire episode over Valencia (Spain)

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Source: Volumes 599–600, 1 December 2017, Pages 2121-2134

<https://doi.org/10.1016/j.scitotenv.2017.05.041>

The most destructive wildfire experienced in Spain since 2004 occurred close to Valencia in summer 2012. A total of 48.500 ha were affected by two wildfires, which were mostly active during 29–30 June. The fresh smoke plume was detected at the Burjassot measurement station simultaneously to a severe dust episode. We propose an empirical method to evaluate the dust and smoke mixing and its impact on the microphysical and optical properties. For this, we combine direct-sun measurements with a Cimel CE-318 sun-photometer with an inversion methodology, and the Mie theory to derive the column-integrated size distribution, single scattering albedo (SSA) and asymmetry parameter (g). The mixing of dust and smoke greatly increased the aerosol load and modified the background aerosol properties. Mineral dust increased the aerosol optical depth (AOD) up to 1, while the smoke plume caused an extreme AOD peak of 8. The size distribution of the mixture was bimodal, with a fine and coarse modes dominated by the smoke particles and mineral dust, respectively. The SSA and g for the dust-smoke mixture show a marked sensitivity on the smoke mixing-ratio, mainly at longer wavelengths. Mineral dust and smoke share a similar SSA at 440 nm (~ 0.90), but with opposite spectral dependency. A small dust contribution to the total AOD substantially affects the SSA of the mixture, and also SSA at 1020 nm increases from 0.87 to 0.95. This leads to a different spectral behaviour of SSA that changes from positive (smoke plume) to negative (dust), depending on the dust and smoke mixing-ratio.

Keywords : Wildfire, Dust and smoke mixing, Aerosol properties

Acid rain footprint three decades after peak deposition: Long-term recovery from pollutant sulphate in the Uhlirská catchment (Czech Republic)

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Source: Volume 598, 15 November 2017, Pages 1037-1049

<https://doi.org/10.1016/j.scitotenv.2017.04.109>

The granitic Uhlirská headwater catchment with a size of 1.78 km² is located in the Jizera Mountains in the northern Czech Republic and received among the highest inputs of anthropogenic acid depositions in Europe. An analysis of sulphate (SO₄²⁻) distribution in deposition, soil water, stream water and groundwater compartments allowed to establish a SO₄²⁻ mass-balance (deposition input minus surface water export) and helped to evaluate which changes occurred since the last evaluation of the catchment in 1997. The determined SO₄²⁻ concentrations decreased in the following order: wetland groundwater > groundwater from 20 m below ground level (bgl) > groundwater from 30 m bgl > stream water > groundwater from 10 m bgl > hillslope soil water > wetland soil water > bulk deposition with median values of 0.24, 0.21, 0.17, 0.15, 0.11, 0.07, 0.03 and 0.01 mmol L⁻¹, respectively. Our results show that average deposition reductions of 62% did not result in equal changes of the sulphate mass-balance, which changed by only 47%. This difference occurs because sulphate originates from internal sources such as the groundwater and soil water. The Uhlirská catchment is subject to delayed recovery from anthropogenic acid depositions and remains a net source of stored sulphur even after three decades of declining inputs. The wetland groundwater and soil water provide environmental memories of legacy pollutant sulphate. Elevated stream water sulphate concentrations after the unusually dry summer 2015 imply importance of weather and climate patterns for future recovery from acidification.

Keywords : Mass balance, Export, Catchment, Soil water, Groundwater, Sulphur

Impacts of Himalayas on black carbon over the Tibetan Plateau during summer monsoon

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Source: Volume 598, 15 November 2017, Pages 307-318

<https://doi.org/10.1016/j.scitotenv.2017.04.101>

The Tibetan Plateau (TP) plays important roles in global climate and environment. This study combines in-situ BC measurements in the Himalayas and the Indo-Gangetic Plain (IGP) with a regional dynamical and chemical model (WRF-Chem model) to investigate the effect of the trans-

Himalayas on black carbon (BC) from the IGP to the TP during Indian summer monsoon. To determine topographic effects of the trans-Himalayas on BC concentrations over the TP, sensitive experiments were conducted by applying the WRF-Chem model. The results showed that the reduction of the altitude of the Himalayas had an important effect on the trans-Himalayas transport of BC. There was an obvious increase in BC concentration over the trans-Himalayas region, but no significant increase over the TP because the TP (a.m.s.l ~ 4 km) always acted as a wall to prevent BC transport from the IGP to the TP. The trans-Himalayas transport of BC was strongly dependent upon meteorological conditions over the IGP. During summer monsoon, there were three types of cyclones at different locations and one kind of convergent circulation in the IGP. Under the condition of convergent airflows, a strong northeastward wind produced the trans-Himalayas transport of BC. As a result, BC concentrations in the southeastern TP significantly increased to $0.6\text{--}0.8\text{ }\mu\text{g m}^{-3}$. When the cyclone located in the eastern IGP, high BC concentrations over the IGP were transported along the foothill of the Himalayas, resulting in a significant reduction of the trans-Himalayas transport. When the cyclone moved to the west, the dynamical perturbations for the trans-Himalayas transport were weaker than the eastern cyclone, and the trans-Himalayas transport were enhanced in the middle and eastern Himalayas. This study will be helpful to assess the impacts of BC particles emitted from South Asia on regional climate change and ecological environment over the TP in the future.

Keywords : Black carbon, Monsoon, The Himalayas and Tibetan Plateau, WRF-Chem model

Airborne dust absorption by semi-arid forests reduces PM pollution in nearby urban environments

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Source: Volume 598, 15 November 2017, Pages 984-992

<https://doi.org/10.1016/j.scitotenv.2017.04.162>

Dust storms are a major source of global atmospheric particulate matter (PM), having significant impacts on air pollution and human health. During dust storms, daily averages of atmospheric PM concentrations can reach high levels above the World Health Organization (WHO) guideline for air quality. The objective of this study was to explore the impact of forests on PM distribution following dust events in a region that is subjected to frequent dust storms (Northern Negev, Israel). Dust was measured in a forest transect including urban environments that are nearby the forest and at a distal location. During a background period, without dust events, the forest with its surrounding areas were characterized by lower monthly average of PM concentrations ($38\text{ }\mu\text{g}/\text{m}^3$) compared with areas that are not affected by the forest ($54\text{ }\mu\text{g}/\text{m}^3$). Such difference can be meaningful for long-term human health exposure. A reduction in PM levels in the forest transect was evident at most measured dust events, depending on the storm intensity and the locations of the protected areas. A significant reduction in PM_{2.5}/PM₁₀ during dust events, indicates the high efficiency of the forest trees to absorb airborne PM_{2.5}. Analysis of dust particles absorbed on the foliage revealed a total dust deposits of $8.1\text{--}9.2\text{ g}/\text{m}^2$, which is equal to a minimum of 418.2 tons removed from the atmosphere per a forest foliage area (30 km^2). The findings can support environmental strategies

to enhance life quality in regions that are subjected to dust storms, or under potential risk of dust-related PM due to land use and/or climate changes.

Keywords : Particulate matter, Air quality, Dust deposition, Ecosystem service, Atmospheric dust, Forest

Review and analysis of strengths and weaknesses of agro-ecosystem models for simulating C and N fluxes

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Source: Volume 598, 15 November 2017, Pages 445-470
<https://doi.org/10.1016/j.scitotenv.2017.03.208>

Biogeochemical simulation models are important tools for describing and quantifying the contribution of agricultural systems to C sequestration and GHG source/sink status. The abundance of simulation tools developed over recent decades, however, creates a difficulty because predictions from different models show large variability. Discrepancies between the conclusions of different modelling studies are often ascribed to differences in the physical and biogeochemical processes incorporated in equations of C and N cycles and their interactions. Here we review the literature to determine the state-of-the-art in modelling agricultural (crop and grassland) systems. In order to carry out this study, we selected the range of biogeochemical models used by the CN-MIP consortium of FACCE-JPI (<http://www.faccejpi.com>): APSIM, CERES-EGC, DayCent, DNDC, DSSAT, EPIC, PaSim, RothC and STICS. In our analysis, these models were assessed for the quality and comprehensiveness of underlying processes related to pedo-climatic conditions and management practices, but also with respect to time and space of application, and for their accuracy in multiple contexts. Overall, it emerged that there is a possible impact of ill-defined pedo-climatic conditions in the unsatisfactory performance of the models (46.2%), followed by limitations in the algorithms simulating the effects of management practices (33.1%). The multiplicity of scales in both time and space is a fundamental feature, which explains the remaining weaknesses (i.e. 20.7%). Innovative aspects have been identified for future development of C and N models. They include the explicit representation of soil microbial biomass to drive soil organic matter turnover, the effect of N shortage on SOM decomposition, the improvements related to the production and consumption of gases and an adequate simulations of gas transport in soil. On these bases, the assessment of trends and gaps in the modelling approaches currently employed to represent biogeochemical cycles in crop and grassland systems appears an essential step for future research.

Keywords : Biogeochemical models, C cycle, N cycle, Management, Pedo-climate

Substantial air quality and climate co-benefits achievable now with sectoral mitigation strategies in China

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Source: Volume 598, 15 November 2017, Pages 1076-1084

<https://doi.org/10.1016/j.scitotenv.2017.03.287>

China is the world's top carbon emitter and suffers from severe air pollution. We examine near-term air quality and CO₂ co-benefits of various current sector-based policies in China. Using a 2015 base case, we evaluate the potential benefits of four sectoral mitigation strategies. All scenarios include a 20% increase in conventional air pollution controls as well as the following sector-specific fuel switching or technology upgrade strategies. Power sector (POW): 80% replacement of small coal power plants with larger more efficient ones; Industry sector (IND): 10% improvement in energy efficiency; Transport sector (TRA): replacement of high emitters with average vehicle fleet emissions; and Residential sector (RES): replacement of 20% of coal-based stoves with stoves using liquefied petroleum gas (LPG). Conducting an integrated assessment using the regional air pollution model WRF-Chem, we find that the IND scenario reduces national air-pollution-related deaths the most of the four scenarios examined (27,000, 24,000, 13,000 and 23,000 deaths reduced annually in IND, POW, TRA and RES, respectively). In addition, the IND scenario reduces CO₂ emissions more than 8 times as much as any other scenario (440, 53, 0 and 52 Mt CO₂ reduced in IND, POW, TRA and RES, respectively). We also examine the benefits of an industrial efficiency improvement of just 5%. We find the resulting air quality and health benefits are still among the largest of the sectoral scenarios, while the carbon mitigation benefits remain more than 3 times larger than any other scenario. Our analysis hence highlights the importance of even modest industrial energy efficiency improvements and air pollution control technology upgrades for air quality, health and climate benefits in China

Keywords : Air pollution, Climate change, Co-benefits, Sectoral mitigation, Industrial energy efficiency, WRF-Chem

Ozone exposure affects tree defoliation in a continental climate

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Source: Volumes 596–597, 15 October 2017, Pages 396-404

<https://doi.org/10.1016/j.scitotenv.2017.03.135>

Ground-level ozone (O₃) affects trees through visible leaf injury, accelerating leaf senescence, declining foliar chlorophyll content, photosynthetic activity, growth, carbon sequestration, predisposing to pests attack and a variety of other physiological effects. Tree crown defoliation is one of the most important parameters that is representative of forest health and vitality. Effects of

air pollution on forests have been investigated through manipulative experiments that are not representative of the real environmental conditions observed in the field. In this work we investigated the role of O₃ concentration and other metrics (AOT40 and POD0) in affecting crown defoliation in temperate Romanian forests. The impacts of O₃ were estimated in combination with nitrogen pollutants, climatic factors and orographic conditions, by applying a non-linear modelling approach (Random Forest and Generalised Regression Models). Ozone concentration and AOT40 under Romanian conditions were more important than meteorological parameters in affecting crown defoliation. In these particular conditions, POD0 never exceeded the critical level suggested by previous literature for forest protection, and thus was not important in affecting crown defoliation.

Keywords : Air pollution impacts, Crown transparency, Stomatal ozone uptake, Forests, General regression models

Source-receptor relationships for PM_{2.5} during typical pollution episodes in the Pearl River Delta city cluster, China

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Source: Volumes 596–597, 15 October 2017, Pages 194-206

<https://doi.org/10.1016/j.scitotenv.2017.03.255>

Located in the Southern China monsoon region, pollution days in Pearl River Delta (PRD) were classified into “Western type”, “Central type” or “Eastern type”, with a relative percentage of 67%, 24% and 9%, respectively. Using this classification system, three typical pollution events were selected for numerical simulations using the WRF-Chem model. The source sensitivity method for anthropogenic emissions of PM_{2.5} and its precursors was applied to identify the source-receptor relationships for PM_{2.5} among 9 cities in PRD. For “Western type” case, the PRD region was under control of a high-pressure system with easterly prevailing winds. The PM_{2.5} concentrations in the western PRD region were higher than those in the eastern region, with emissions from cities in the eastern PRD region having higher contributions. Within the PRD’s urban cluster, PM_{2.5} in Huizhou, Dongguan and Shenzhen was mainly derived from local emissions, whereas the PM_{2.5} in the other cities was primarily derived from external transport. For “Eastern type” case, the PRD was influenced by Typhoon Soulik with westerly prevailing winds. Emissions from cities in the western PRD region had the highest impacts on the overall PM_{2.5} concentration. PM_{2.5} in Jiangmen and Foshan was primarily derived from local emissions. Regarding “Central type” case, the PRD region was under control of a uniform pressure field with low wind speed. PM_{2.5} concentrations of each city were primarily caused by local emissions. Overall, wind flows played a significant role in the transport and spatial distribution of PM_{2.5} across the PRD region. Ideally, local governments would be wise to establish joint prevention and control measures to reduce regional atmospheric pollution, especially for “Western type” pollution.

Keywords : Regional transport, Source-receptor relationships, Contribution, WRF-Chem

Biomass burning sources and their contributions to the local air quality in Hong Kong

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Source: Volumes 596–597, 15 October 2017, Pages 212–221
<https://doi.org/10.1016/j.scitotenv.2017.04.091>

In this paper, we present a quantitative estimation of the impacts of biomass burning emissions from different source regions to the local air quality in Hong Kong in 2014 using global chemistry transport model simulations, sun photometer measurements, satellite observations and local monitoring network data. This study focuses on two major biomass burning pollutants, black carbon aerosols and carbon monoxide (CO). The model simulations of atmospheric black carbon and CO show excellent agreement with sun photometer aerosol optical depth (AOD) measurements, satellite CO columns observations and local monitoring stations data. From the model simulation results, we estimated that biomass burning contributes 12% of total black carbon and 16% of atmospheric CO in Hong Kong on annual average. South East Asia shows the largest influence to the black carbon and CO levels in Hong Kong, accounts for 11% of the total atmospheric black carbon and 8% of CO. Biomass burning in North East Asia and Africa also show significant impacts to Hong Kong. Elevated levels of atmospheric black carbon aerosols and CO were observed during springtime (March and April) which is mainly due to the enhancement of biomass burning contributions. Black carbon and CO originating from biomass burning sources are estimated to contribute 40% of atmospheric black carbon and 28% of CO in Hong Kong during March 2014. An investigation focusing on the biomass burning pollution episode during springtime suggests the intensified biomass burning activities in the Indochinese Peninsula are the major sources of black carbon and CO in Hong Kong during the time.

Keywords : Biomass burning, Source contributions, Long range transport, Black carbon, Carbon monoxide

Atmospheric removal of PM_{2.5} by man-made Three Northern Regions Shelter Forest in Northern China estimated using satellite retrieved PM_{2.5} concentration

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Source: Volumes 593–594, 1 September 2017, Pages 713–721
<https://doi.org/10.1016/j.scitotenv.2017.03.206>

Atmospheric removal of PM_{2.5} by the Three Northern Regions Shelter Forest (TNRSF) – the so called Green Great Wall (GGW) in northern China through dry deposition process was estimated using a bulk big-leaf model and a vegetation collection model. Decadal trend of PM_{2.5} dry deposition flux from 1999 to 2010 was calculated from modeled dry deposition velocity and air concentration retrieved from the satellite remote sensing. Dry deposition velocities of PM_{2.5} calculated using the two deposition models increased in many places of the TNRSF over the last decade due to increasing vegetation coverage of the TNRSF. Both increasing deposition velocity

due to forest expansion and PM_{2.5} atmospheric level contributed to the increasing deposition flux of PM_{2.5}. The highest atmospheric deposition flux of PM_{2.5} was found in the Central-north region covering Beijing-Tianjin-Hebei area, followed by the Northwestern and the Northeastern regions of the TNRSF. While greater collection of PM_{2.5} by vegetation was identified in the Northeastern region of the TNRSF due to higher forest coverage over this region, the most significant incline of the PM_{2.5} atmospheric removal due to vegetation collection was discerned in the Central-north region because of the most rapid increase in the vegetation coverage in this region. A total mass of 2.85×10^7 t PM_{2.5} was estimated to be removed from the atmosphere through dry deposition process over the TNRSF from 1999 to 2010. The two deposition models simulated similar magnitude and spatial patterns of PM_{2.5} dry deposition fluxes. Our results suggest that the TNRSF plays a moderate role in PM_{2.5} uptake, but enhances PM_{2.5} atmospheric removal by 30% in 2010 than in 1980.

Keywords : Dry deposition, Model intercomparison, Trend analysis, Vegetation collection

Physicochemical characterization of ambient PM_{2.5} in Tehran air and its potential cytotoxicity in human lung epithelial cells (A549)

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Source: Volumes 593–594, 1 September 2017, Pages 182-190
<https://doi.org/10.1016/j.scitotenv.2017.03.150>

As air pollution is a major problem in Tehran, this study aimed to investigate the physicochemical characterization of the water-soluble and organic contents of ambient PM_{2.5} in Tehran and determine its in vitro toxicological impact on human lung epithelial cells (A549). A total of 11 sampling stations were selected, including three categories: traffic, urban, and suburban. All sampling was carried out in the spring and summer of 2015. Ion chromatography (IC), inductively coupled plasma atomic emission spectroscopy (ICP-AES), and GC-MS were used to analyze ionic compounds, heavy metals, and polycyclic aromatic hydrocarbons (PAHs), respectively, and an ELISA reader was used for cytotoxicity analysis. The most prevalent ionic species found for all three categories was SO₄²⁻. PAH concentrations were 43.45 ± 32.71 , 50.51 ± 37.27 , and 29.13 ± 33.29 ng/m³ for traffic, urban, and suburban stations, respectively. For all sampling stations, Al and Fe had the highest values among the investigated heavy metals. Cell viability measurements, carried out using the MTT assay, showed that all three categories of samples cause cytotoxicity, although the urban station samples showed higher cytotoxicity than those from the other stations ($p < 0.05$). Based on the results of the present study, organic compounds and insoluble particles could be the main causes of cytotoxicity.

Keywords : Cytotoxicity, Lung epithelial cells (A549), MTT assay

Temporal patterns of CO₂ and CH₄ in a rural area in northern Spain described by a harmonic equation over 2010–2016

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Source: Volumes 593–594, 1 September 2017, Pages 1–9
<https://doi.org/10.1016/j.scitotenv.2017.03.132>

The present paper seeks to improve our knowledge concerning the evolution of CO₂ and CH₄ in terms of monthly trends, growth rate and seasonal variations in the lower atmosphere. Dry continuous measurements of CO₂ and the CH₄ mixing ratio were carried out over five and a half years (from 15 October 2010 to 29 February 2016) by multi-point sampling at 1.8, 3.7 and 8.3 m, using a Picarro analyzer at a rural site in the Low Atmosphere Research Centre (CIBA), on the upper Spanish plateau. Data were divided into diurnal and nocturnal records. The mathematical equation proposed to analyze the overall data was a harmonic one, comprising a polynomial (trend) and a series of harmonics (seasonal cycle). Amplitude was considered as a constant and variable term over time. Quite different behaviour was found between day and night measurements in both climate forcing agents. CO₂ showed an accelerating trend in autumn, whereas CH₄ trends were higher during the winter. Increasing growth rates were reported for CO₂ and CH₄ over the whole study period. Nocturnal CO₂ amplitudes are higher than diurnal ones except in winter for both gases, and also in the autumn for CH₄.

Keywords : Upper Spanish plateau, Greenhouse gases, Series of harmonics, Daytime, Night-time

Influence of seasonality, air mass origin and particulate matter chemical composition on airborne bacterial community structure in the Po Valley, Italy

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Source: Volumes 593–594, 1 September 2017, Pages 677–687
<https://doi.org/10.1016/j.scitotenv.2017.03.199>

The integration of chemical and biological data in aerosol studies represents a new challenge in atmospheric science. In this perspective it will be possible to gain a clearer and deeper comprehension of biogeochemical cycles in the atmosphere.

In this view, this study aimed to investigate the relationships occurring between bacterial populations and PM chemical composition in one of the most polluted and urbanized areas in Europe: the Po Valley (Italy). Moreover, seasonality, long- and short-range transports were also evaluated to investigate the influence on airborne bacterial communities.

PM samples were collected in two cities of the Po Valley (Milan and Venice) characterized by different meteorological conditions and atmospheric pollutant sources. Samples were analysed for water-soluble inorganic ions (WSIIs) and bacterial community structure. Chemical and biological

data were jointly processed by using redundancy discriminate analysis (RDA), while the influence of atmospheric circulation was evaluated by using wind ground data and back-trajectories analysis. Results showed strong seasonal shifts of bacterial community structure in both cities, while a different behaviour was observed for air mass circulation at Milan and Venice sites: long-range transport significantly affected bacterial populations in Milan whereas local ground wind had more influence in the Venice area. Moreover, difference in taxonomic composition can be mostly addressed to the characteristics of sampling sites. This evidence could suggest that, while PM composition is influenced by long-range transport, bacterial populations are affected, besides transport, by other factors (i.e., season and sampling site location). This perspective allows to better understand and explain airborne bacterial community behaviour.

Keywords : Airborne bacteria, WSIs, Ground wind circulation, Back-trajectories analysis

Long-term exposure to high air pollution induces cumulative DNA damages in traffic policemen

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*Source: Volumes 593–594, 1 September 2017, Pages 330–336
<https://doi.org/10.1016/j.scitotenv.2017.03.179>*

The specific effects of long-term exposure to high air pollution on human health and biological remain unclear. To explore the adverse health effects as well as biological mechanisms and biomarkers for durative exposure to air pollution, 183 traffic policemen and 88 office policemen were enrolled in this study. The concentration of PM_{2.5} in both the traffic and office policemen's working environments were obtained. Detailed personal questionnaires were completed and levels of inflammation, oxidative stress and DNA damage markers of all participants were analyzed in this study. The average PM_{2.5} concentration of the intersections of main roads and the offices of control group were $132.4 \pm 48.9 \mu\text{g}/\text{m}^3$ and $50.80 \pm 38.6 \mu\text{g}/\text{m}^3$, respectively. The traffic policemen, who stably exposed to at least 2 times higher PM_{2.5} in their work area as compared with the control group, have a median average duration of 7.00 years, and average cumulative intersection duty time reached 8030 h. No statistically significant differences in the levels of inflammation markers were observed between the traffic and office policemen. However, the DNA damage markers in traffic policemen shared significant positive correlation with cumulative intersection duty time and higher than those in the office policemen. Multiple linear regression analysis demonstrated that the increase of cumulative intersection duty time by 1 h per day for one year was associated with the increase in 8-hydroxy-2'-deoxyguanosine of 0.329% (95% CI: 0.249% to 0.409%), tail DNA of 0.051% (95% CI: 0.041% to 0.061%), micronucleus frequency of 0.036‰ (95% CI: 0.03‰ to 0.043‰), and a decrease in glutathione of 0.482% (95% CI: - 0.652% to - 0.313%). These findings suggest that long-term exposure to high air pollution could induce cumulative DNA damages, supporting the hypothesis that durative exposure to air pollution is associated with an increased risk of cancer.

Keywords : Air pollution, DNA damage, Long-term exposure, PM_{2.5}, Inflammation, Oxidative stress

Ambient PM_{2.5} in the residential area near industrial complexes: Spatiotemporal variation, source apportionment, and health impact

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Source: Volumes 590–591, 15 July 2017, Pages 204–214
<https://doi.org/10.1016/j.scitotenv.2017.02.212>

This study systemically investigated the ambient PM_{2.5} (n = 108) with comprehensive analyses of the chemical composition, identification of the potential contributors, and estimation of the resultant respiratory physician visits in the residential regions near energy-consuming and high-polluting industries in central Taiwan. The positive matrix fraction (PMF) model with chemical profiles of trace metals, water-soluble ions, and organic/elemental carbons (OC/EC) was applied to quantify the potential sources of PM_{2.5}. The influences of local sources were also explored using the conditional probability function (CPF). Associations between the daily PM_{2.5} concentration and the risk of respiratory physician visits for the elderly (≥ 65 years of age) were estimated using time-series analysis. A seasonal variation, with higher concentrations of PM_{2.5}, metals (As, Cd, Sb, and Pb), OC/EC and ions (i.e., NO₃⁻, SO₄²⁻ and NH₄⁺) in the winter than in the spring and summer, was observed. Overall, an increase of 10 $\mu\text{g m}^{-3}$ in the same-day PM_{2.5} was associated with an $\sim 2\%$ (95% CI: 1.5%–2.5%) increase in respiratory physician visits. Considering the health benefits of an effective reduction, we suggest that the emission from coal combustion (23.5%), iron ore and steel industry (17.1%), and non-ferrous metallurgy (14.4%), accounting for $\sim 70\%$ of the primary PM_{2.5} in the winter are prioritized to control.

Keywords : Fine particle, Source apportionment, Chemical constituents, Respiratory physician visits

Indoor air pollutants, ventilation rate determinants and potential control strategies in Chinese dwellings: A literature review

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Source: Volume 586, 15 May 2017, Pages 696–729
<https://doi.org/10.1016/j.scitotenv.2017.02.047>

After nearly twenty years of rapid modernization and urbanization in China, huge achievements have transformed the daily lives of the Chinese people. However, unprecedented environmental consequences in both indoor and outdoor environments have accompanied this progress and have triggered public awareness and demands for improved living standards, especially in residential environments. Indoor pollution data measured for > 7000 dwellings (approximately 1/3 were newly decorated and were tested for volatile organic compound (VOC) measurements, while the rest were tested for particles, phthalates and other semi-volatile organic compounds (SVOCs), moisture/mold, inorganic gases and radon) in China within the last ten years were reviewed, summarized and compared with indoor concentration recommendations based on sensory or health end-points. Ubiquitous pollutants that exceed the concentration recommendations, including particulate matter, formaldehyde, benzene and other VOCs, moisture/mold, inorganic gases and

radon, were found, indicating a common indoor air quality (IAQ) issue in Chinese dwellings. With very little prevention, oral, inhalation and dermal exposure to those pollutants at unhealthy concentration levels is almost inevitable. CO₂, VOCs, humidity and radon can serve as ventilation determinants, each with different ventilation demands and strategies, at typical occupant densities in China; and particle reduction should be a prerequisite for determining ventilation requirements. Two directional ventilation modes would have profound impacts on improving IAQ for Chinese residences are: 1) natural (or window) ventilation with an air cleaner and 2) mechanical ventilation with an air filtration unit, these two modes were reviewed and compared for their applicability and advantages and disadvantages for reducing human exposure to indoor air pollutants. In general, mode 2 can more reliably ensure good IAQ for occupants; while mode 1 is more applicable due to its low cost and low energy consumption. However, besides a roadmap, substantial efforts are still needed to develop affordable, applicable and general ventilation solutions to improve the IAQ of residential buildings in China.

Keywords : Bioeffluent, Particular matter, VOC, SVOC, Mold, Radon

Extreme air pollution with contaminants originating from the mining-metallurgical processes

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Source: Volume 586, 15 May 2017, Pages 1066-1075
<https://doi.org/10.1016/j.scitotenv.2017.02.091>

Levels of SO₂ and metals/metalloids in the air near a copper smelter in Bor (Serbia) from 2009 to 2015 were presented in this study. Annual levels of SO₂ were constantly above the proposed limit value (LV), at almost all the measuring sites. SO₂ concentrations on an annual level measured in different zones in Bor were several times higher compared to the LV in 2011, 2012, 2014 and 2015. Enormously high daily SO₂ concentrations measured at the suburban zone (3734 µg m⁻³) was 187 times higher than the LV given by the World Health Organization. Annual arsenic concentrations exceeded the LV at all the measuring sites during the study period. Extremely high annual As level in 2012 was 21 times higher than the LV proposed by the European Union. The annual lead and cadmium concentrations frequently exceeded the LV. The vicinity of the measuring sites to the copper smelter and the location of the sites in regard to the prevailing wind directions contribute to higher content of air pollutants. The data presented in this study revealed that extremely high concentrations of air polluting substances could rank the town of Bor as one of the most polluted regions in Serbia and beyond.

Keywords : Air pollution, SO₂, Arsenic, Lead, Cadmium

Linking potential heat source and sink to urban heat island: Heterogeneous effects of landscape pattern on land surface temperature

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Source: Volume 586, 15 May 2017, Pages 457-465
<https://doi.org/10.1016/j.scitotenv.2017.01.191>

Rapid urbanization has significantly contributed to the development of urban heat island (UHI). Regulating landscape composition and configuration would help mitigate the UHI in megacities. Taking Shenzhen, China, as a case study area, we defined heat source and heat sink and identified strong and weak sources as well as strong and weak sinks according to the natural and socioeconomic factors influencing land surface temperature (LST). Thus, the potential thermal contributions of heat source and heat sink patches were differentiated. Then, the heterogeneous effects of landscape pattern on LST were examined by using semiparametric geographically weighted regression (SGWR) models. The results showed that landscape composition has more significant effects on thermal environment than configuration. For a strong source, the percentage of patches has a positive impact on LST. Additionally, when mosaicked with some heat sink, even a small improvement in the degree of dispersion of a strong source helps to alleviate UHI. For a weak source, the percentage and density of patches have positive impacts on LST. For a strong sink, the percentage, density, and degree of aggregation of patches have negative impacts on LST. The effects of edge density and patch shape complexity vary spatially with the fragmentation of a strong sink. Similarly, the impacts of a weak sink are mainly exerted via the characteristics of percent, density, and shape complexity of patches.

Keywords : Urban heat island, Land surface temperature, Source·Sink·Heterogeneity, SGWR model

Vehicle emission trends in China's Guangdong Province from 1994 to 2014

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Source: Volume 586, 15 May 2017, Pages 512-521
<https://doi.org/10.1016/j.scitotenv.2017.01.215>

Exploring vehicle emission trends within and outside the Pearl River Delta (PRD) region during a long period was scientific and practical, for the economic rapid unbalanced development, continuous implements of severe reducing vehicle emissions measures in Guangdong province. Multi-year inventories of vehicle emissions from 1994 to 2014 were estimated based on the emissions factors of different emissions standards and vehicle kilometers travelled for all types of vehicles. The trends and characteristics of the emissions of carbon monoxide (CO), volatile organic compounds (VOCs), nitrogen oxides (NO_x), fine particulate matter (PM_{2.5}) and course particulate matter (PM₁₀) were then analyzed within and outside the PRD region. In the above two regions, the total amount of the five pollutant emissions varied greatly with gross domestic product (GDP) from 1994 to 2014, showing the overall performance of the first increasing up to 1.6–3.0 times

before 2002, and then decreasing. However, the five pollutant emissions in the PRD region were 2.4–3.3 times more than in the non-PRD region. In both regions, light passenger cars and motorcycles were the main contributors to CO and VOC emissions (65%–80%), and heavy duty trucks and passenger cars were the main contributors to NO_x, PM_{2.5} and PM₁₀ emissions (around 42%–50%). Moreover, compared to CO and VOCs emissions, the changes in the contribution of every vehicles type to NO_x, PM_{2.5} and PM₁₀ emissions were more obvious, and coincided with the implementation time of emission and fuel standards in the non-PRD region. It was noted that CO and VOC emission variations was correlated closely with the population of yellow-label light passenger cars and motorcycles, whereas those of NO_x and PM_{2.5} was coincided that of yellow-label heavy passenger cars and trucks.

Keywords : Vehicle emissions, Multi-year inventories trends, PRD and non-PRD region, GDP

Heavy pollution episodes, transport pathways and potential sources of PM_{2.5} during the winter of 2013 in Chengdu (China)

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Source: Volumes 584–585, 15 April 2017, Pages 1056-1065
<https://doi.org/10.1016/j.scitotenv.2017.01.160>

Air mass concentration data from 8 environmental quality monitoring sites and meteorological data of Chengdu from 1 December 2013 to 28 February 2014 were used in this study. Chengdu suffered five continuous heavy pollutions during this winter due to the basin terrain and the meteorological conditions of low wind speed, low precipitation and high relative humidity. Analysing the hourly resolution time series of pollutants' concentrations, variation of PM_{2.5} in the urban area followed a growing “saw-tooth cycle” pattern during the heavy pollution, with a daily cycle of bimodal distribution. The massive letting-off of fireworks within a short period of time on the Eve of the Lunar New Year under the unfavourable diffusion conditions resulted in an extreme pollution event. The sharply rising Longmen-Qionglai Mountains to the west of the Sichuan Basin not only acted as a huge barrier to block the air mass from the east but also favoured the formation of a local circulation. The cluster analysis of back trajectories revealed that up to 77% of them came from the inner part of the Basin. Combining the concentration data of PM_{2.5} with air mass back trajectories, a potential source contribution function (PSCF) model and a concentration-weighted trajectory (CWT) method were used to evaluate the transport pathways and sources over PM_{2.5} of Chengdu, revealing that the main potential sources of PM_{2.5} were located in southeast cities and the western margin of the Sichuan Basin. The result provided advice for the government to take measures in improving air quality.

Keywords :Air mass trajectories, Heavy pollution, Chengdu

Particulate pollution in urban Chongqing of southwest China: Historical trends of variation, chemical characteristics and source apportionment

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Source: Volumes 584–585, 15 April 2017, Pages 523-534
<https://doi.org/10.1016/j.scitotenv.2017.01.060>

Chongqing, the largest megacity in southwest China, faces serious aerosol pollution but lacks information on particle characteristics and its sources. Official data released by Chongqing Environmental Protection Bureau demonstrated that urban PM₁₀ concentrations decreased remarkably from 150 $\mu\text{g m}^{-3}$ in 2000 to 90 $\mu\text{g m}^{-3}$ in 2012. However, only several peer-reviewed studies paid attention to local fine particle (PM_{2.5}) pollution. In the study, PM_{2.5} samples were obtained and subjected to chemical analysis in an urban site of the city during 2012 to 2013. The annual mean PM₁₀ and PM_{2.5} concentrations in urban Chongqing were 103.9 ± 52.5 and 75.4 ± 42.2 $\mu\text{g m}^{-3}$, respectively. PM_{2.5} showed a distinct seasonality of high concentration in winter and similar levels in other seasons. The average OC/EC (organic carbon/element carbon) ratio was 3.7 with more high-OC/EC ratio sources contribution in autumn and winter. The varying sources and formation mechanisms resulted in SO₄²⁻ and NH₄⁺ peaks in both summer and winter, whereas high nitrate concentration was only observed in winter. In the average mass closure, PM_{2.5} was composed of 23.0% SO₄²⁻, 11.7% NO₃⁻, 10.9% NH₄⁺, 30.8% OM (organic matter), 5.2% EC, 8.2% mineral dust, 0.6% TEQ (trace elements), 1.0% Cl⁻ and 1.1% K⁺, while exhibiting large seasonal variability. Using positive matrix factorization (PMF), six sources were apportioned in PM_{2.5}: secondary inorganic aerosols, coal combustion, other industrial pollution, soil dust, vehicular emission, and metallurgical industry. The annual mean contribution of above sources to PM_{2.5} was 37.5, 22.0, 17.5, 11.0, 9.8 and 2.2%, respectively. Coal combustion was identified by As tracer and dominated the primary sources of PM_{2.5}, while the two different industrial sources were characterized by Cr and Mo, Co, Ni, and Se, respectively. The study is of great importance in characterizing the historical trends, current chemical characteristics and sources of fine particles in urban Chongqing.

Keywords : Chemical composition, Source apportionment, Chongqing

Emission factors for PM_{2.5}, CO, CO₂, NO_x, SO₂ and particle size distributions from the combustion of wood species using a new controlled combustion chamber 3CE

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Source: Volumes 584–585, 15 April 2017, Pages 901-910
<https://doi.org/10.1016/j.scitotenv.2017.01.136>

The objective of this research was to determine emission factors (EF) for particulate matter (PM_{2.5}), combustion gases and particle size distribution generated by the combustion of *Eucalyptus globulus* (EG), *Nothofagus obliqua* (NO), both hardwoods, and *Pinus radiata* (PR), softwood, using a

controlled combustion chamber (3CE). Additionally, the contribution of the different emissions stages associated with the combustion of these wood samples was also determined. Combustion experiments were performed using shaving size dried wood (0% humidity). The emission samples were collected with a tedlar bag and sampling cartridges containing quartz fiber filters. High reproducibility was achieved between experiment repetitions ($CV < 10\%$, $n = 3$). The EF for PM_{2.5} was 1.06 g kg⁻¹ for EG, 1.33 g kg⁻¹ for NO, and 0.84 g kg⁻¹ for PR. Using a laser aerosol spectrometer (0.25–34 µm), the contribution of particle emissions (PM_{2.5}) in each stage of emission process (SEP) was sampled in real time. Particle size of 0.265 µm were predominant during all stages, and the percentages emitted were PR (33%), EG (29%), and NO (21%). The distributions of EF for PM_{2.5} in pre-ignition, flame and smoldering stage varied from predominance of the flame stage for PR (77%) to predominance of the smoldering stage for NO (60%). These results prove that flame phase is not the only stage contributing to emissions and on the contrary, pre-ignition and in especial post-combustion smoldering have also very significant contributions. This demonstrates that particle concentrations measured only in stationary state during flame stage may cause underestimation of emissions.

Keywords : Particulate matter, Emissions factors, Residential wood combustion, Combustion chamber

Particulate matter pollution over China and the effects of control policies

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Source: Volumes 584–585, 15 April 2017, Pages 426–447
<https://doi.org/10.1016/j.scitotenv.2017.01.027>

China is one of the regions with highest PM_{2.5} concentration in the world. In this study, we review the spatio-temporal distribution of PM_{2.5} mass concentration and components in China and the effect of control measures on PM_{2.5} concentrations. Annual averaged PM_{2.5} concentrations in Central-Eastern China reached over 100 µg m⁻³, in some regions even over 150 µg m⁻³. In 2013, only 4.1% of the cities attained the annual average standard of 35 µg m⁻³. Aitken mode particles tend to dominate the total particle number concentration. Depending on the location and time of the year, new particle formation (NPF) has been observed to take place between about 10 and 60% of the days. In most locations, NPF was less frequent at high PM mass loadings. The secondary inorganic particles (i.e., sulfate, nitrate and ammonium) ranked the highest fraction among the PM_{2.5} species, followed by organic matters (OM), crustal species and element carbon (EC), which accounted for 6–50%, 15–51%, 5–41% and 2–12% of PM_{2.5}, respectively. In response to serious particulate matter pollution, China has taken aggressive steps to improve air quality in the last decade. As a result, the national emissions of primary PM_{2.5}, sulfur dioxide (SO₂), and nitrogen oxides (NO_x) have been decreasing since 2005, 2006, and 2011, respectively. The emission control policies implemented in the last decade could result in noticeable reduction in PM_{2.5} concentrations, contributing to the decreasing PM_{2.5} trends observed in Beijing, Shanghai, and Guangzhou. However, the control policies issued before 2010 are insufficient to improve PM_{2.5} air quality notably in future. An optimal mix of energy-saving and end-of-pipe control measures should be implemented, more ambitious control policies for NMVOC and NH₃ should be

enforced, and special control measures in winter should be applied. 40–70% emissions should be cut off to attain PM_{2.5} standard.

Keywords : Metropolitan regions, Chemical speciation, Control policies, China

Regional transport of anthropogenic pollution and dust aerosols in spring to Tianjin — A coastal megacity in China

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Source: Volumes 584–585, 15 April 2017, Pages 381–392.
<https://doi.org/10.1016/j.scitotenv.2017.01.016>

Simultaneous measurements of columnar aerosol microphysical and optical properties, as well as PM_{2.5} chemical compositions, were made during two types of spring pollution episodes in Tianjin, a coastal megacity of China. The events were investigated using field observations, satellite data, model simulations, and meteorological fields. The lower Ångström Exponent and the higher aerosol optical depth on 29 March, compared with the earlier event on 26 March, implied a dominance of coarse mode particles — this was consistent with the differences in volume-size distributions. Based on the single scattering spectra, the dominant absorber (at blue wavelength) changed from black carbon during less polluted days to brown carbon on 26 March and dust on 29 March. The concentrations of major PM_{2.5} species for these two episodes also differed, with the earlier event enriched in pollution-derived substances and the later with mineral dust elements. The formation mechanisms of these two pollution episodes were also examined. The 26 March episode was attributed to the accumulation of both local emissions and anthropogenic pollutants transported from the southwest of Tianjin under the control of high pressure system. While the high aerosol loading on 29 March was caused by the mixing of transported dust from northwest source region with local urban pollution. The mixing of transported anthropogenic pollutants and dust with local emissions demonstrated the complexity of springtime pollution in Tianjin. The synergy of multi-scale observations showed excellent potential for air pollution study.

Keywords : Multi-scale observations, Urban pollution, Dust transport, Formation mechanism

Utilising green and bluespace to mitigate urban heat island intensity

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Source: Volumes 584–585, 15 April 2017, Pages 1040–1055
<https://doi.org/10.1016/j.scitotenv.2017.01.158>

It has long been recognised that cities exhibit their own microclimate and are typically warmer than the surrounding rural areas. This ‘mesoscale’ influence is known as the urban heat island (UHI) effect and results largely from modification of surface properties leading to greater absorption of

solar radiation, reduced convective cooling and lower water evaporation rates. Cities typically contain less vegetation and bodies of water than rural areas, and existing green and bluespace is often under threat from increasing population densities. This paper presents a meta-analysis of the key ways in which green and bluespace affect both urban canopy- and boundary-layer temperatures, examined from the perspectives of city-planning, urban climatology and climate science. The analysis suggests that the evapotranspiration-based cooling influence of both green and bluespace is primarily relevant for urban canopy-layer conditions, and that tree-dominated greenspace offers the greatest heat stress relief when it is most needed. However, the magnitude and transport of cooling experienced depends on size, spread, and geometry of greenspaces, with some solitary large parks found to offer minimal boundary-layer cooling. Contribution to cooling at the scale of the urban boundary-layer climate is attributed mainly to greenspace increasing surface roughness and thereby improving convection efficiency rather than evaporation. Although bluespace cooling and transport during the day can be substantial, nocturnal warming is highlighted as likely when conditions are most oppressive. However, when both features are employed together they can offer many synergistic ecosystem benefits including cooling. The ways in which green and bluespace infrastructure is applied in future urban growth strategies, particularly in countries expected to experience rapid urbanisation, warrants greater consideration in urban planning policy to mitigate the adverse effects of the UHI and enhance climate resilience.

Keywords: Urban heat island, Greenspace, Bluespace, Evapotranspiration, Green infrastructure, Blue infrastructure

An emission processing system for air quality modelling in the Mexico City metropolitan area: Evaluation and comparison of the MOBILE6.2-Mexico and MOVES-Mexico traffic emissions

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Source: Volumes 584–585, 15 April 2017, Pages 882-900
<https://doi.org/10.1016/j.scitotenv.2017.01.135>

This article describes the High-Selective Resolution Modelling Emission System for Mexico (HERMES-Mex) model, an emission processing tool developed to transform the official Mexico City Metropolitan Area (MCMA) emission inventory into hourly, gridded (up to 1 km²) and speciated emissions used to drive mesoscale air quality simulations with the Community Multi-scale Air Quality (CMAQ) model. The methods and ancillary information used for the spatial and temporal disaggregation and speciation of the emissions are presented and discussed. The resulting emission system is evaluated, and a case study on CO, NO₂, O₃, VOC and PM_{2.5} concentrations is conducted to demonstrate its applicability. Moreover, resulting traffic emissions from the Mobile Source Emission Factor Model for Mexico (MOBILE6.2-Mexico) and the Motor Vehicle Emission Simulator for Mexico (MOVES-Mexico) models are integrated in the tool to assess and compare their performance. NO_x and VOC total emissions modelled are reduced by 37% and 26% in the MCMA when replacing MOBILE6.2-Mexico for MOVES-Mexico traffic emissions. In terms of air quality, the system composed by the Weather Research and Forecasting model (WRF) coupled with the HERMES-Mex and CMAQ models properly reproduces the pollutant levels and patterns measured in

the MCMA. The system's performance clearly improves in urban stations with a strong influence of traffic sources when applying MOVES-Mexico emissions. Despite reducing estimations of modelled precursor emissions, O₃ peak averages are increased in the MCMA core urban area (up to 30 ppb) when using MOVES-Mexico mobile emissions due to its VOC-limited regime, while concentrations in the surrounding suburban/rural areas decrease or increase depending on the meteorological conditions of the day. The results obtained suggest that the HERMES-Mex model can be used to provide model-ready emissions for air quality modelling in the MCMA.

Keywords : Emission processing system, Road transport emissions, Air quality modeling, MOVES-Mexico, MOBILE6.2-Mexico, Ozone

An anomalous African dust event and its impact on aerosol radiative forcing on the Southwest Atlantic coast of Europe in February 2016

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Source: Volume 583, 1 April 2017, Pages 269-279

<https://doi.org/10.1016/j.scitotenv.2017.01.064>

A desert dust (DD) event that had its origin in North Africa occurred on the 20th–23rd of February 2016. The dust transport phenomenon was exceptional because of its unusual intensity during the coldest season. A historical dataset (2006–2015) of February meteorological scenarios using ECMWF fields, meteorological parameters, aerosol optical properties, surface O₃ and AOD retrieved from MODIS at the El Arenosillo observatory (southwestern Spain) were analysed and compared with the levels during the DD event to highlight its exceptionality. Associated with a low-pressure system in western North Africa, flows transported air from the Sahel to Algeria and consequently increased temperatures from the surface to 700 hPa by up to 7–9 °C relative to the last decade. These conditions favoured the formation of a Saharan air layer. Dust was transported to the north and reached the Western Mediterranean Basin and the Iberian Peninsula. The arrival of the DD event at El Arenosillo did not affect the surface weather conditions or ozone but did impact the aerosol radiative forcing at the top of atmosphere (RFTOA). Aerosol radiative properties did not change relative to historical; however, the particle size and the amount of the aerosol were significantly higher. The DD event caused an increase (in absolute terms) of the mean aerosol RFTOA to a value of -8.1 W m^{-2} (long-term climatological value $\sim -1.5 \text{ W m}^{-2}$). The aerosol RFTOA was not very large relative other DD episodes; however, our analysis of the historical data concluded that the importance of this DD event lay in the month of occurrence. European phenological datasets related to extreme atmospheric events predominantly reflect changes that are probably associated with climate change. This work is an example of this phenomenon, showing an event that occurred in a hotspot, the Saharan desert, and its impact two thousand km away.

Keywords : Saharan desert dust, Aerosol optical properties, Extreme meteorological event, Climate change

Long-range and regional transported size-resolved atmospheric aerosols during summertime in urban Shanghai

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Source: Volume 583, 1 April 2017, Pages 334-343
<https://doi.org/10.1016/j.scitotenv.2017.01.073>

In this study, the concentrations of water soluble ions (WSI), organic carbon (OC), and elemental carbon (EC) of size-resolved (0.056–18 μm) atmospheric aerosols were measured in July and August 2015 in Shanghai, China. Backward trajectory model and potential source contribution function (PSCF) model were used to identify the potential source distributions of size-resolved particles and PM_{1.8}-associated atmospheric inorganic and carbonaceous aerosols. The results showed that the average mass concentrations of PM_{0.1}, PM₁, and PM_{1.8} were 21.21, 82.90, and 100.1 $\mu\text{g m}^{-3}$ in July and 7.00, 29.21, and 35.10 $\mu\text{g m}^{-3}$ in August, respectively, indicating that the particulate matter pollution was more serious in July than in August in this study due to the strong dependence of the aerosol species on the air mass origins. The trajectory cluster analysis revealed that the air masses originated from heavily industrialized areas including the Pearl River Delta (PRD) region, the Yangtze River Delta (YRD) region and the Beijing-Tianjin region were characterised with high OC and SO₄²⁻ loadings. The results of PSCF showed that the pollution in July was mainly influenced by long-range transport while it was mainly associated to local and intra-regional transport in August. Besides the contributions of anthropogenic sources from YRD and PRD region, ship emissions from the East China Sea also made a great contribution to the high loadings of PM_{1.8} and PM_{1.8}-associated NO₃⁻, NH₄⁺, and EC in July. SO₄²⁻ in Shanghai was dominantly ascribed to anthropogenic sources and the high PSCF values for PM_{1.8}-associated SO₄²⁻ observed in August was mainly due to the ship emissions of Shanghai port, such as Wusong port and Yangshan deep-water port. These results indicated that the particulate pollutants from long-range transported air masses and shipping made a significant contribution to Shanghai's air pollution.

Keywords : Size-resolved aerosol, Regional transport, Long-range transport, PSCF

Estimating the contribution of regional transport to PM_{2.5} air pollution in a rural area on the North China Plain

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Source: Volume 583, 1 April 2017, Pages 280-291
<https://doi.org/10.1016/j.scitotenv.2017.01.066>

PM_{2.5} air pollution in metropolises as well as some medium-sized cities in the North China Plain have aroused many researchers' interest, but less attention has been paid to the rural areas of this region. In this study, four months of daily PM_{2.5} samples were collected from a rural site in Lingcheng (a district of Dezhou City in Shandong Province) during different seasons in 2013 and

2014. Analysis of the samples indicates that the PM_{2.5} air pollution was severe over this area with the four-month average concentration of 105.9 µg/m³, three times higher than China's guideline for this pollutant (35 µg/m³). In winter, the monthly average concentration was as high as 151.2 µg/m³. In order to identify the potential source regions, the Integrated Source Apportionment Method within Community Multiscale Air Quality model (CMAQ-ISAM) was applied during the wintertime. The regional source apportionment results show that local emissions in Lingcheng only contributed 15.4% to PM_{2.5} concentrations, with 12.6% and 28.1% from its circumjacent areas in Dezhou City and the six surrounding cities, respectively. Regional transport from areas farther away and the boundaries account for 31.6% and 11.1%, respectively. This indicates that the ambient PM_{2.5} at Lingcheng is not affected only by emissions from local and circumjacent areas; regional and long-range transport should also be considered. Further analysis indicated that with increasing degrees of pollution, the contributions from local and circumjacent regions showed a clear downward trend, while the contributions from northern and southwestern areas, which most of the trajectories passed through during periods of heavy haze, showed an obvious upward trend.

Keywords : Source apportionment, CMAQ-ISAM, Regional transport

The analysis and application of a new hybrid pollutants forecasting model using modified Kolmogorov–Zurbenko filter

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Source: Volume 583, 1 April 2017, Pages 228-240
<https://doi.org/10.1016/j.scitotenv.2017.01.057>

Cities in China suffer from severe smog and haze, and a forecasting system with high accuracy is of great importance to foresee the concentrations of the airborne particles. Compared with chemical transport models, the growing artificial intelligence models can simulate nonlinearities and interactive relationships and getting more accurate results. In this paper, the Kolmogorov–Zurbenko (KZ) filter is modified and firstly applied to construct the model using an artificial intelligence method. The concentration of inhalable particles and fine particulate matter in Dalian are used to analyze the filtered components and test the forecasting accuracy. Besides, an extended experiment is made by implementing a comprehensive comparison and a stability test using data in three other cities in China. Results testify the excellent performance of the developed hybrid models, which can be utilized to better understand the temporal features of pollutants and to perform a better air pollution control and management.

Keywords : Kolmogorov–Zurbenko filter, Inhalable particles, Fine particulate matter, Decomposition-ensemble technique

Trends in population exposure to particulate matter in urban areas of Greece during the last decade

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Source: Volumes 581–582, 1 March 2017, Pages 399–412
<https://doi.org/10.1016/j.scitotenv.2016.12.148>

Assessment of population exposure is very important for the evaluation of the adverse health effects of particulate matter (PM) ambient levels. In this work population exposure to PM₁₀ and PM_{2.5} has been evaluated in outdoor environments in the Athens (AMA) and Thessaloniki (TMA) metropolitan areas during the period 2001–2010. The methodology used is based on combining spatiotemporally allocated PM₁₀ and PM_{2.5} concentration fields with the geographical distribution of population. The results showed that the number of people living in the AMA exposed to PM₁₀ and PM_{2.5} concentrations above the annual air quality standards (AQS), has dropped 18% since 2001 and 98% since 2008, respectively. Likewise, in 2010 8% less, compared to 2001, of the AMA population lived in areas where the daily AQS for PM₁₀ was exceeded > 35 times a year. The results as regards TMA indicated a decrease in the number of people exposed to PM₁₀ concentrations over the annual AQS value (78% decrease). However, the number of people living in areas with PM₁₀ concentrations over the daily AQS for > 35 times in a year doubled since 2001. Finally, the spatial distribution of the normalised population load which reflects populated areas with concentrations above the daily AQS was evaluated. The hot spots for both AMA and TMA areas correspond to urban areas and areas with significant primary PM₁₀ emissions.

Keywords : Population exposure, Particulate matter, Aerosol emissions, Urban air quality

Shipping emissions and their impacts on air quality in China

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Source: Volumes 581–582, 1 March 2017, Pages 186–198
<https://doi.org/10.1016/j.scitotenv.2016.12.098>

China has > 400 ports, is home to 7 of 10 biggest ports in the world and its waterway infrastructure construction has been accelerating over the past years. But the increasing number of ports and ships means increasing emissions, and in turn, increasing impact on local and regional air pollution. This paper presents an overview of the broad field of ship emissions in China and their atmospheric impacts, including topics of ship engine emissions and control, ship emission factors and their measurements, developing of ship emission inventories, shipping and port emissions of the main shipping areas in China, and quantitative contribution of shipping emissions to the local and regional air pollution.

There have been an increasing number of studies published on all the above aspects, yet, this review identified some critical research gaps, filling of which is necessary for better control of ship emissions, and for lowering their impacts. In particular, there are very few studies on inland ports and river ships, and there are few national scale ship emission inventories available for China.

While advanced method to estimate ship emission based on ship AIS activities makes it now possible to develop high spatial- and temporal-resolution emission inventories, the ship emission factors used in Chinese studies have been based mainly on foreign measurements. Further, the contribution of ship emissions to air pollution in coastal cities, the dispersion of pollution plumes emitted by ships, or the chemical evolution process along the transmission path, have so far not been systematically studied in China.

Keywords : Ship emission, China, Air pollution, Emission factor

Characteristics of PAHs in street dust of Beijing and the annual wash-off load using an improved load calculation method

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Source: Volumes 581–582, 1 March 2017, Pages 328-336
<https://doi.org/10.1016/j.scitotenv.2016.12.133>

A significant amount of polycyclic aromatic hydrocarbons (PAHs) adsorbed in street dust ends up in runoff. Accordingly, evaluating the content, possible wash-off load and constituent risks associated with street dust is critical for urban runoff usage. Based on the PAH concentrations examined in this study, different risk assessment methods were applied and the differences among results were analyzed. An improved PAH wash-off calculation method was established and the annual PAH wash-off load was obtained. In addition, emission sources were diagnosed based on isomer ratios and PMF methods. The overall mean Σ PAHs (sum of 16 individual PAHs) concentration in the street dust was 3.70 $\mu\text{g/g}$, with the highest mean concentrations found for main roads (5.18 $\mu\text{g/g}$). Adjacent anthropogenic activities had a greater effect on pollution characteristics of street dust than land use types. The toxic risk order was park (0.64 $\mu\text{g/g}$) > main road (0.57 $\mu\text{g/g}$) > residence (0.32 $\mu\text{g/g}$) > street (0.29 $\mu\text{g/g}$) > commerce (0.23 $\mu\text{g/g}$), while that of the ecological risk index was main road (1278) > street (920) > residence (904) > commerce (713) > park (195). Although the park sites had a high level of toxic risk, they showed low ecological risk because they had less dust mass per unit area, indicating a great difference in the risk evaluation results and the difference methods. Using different values for different magnitudes of the antecedent dry days, rainfall event amounts, and dust mass fraction of different size ranges, the average wash-off load of Σ PAHs from street dust was calculated to be 23 kg per year in Beijing from 2000 to 2014 with an improved wash-off load calculation method. The main sources of PAHs in the street dust of Beijing's center were identified as gasoline emissions, diesel emissions, coal combustion and unburned petroleum.

Keywords : PAHs, Risk assessment, Wash-off load, Source signature

Long-term trend of chemical composition of atmospheric precipitation at a regional background station in Northern China

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Source: Volume 580, 15 February 2017, Pages 1340-1350
<https://doi.org/10.1016/j.scitotenv.2016.12.097>

Understanding the trend of chemical composition of precipitation is of great importance for air pollution control strategies in Northern China. A comprehensive study on the long-term chemical compositions of precipitation was carried out from 2003 to 2014 at the Shangdianzi (SDZ) regional background station in northern China. All samples were analyzed for pH, electrical conductivity and major ions (F^- , Cl^- , NO_3^- , SO_4^{2-} , NH_4^+ , Mg^{2+} , Ca^{2+} , K^+ and Na^+). The average pH during this period was 4.53 ± 0.35 , which is considerably lower than those reported in other background stations in China (Linan, Waliguan and Longfengshan). NH_4^+ , SO_4^{2-} , Ca^{2+} and NO_3^- were the dominant ions in precipitation, with concentrations (volume-weighted mean) of $212.99 \mu eq L^{-1}$, $200.20 \mu eq L^{-1}$, $116.88 \mu eq L^{-1}$ and $98.56 \mu eq L^{-1}$, respectively. The ion concentrations at SDZ were much higher than those of other background stations and megacities in China. A significantly increasing trend was observed for NO_3^- ($7.26\% year^{-1}$), and a decreasing trend was observed for SO_4^{2-}/NO_3^- , suggesting that the precipitation of SDZ evolved from a sulfuric acid type to a mixed type dominated by both sulfuric and nitric acid. The source identification indicated that SO_4^{2-} , NO_3^- , NH_4^+ and F^- were dominated by secondary sources, Mg^{2+} , Ca^{2+} and Na^+ mostly originated from natural sources, and K^+ and Cl^- probably associated with anthropogenic sources. Long-range transport of air masses could influence the acidity, electrical conductivity and ion concentrations of precipitation at SDZ. The higher acidity and ion concentrations mainly occurred in the southerly and westerly trajectory pathways and partially in northwest pathways. Anthropogenic pollutants and crustal sources along these pathways were significant contributors to the chemical composition of precipitation in SDZ.

Keywords : Chemical composition, Precipitation, Source, Background station

Statistical analysis of Seoul air quality to assess the efficacy of emission abatement strategies since 1987

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Source: Volume 580, 15 February 2017, Pages 105-116
<https://doi.org/10.1016/j.scitotenv.2016.09.151>

The combined influences of recent mitigation measures on urban air quality have been assessed using hourly observations of the criteria air pollutants (NO , NO_2 , O_3 , CO , and SO_2) made from the Yongsan district of Seoul, Korea, over 26 years (1987 to 2013). A number of data selection criteria are proposed in order to minimize variability associated with temporal changes (at diurnal, weekly,

and seasonal timescales) in source strengths, their spatial distribution, and the atmospheric volume into which they mix. The temporal constraints required to better characterize relationships between observed air quality and changes in source strengths in Seoul were identified as: (i) a 5-hour diurnal sampling window (1300–1700 h), (b) weekday measurements (Monday to Friday only), and (c) summer measurements (when pollutant fetch is mostly Korea-specific, and mean wind speeds are the lowest). Using these selection criteria, we were able to closely relate long-term trends identified in criteria pollutants to a number of published changes to traffic-related source strengths brought about by mitigation measures adopted over the last 10–15 years.

Keywords : Air quality, Temporal influences, Sampling window, Mitigation, Criteria pollutants, Source strengths, Long-term trends

Modelling impacts of atmospheric deposition and temperature on long-term DOC trends

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Source: Volume 578, 1 February 2017, Pages 323–336

<https://doi.org/10.1016/j.scitotenv.2016.10.164>

It is increasingly recognised that widespread and substantial increases in Dissolved organic carbon (DOC) concentrations in remote surface, and soil, waters in recent decades are linked to declining acid deposition. Effects of rising pH and declining ionic strength on DOC solubility have been proposed as potential dominant mechanisms. However, since DOC in these systems is derived mainly from recently-fixed carbon, and since organic matter decomposition rates are considered sensitive to temperature, uncertainty persists over the extent to which other drivers that could influence DOC production. Such potential drivers include fertilisation by nitrogen (N) and global warming. We therefore ran the dynamic soil chemistry model MADOC for a range of UK soils, for which time series data are available, to consider the likely relative importance of decreased deposition of sulphate and chloride, accumulation of reactive N, and higher temperatures, on soil DOC production in different soils. Modelled patterns of DOC change generally agreed favourably with measurements collated over 10–20 years, but differed markedly between sites. While the acidifying effect of sulphur deposition appeared to be the predominant control on the observed soil water DOC trends in all the soils considered other than a blanket peat, the model suggested that over the long term, the effects of nitrogen deposition on N-limited soils may have been sufficient to raise the “acid recovery DOC baseline” significantly. In contrast, reductions in non-marine chloride deposition and effects of long term warming appeared to have been relatively unimportant. The suggestion that future DOC concentrations might exceed preindustrial levels as a consequence of nitrogen pollution has important implications for drinking water catchment management and the setting and pursuit of appropriate restoration targets, but findings still require validation from reliable centennial-scale proxy records, such as those being developed using palaeolimnological techniques.

Keywords : Doc, Sulphur deposition, Nitrogen deposition, Warming, Trend, Dynamic modeling

Benefits on public health from transport-related greenhouse gas mitigation policies in Southeastern European cities

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Source: Volume 579, 1 February 2017, Pages 1427-1438
<https://doi.org/10.1016/j.scitotenv.2016.11.142>

Climate change is a major environmental threat of our time. Cities have a significant impact on greenhouse gas emissions as most of the traffic, industry, commerce and more than 50% of world population is situated in urban areas. Southern Europe is a region that faces financial turmoil, enhanced migratory fluxes and climate change pressure. The case study of Thessaloniki is presented, one of the only two cities in Greece with established climate change action plans. The effects of feasible traffic policies in year 2020 are assessed and their potential health impact is compared to a business as usual scenario. Two types of measures are investigated: operation of underground rail in the city centre and changes in fleet composition. Potential co-benefits from reduced greenhouse gas emissions on public health by the year 2020 are computed utilizing state-of-the-art concentration response functions for PM_x, NO₂ and C₆H₆. Results show significant environmental health and monetary co-benefits when the city metro is coupled with appropriate changes in the traffic composition. Monetary savings due to avoided mortality or leukaemia incidence corresponding to the reduction in PM₁₀, PM_{2.5}, NO₂ and C₆H₆ exposure will be 56.6, 45, 37.7 and 1.0 million Euros respectively. Promotion of 'green' transportation in the city (i.e. the wide use of electric vehicles), will provide monetary savings from the reduction in PM₁₀, PM_{2.5}, NO₂ and C₆H₆ exposure up to 60.4, 49.1, 41.2 and 1.08 million Euros. Overall, it was shown that the respective GHG emission reduction policies resulted in clear co-benefits in terms of air quality improvement, public health protection and monetary loss mitigation.

Keywords : Public health co-benefits, Climate change mitigation, Air pollution, Data fusion, Cities, Urban transport

Atmospheric deposition of particles at a sensitive alpine lake: Size-segregated daily and annual fluxes from passive sampling techniques

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Source: Volume 579, 1 February 2017, Pages 1736-1744
<https://doi.org/10.1016/j.scitotenv.2016.11.117>

Lake Tahoe, a North American alpine lake long appreciated for its clear water and geographic setting, has experienced a trend of declining water clarity due to increasing nutrient and particle inputs. Contributions from atmospheric deposition of particulate matter (PM) could be important, yet they are inadequately quantified. This study established a yearlong deposition monitoring network in the northern Lake Tahoe Basin. Dry deposition was quantified on surrogate surfaces while wet deposition was based on particles suspended in precipitation at 24-hour resolution. The particle size ranges by these passive techniques were 1–64 µm and 0.5–20 µm in diameter for dry

and wet deposition, respectively. Dry deposition of submicrometer (0.5–1 μm) particles was also estimated by extrapolation of a lognormal size distribution. Higher daily number deposition fluxes (NDFdry and NDFwet) were found at a near-shore site, confirming substantial impacts of commercial and tourist activities. The two more isolated sites indicated a uniform regional background. On average, daily NDFdry is about one order of magnitude lower than daily NDFwet. Dry deposition velocities increased rapidly with particle size, as evidenced by collocated measurements of NDFdry and ambient particle number concentrations, though it seems less so for wet deposition due to different scavenging mechanisms. Despite fewer “wet” days than “dry” days during the monitoring period, wet processes dominated seasonal particle deposition, particularly in winter and spring when most precipitation occurred. Adopting sediment (insoluble, inorganic) particle fraction estimates from the literature, this study reports an annual particle flux of $2.9\text{--}5.2 \times 10^{10} \# \text{ m}^{-2} \text{ yr}^{-1}$ for sediment particles with 1–20 μm diameter and $6.1\text{--}11 \times 10^{10} \# \text{ m}^{-2} \text{ yr}^{-1}$ for those with 0.5–20 μm diameter. Implications of these findings to the current knowledge of atmospheric deposition in the Lake Tahoe Total Maximum Daily Load (TMDL) are discussed.

Keywords : Surrogate surface, Scanning electron microscopy, Image analysis, Total maximum daily load, Dry deposition, Wet deposition

Formation, features and controlling strategies of severe haze-fog pollutions in China

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Source: Volume 578, 1 February 2017, Pages 121-138

<https://doi.org/10.1016/j.scitotenv.2016.10.201>

With rapid industrialization and urbanization, China is facing a great challenge with regard to severe fog-haze pollutions, which were characterized by high fine particulate concentration level and visibility impairment. The control strategies for atmosphere pollutions in China were not only cutting-edge topics of atmospheric research, but also an urgent issue to be addressed by the Chinese government and the public. Focused on the core scientific issues of the haze and fog pollution, this paper reviews the main studies conducted in China, especially after 2010, including formation mechanisms, evolution features, and factors contributing to the fog-haze pollutions. Present policy and control strategies were synoptically discussed. The major challenges ahead will be stated and recommendations for future research directions are proposed at the end of this Review.

Keywords : Fog-haze pollution, Fine particles, Health, China

Haze, public health and mitigation measures in China: A review of the current evidence for further policy response

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Source: Volume 578, 1 February 2017, Pages 148-157
<https://doi.org/10.1016/j.scitotenv.2016.10.231>

With rapid economic development, China has been plagued by choking air pollution in recent years, and the frequent occurrence of haze episodes has caused widespread public concern. The purpose of this study is to describe the sources and formation of haze, summarize the mitigation measures in force, review the relationship between haze pollution and public health, and to discuss the challenges, potential research directions and policy options. Haze pollution has both natural and man-made causes, though it is anthropogenic sources that are the major contributors. Accumulation of air pollutants, secondary formation of aerosols, stagnant meteorological conditions, and trans-boundary transportation of pollutants are the principal causes driving the formation and evolution of haze. In China, haze includes gaseous pollutants and fine particles, of which PM_{2.5} is the dominant component. Short and long-term exposure to haze pollution are associated with a range of negative health outcomes, including respiratory diseases, cardiovascular and cerebrovascular diseases, mental health problems, lung cancer and premature death. China has paid increasing attention to the improvement of air quality, and has introduced action plans and policies to tackle pollution, but many interventions have only temporary effects. There may be fierce resistance from industry groups and some government agencies, and often it is challenging to enforce relevant control measures and laws. We discuss the potential policy options for prevention, the need for wider public dialogue and the implications for scientific research.

Keywords : Air pollution, Haze, Public health, Mitigation measure, China

How does elevated ozone reduce methane emissions from peatlands?

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Source: Volume 579, 1 February 2017, Pages 60-71
<https://doi.org/10.1016/j.scitotenv.2016.10.188>

The effects of increased tropospheric ozone (O₃) pollution levels on methane (CH₄) emissions from peatlands, and their underlying mechanisms, remain unclear. In this study, we exposed peatland mesocosms from a temperate wet heath dominated by the sedge *Schoenus nigricans* and *Sphagnum papillosum* to four O₃ treatments in open-top chambers for 2.5 years, to investigate the O₃ impacts on CH₄ emissions and the processes that underpin these responses. Summer CH₄ emissions, were significantly reduced, by 27% over the experiment, due to summer daytime (8 h day⁻¹) O₃ exposure to non-filtered air (NFA) plus 35 ppb O₃, but were not significantly affected by year-round, 24 h day⁻¹, exposure to NFA plus 10 ppb or NFA plus 25 ppb O₃. There was no evidence that the reduced CH₄ emissions in response to elevated summer O₃ exposure were caused by

reduced plant-derived carbon availability below-ground, because we found no significant effect of high summer O₃ exposure on root biomass, pore water dissolved organic carbon concentrations or the contribution of recent photosynthate to CH₄ emissions. Our CH₄ production potential and CH₄ oxidation potential measurements in the different O₃ treatments could also not explain the observed CH₄ emission responses to O₃. However, pore water ammonium concentrations at 20 cm depth were consistently reduced during the experiment by elevated summer O₃ exposure, and strong positive correlations were observed between CH₄ emission and pore water ammonium concentration at three peat depths over the 2.5-year study. Our results therefore imply that elevated regional O₃ exposures in summer, but not the small increases in northern hemisphere annual mean background O₃ concentrations predicted over this century, may lead to reduced CH₄ emissions from temperate peatlands as a consequence of reductions in soil inorganic nitrogen affecting methanogenic and/or methanotrophic activity.

Keywords : CH₄, Mires, Sedge, Sphagnum papillosum, Nitrogen, 13C

Factors influencing the atmospheric concentrations of PCBs at an abandoned e-waste recycling site in South China

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Source: Volume 578, 1 February 2017, Pages 34-39
<https://doi.org/10.1016/j.scitotenv.2016.08.131>

The diurnal atmospheric concentrations of polychlorinated biphenyls (PCBs) were investigated at an abandoned e-waste recycling site in South China during winter and summer. Total PCB concentrations during winter and summer were 27.6–212 and 368–1704 pg/m³ in the particulate phase and 270–697 and 3000–15,500 pg/m³ in the gaseous phase, respectively. Both gaseous and particulate PCB concentrations and compositions exhibited significant difference between winter and summer samples, but no diurnal variations during the measurement period. The correlation analysis between PCB concentrations and meteorological conditions, including atmospheric temperature, humidity, and mixing layer height, suggested that the seasonal variability of atmospheric PCB concentrations was strongly temperature-dependent, while the diurnal variability was probably source-dependent. The temperature-driven variations can also be proved by the significant linear correlation between $\ln P$ and $1/T$ in the Clausius-Clapeyron plot. Although government has implemented controls to reduce e-waste pollution, both the relatively high concentrations of PCBs and the diurnal variation in the air suggested that emissions from occasional e-waste recycling activities may still exist in this recycling area. These results underline the importance of continuing e-waste recycling site management long after abandonment.

Keywords : E-waste, PCBs, Diurnal variation, Seasonal variation, Air

Seasonal and temperature modifications of the association between fine particulate air pollution and cardiovascular hospitalization in New York state

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Source: Volume 578, 1 February 2017, Pages 626-632
<https://doi.org/10.1016/j.scitotenv.2016.11.008>

It is known that extreme temperature and ambient air pollution are each independently associated with human health outcomes. However, findings from the few studies that have examined modified effects by seasons and the interaction between air pollution and temperature on health endpoints are inconsistent. This study examines the effects of short-term PM_{2.5} (particulate matter less than or equal to 2.5 μm in aerodynamic diameter) on hospitalization for cardiovascular diseases (CVDs), its modifications by season and temperature, and whether these effects are heterogeneous across different regions in New York State (NYS). We used daily average temperature and PM_{2.5} concentrations as exposure indicators and performed a time series analysis with a quasi-Poisson model, controlling for possible confounders, such as time-relevant variables and dew point, for CVDs in NYS, 1991–2006. Stratification parametric models were applied to evaluate the modifying effects by seasons and temperature. Across the whole year, a 10- $\mu\text{g}/\text{m}^3$ increment in PM_{2.5} concentration accounted for a 1.37% increase in CVDs (95% confidence interval (CI): 0.90%, 1.84%) in New York City, Long Island & Hudson. The PM_{2.5} effect was strongest in winter, with an additional 2.06% (95% CI: 1.33%, 2.80%) increase in CVDs observed per 10- $\mu\text{g}/\text{m}^3$ increment in PM_{2.5}. Temperature modified the PM_{2.5} effects on CVDs, and these modifications by temperature on PM_{2.5} effects on CVDs were found at low temperature days. These associations were heterogeneous across four PM_{2.5} concentration regions. PM_{2.5} was positively associated with CVD hospitalizations. The short-term PM_{2.5} effect varied with season and temperature levels, and stronger effects were observed in winter and at low temperature days.

Keywords : CVD, Temperature, Season, Modification

Seasonal analysis of the short-term effects of air pollution on daily mortality in Northeast Asia

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Source: Volume 576, 15 January 2017, Pages 850-857
<https://doi.org/10.1016/j.scitotenv.2016.10.036>

The constituents and concentrations of pollutants, individual exposures, and biologic responses to air pollution may vary by season and meteorological conditions. However, evidence regarding seasonality of the acute effects of air pollution on mortality is limited and inconsistent. Herein, we examined seasonal patterns in the short-term associations of particulate matter (PM) smaller than 10 μm (PM₁₀) with daily mortality in 29 cities of three northeast Asian countries. Stratified time-

series models were used to determine whether season altered the effect of PM₁₀ on mortality. This effect was first quantified within each season and at each location using a time-series model, after which city-specific estimates were pooled using a hierarchical Bayesian model. In all data sets, 3,675,348 non-accidental deaths were registered from 1993 to 2009. In Japan, a 10 µg/m³ increase in PM₁₀ was significantly associated with increases in non-accidental mortality of 0.44% (95% confidence interval [CI]: 0.03%, 0.8%) in spring and 0.42% (0.02%, 0.82%) in fall. In South Korea, a 10 µg/m³ increase in PM₁₀ was significantly associated with increases in non-accidental mortality of 0.51% (0.01%, 1.01%) in summer and 0.45% (0.03%, 0.87%) in fall, in cardiovascular disease mortality of 0.96% (0.29%, 1.63%) in fall, and in respiratory disease mortality of 1.57% (0.40%, 2.75%) in fall. In China, a 10 µg/m³ increase in PM₁₀ was associated with increases in non-accidental mortality of 0.33% (0.01%, 0.66%) in summer and 0.41% (0.09%, 0.73%) in winter, in cardiovascular disease mortality of 0.41% (0.08%, 0.74%) in spring and 0.33% (0.02%, 0.64%) in winter, and in respiratory diseases mortality of 0.78% (0.27%, 1.30%) in winter. Our analyses suggest that the acute effect of particulate air pollution could vary seasonally and geographically.

Keywords : Air pollution, Mortality, Particulate matter, Seasonality, Time series

Exposures to and origins of carbonaceous PM_{2.5} in a cookstove intervention in Northern Ghana

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Source: Volume 576, 15 January 2017, Pages 178-192
<https://doi.org/10.1016/j.scitotenv.2016.10.069>

REACCTING (Research on Emissions Air Quality, Climate, and Cooking Technologies in Northern Ghana) was a 200-home cookstove intervention study from 2013 to 2015. Study households were divided into four groups: a control group, a group given two locally made rocket stoves, a group given two Philips forced draft stoves, and a group given a locally made rocket stove and a Philips stove. In a subset of study households, 48-hour PM_{2.5} exposure samples were collected for adults and children, as well as in the primary cooking area. Further, weekly ambient background PM_{2.5} samples were collected for the first nine months of the study. All PM_{2.5} samples were analyzed for elemental and organic carbon (EC/OC), and a subset was also analyzed for organics. Mixed effects modeling was applied to quantify differences in PM exposures between the groups and to assess relationships between exposures and cooking area measurements. Results showed that personal OC exposure for the intervention groups was 56.6% lower than the control group ($p \leq 0.01$). Both intervention groups given Philips stoves had significantly lower EC exposure than the control group (60.6% reduction, $p \leq 0.02$). Only weak relationships were found between personal and cooking area EC or OC. Source apportionment modeling was performed on both the personal/microenvironment and the ambient organics PM_{2.5} data sets to assess the sources of the observed PM. We identified six PM sources. The identified source factors were similar among the data sets, as well as with previous work in Navrongo. Two sources, one characterized by the presence of methoxyphenols, and one by the presence of polyaromatic hydrocarbons and EC, were associated with biomass burning, and accounted for a median of 9.2% of OC and 15.3% of EC personal exposure. Here, we demonstrate the utility of using the cooking-related source

apportionment factors within a mixed effects model for more precise estimation of exposures due to cooking, rather than other combustion sources unrelated to the intervention.

Impacts of household coal and biomass combustion on indoor and ambient air quality in China: Current status and implication

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Source: Volume 576, 15 January 2017, Pages 347-361
<https://doi.org/10.1016/j.scitotenv.2016.10.080>

This review briefly introduces current status of indoor and ambient air pollution originating from household coal and biomass combustion in mainland China. Owing to low combustion efficiency, emissions of CO, PM_{2.5}, black carbon (BC), and polycyclic aromatic hydrocarbons have significant adverse consequences for indoor and ambient air qualities, resulting in relative contributions of more than one-third in all anthropogenic emissions. Their contributions are higher in less economically developed regions, such as Guizhou (61% PM_{2.5}, 80% BC), than that in more developed regions, such as Shanghai (4% PM_{2.5}, 17% BC). Chimneys can reduce ~80% indoor PM_{2.5} level when burning dirty solid fuels, such as plant materials. Due to spending more time near stoves, housewives suffer much more (~2 times) PM_{2.5} than the adult men, especially in winter in northern China (~4 times). Improvement of stove combustion/thermal efficiencies and solid fuel quality are the two essential methods to reduce pollutant emissions. PM_{2.5} and BC emission factors (EFs) have been identified to increase with volatile matter content in traditional stove combustion. EFs of dirty fuels are two orders higher than that of clean ones. Switching to clean ones, such as semi-coke briquette, was identified to be a feasible path for reducing >90% PM_{2.5} and BC emissions. Otherwise, improvement of thermal and combustion efficiencies by using under-fire technology can reduce ~50% CO₂, 87% NH₃, and 80% PM_{2.5} and BC emissions regardless of volatile matter content in solid fuel. However, there are still some knowledge gaps, such as, inventory for the temporal impact of household combustion on air quality, statistic data for deployed clean solid fuels and advanced stoves, and the effect of socioeconomic development. Additionally, further technology research for reducing air pollution emissions is urgently needed, especially low cost and clean stove when burning any type of solid fuel. Furthermore, emission-abatement oriented policy should base on sound scientific evidence to significantly reduce pollutant emissions.

Keywords : Particulate matter emissions, Coal burning emissions, Household stove, Human exposure, Air quality control, Pollution emissions from solid fuel combustion

Effects of landscape composition and pattern on land surface temperature: An urban heat island study in the megacities of Southeast Asia

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Source: Volume 577, 15 January 2017, Pages 349-359
<https://doi.org/10.1016/j.scitotenv.2016.10.195>

Due to its adverse impacts on urban ecological environment and the overall livability of cities, the urban heat island (UHI) phenomenon has become a major research focus in various interrelated fields, including urban climatology, urban ecology, urban planning, and urban geography. This study sought to examine the relationship between land surface temperature (LST) and the abundance and spatial pattern of impervious surface and green space in the metropolitan areas of Bangkok (Thailand), Jakarta (Indonesia), and Manila (Philippines). Landsat-8 OLI/TIRS data and various geospatial approaches, including urban-rural gradient, multiresolution grid-based, and spatial metrics-based techniques, were used to facilitate the analysis. We found a significant strong correlation between mean LST and the density of impervious surface (positive) and green space (negative) along the urban-rural gradients of the three cities, depicting a typical UHI profile. The correlation of impervious surface density with mean LST tends to increase in larger grids, whereas the correlation of green space density with mean LST tends to increase in smaller grids, indicating a stronger influence of impervious surface and green space on the variability of LST in larger and smaller areas, respectively. The size, shape complexity, and aggregation of the patches of impervious surface and green space also had significant relationships with mean LST, though aggregation had the most consistent strong correlation. On average, the mean LST of impervious surface is about 3 °C higher than that of green space, highlighting the important role of green spaces in mitigating UHI effects, an important urban ecosystem service. We recommend that the density and spatial pattern of urban impervious surfaces and green spaces be considered in landscape and urban planning so that urban areas and cities can have healthier and more comfortable living urban environments

Keywords : Urban heat island, Land surface temperature, Impervious surface, Greenspace, Landscape pattern, Megacities

Air quality across a European hotspot: Spatial gradients, seasonality, diurnal cycles and trends in the Veneto region, NE Italy

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Source: Volume 576, 15 January 2017, Pages 210-224
<https://doi.org/10.1016/j.scitotenv.2016.10.042>

The Veneto region (NE Italy) lies in the eastern part of the Po Valley, a European hotspot for air pollution. Data for key air pollutants (CO, NO, NO₂, O₃, SO₂, PM₁₀ and PM_{2.5}) measured over 7 years (2008/2014) across 43 sites in Veneto were processed to characterise their spatial and

temporal patterns and assess the air quality. Nitrogen oxides, PM and ozone are critical pollutants frequently breaching the EC limit and target values. Intersite analysis demonstrates a widespread pollution across the region and shows that primary pollutants (nitrogen oxides, CO, PM) are significantly higher in cities and over the flat lands due to higher anthropogenic pressures. The spatial variation of air pollutants at rural sites was then mapped to depict the gradient of background pollution: nitrogen oxides are higher in the plain area due to the presence of strong diffuse anthropogenic sources, while ozone increases toward the mountains probably due to the higher levels of biogenic ozone-precursors and low NO emissions which are not sufficient to titrate out the photochemical O₃. Data-depth classification analysis revealed a poor categorization among urban, traffic and industrial sites: weather and urban planning factors may cause a general homogeneity of air pollution within cities driving this poor classification. Seasonal and diurnal cycles were investigated: the effect of primary sources in populated areas is evident throughout the region and drives similar patterns for most pollutants: road traffic appears the predominant potential source shaping the daily cycles. Trend analysis of experimental data reveals a general decrease of air pollution across the region, which agrees well with changes assessed by emission inventories. This study provides key information on air quality across NE Italy and highlights future research needs and possible developments of the regional monitoring network.

Keywords : Air pollution, Nitrogen oxides, Particulate matter, Veneto, Trends

A synchronous observation of enhanced aerosol and NO₂ over Beijing, China, in winter 2015

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Despite extensive efforts into the characterization of air pollution during the past decade, concurrent real-time characterization of aerosol and NO₂ as well as satellite observation above the urban canopy in the megacity of Beijing has sparsely been performed to date. We conducted a simultaneous real-time measurement of aerosol and NO₂ in urban Beijing in a pollution episode from November 25, 2015 to December 2, 2015. The aerosol extinction coefficient was measured by a 532 Mie lidar and the NO₂ concentration by a Multi-Axis Differential Optical Absorption Spectroscopy (MAX-DOAS). The highest aerosol extinction coefficient was 1.55 (\pm 0.16) km⁻¹ at 532 m at a height of 0.27 km. The high average aerosol extinction was observed throughout the pollution period, probably due to the favorable static weather conditions under high surface high-pressure system and transportation sources. NO₂ showed similar temporal variations between ground level and 0.3 km, whereas a much weaker diurnal evolution was found for NO₂. The boundary layer structures were observed, and the relatively homogeneous mixture maintained the pollution within the layer under 0.5 km. Ozone Monitoring Instrument (OMI) NO₂ products based on well-known DOAS trace gas fitting algorithm and WRF-Chem model further illustrated the simultaneous enhancement of NO₂ during the observation period. Backward trajectories suggest that long-range pollution transportation from northwest China contributes more to this pollution event.

Keywords : Aerosol, NO₂, Lidar, MAX-DOAS

Ozone pollution in China: A review of concentrations, meteorological influences, chemical precursors, and effects

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Source: Volume 575, 1 January 2017, Pages 1582-1596
<https://doi.org/10.1016/j.scitotenv.2016.10.081>

High concentrations of ozone in urban and industrial regions worldwide have long been a major air quality issue. With the rapid increase in fossil fuel consumption in China over the past three decades, the emission of chemical precursors to ozone—nitrogen oxides and volatile organic compounds—has increased sharply, surpassing that of North America and Europe and raising concerns about worsening ozone pollution in China. Historically, research and control have prioritized acid rain, particulate matter, and more recently fine particulate matter (PM_{2.5}). In contrast, less is known about ozone pollution, partly due to a lack of monitoring of atmospheric ozone and its precursors until recently. This review summarizes the main findings from published papers on the characteristics and sources and processes of ozone and ozone precursors in the boundary layer of urban and rural areas of China, including concentration levels, seasonal variation, meteorology conducive to photochemistry and pollution transport, key production and loss processes, ozone dependence on nitrogen oxides and volatile organic compounds, and the effects of ozone on crops and human health. Ozone concentrations exceeding the ambient air quality standard by 100–200% have been observed in China's major urban centers such as Jing-Jin-Ji, the Yangtze River delta, and the Pearl River delta, and limited studies suggest harmful effect of ozone on human health and agricultural crops; key chemical precursors and meteorological conditions conducive to ozone pollution have been investigated, and inter-city/region transport of ozone is significant. Several recommendations are given for future research and policy development on ground-level ozone.

Keywords : Ozone pollution in China, A review, Meteorological influences, Chemical precursors, Effect on crops and human health

The covariance of air quality conditions in six cities in Southern Germany - The role of meteorology

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Source: Volume 574, 1 January 2017, Pages 1611-1621
<https://doi.org/10.1016/j.scitotenv.2016.08.200>

This paper analyzed air quality in six cities in Southern Germany (Ulm, Augsburg, Konstanz, Freiburg, Stuttgart and Munich), in conjunction with the prevailing synoptic conditions. Air quality was estimated through the calculation of a daily Air Stress Index (ASI) constituted by five independent components, each one expressing the contribution of one of the five main pollutants (PM₁₀, O₃, SO₂, NO₂ and CO) to the total air stress. As it was deduced from ASI components, PM₁₀ from combustion sources and photochemically produced tropospheric O₃ are the most

hazardous pollutants at the studied sites, throughout cold and warm periods respectively, yet PM10 contribute substantially to the overall air stress during both seasons. The influence of anticyclonic high pressure systems, leading to atmospheric stagnation, was associated with increased ASI values, mainly due to the entrapment of PM10. Moderate air stress was generally estimated in all cities however a cleaner atmosphere was detected principally in Freiburg when North Europe was dominated by low pressure systems. Daily events of notably escalated ASI values were further analyzed with backward air mass trajectories. Throughout cold period, ASI episodes were commonly related to eastern airflows carrying exogenous PM10 originated from eastern continental Europe. During warm period, ASI episodes were connected to the arrival of regionally circulated air parcels reflecting lack of dispersion and accumulation of pollutants in accordance with the synoptic analysis.

Keywords : Air quality, Synoptic circulation, Air mass trajectories, Air stress index, South Germany

Forest soils in France are sequestering substantial amounts of carbon

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Source: Volume 574, 1 January 2017, Pages 616-628
<https://doi.org/10.1016/j.scitotenv.2016.09.028>

The aim of this study was to assess whether French forest soils are sources or sinks of carbon and to quantify changes in soil organic carbon (SOC) stocks over time by resampling soil in long-term forest monitoring plots. Within each plot, and for each survey, soils were sampled at five points selected in five subplots and divided into layers. Composite samples were produced for each layer and subplot, then analysed for mass, bulk density and SOC. Linear mixed models were used to estimate SOC changes over 15 years between two soil surveys carried out in 102 plots in France. A factor analysis and a budget approach were also used to identify which factors and processes were primarily responsible for SOC dynamics. Forest soils throughout France substantially accumulated SOC (+ 0.35 MgC ha⁻¹ yr⁻¹) between 1993 and 2012. The SOC sequestration rate declined with stand age and was affected by stand structure. Uneven-aged stands sequestered more SOC than did even-aged stands ($p < 0.001$). For the forest floor, the SOC sequestration rate estimated by the budget approach was in agreement with that based on stock comparison. This increasing SOC stock in the forest floor can be explained by recent changes in certain factors affecting litter decomposition (climate and litter quality). For the mineral soil, the budget approach was unable to replicate the observed SOC sequestration rate, probably because SOC stocks were not yet at equilibrium with litter inputs at the beginning of the monitoring period (contrary to our steady-state assumption). This explanation is also supported by the fact that the SOC sequestration rate decreased with stand age. As the SOC sequestration rate declines with stand age and is higher in uneven-aged stands, forest management has the potential to influence this carbon sink.

Keywords : Soil organic carbonForest, Stand age, Stand structure, Soil carbon budget, Trend analysis

Optical and radiative properties of aerosols over Desalpar, a remote site in western India: Source identification, modification processes and aerosol type discrimination

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Source: Volume 575, 1 January 2017, Pages 612-627
<https://doi.org/10.1016/j.scitotenv.2016.09.023>

Aerosol optical properties are analyzed for the first time over Desalpar (23.74°N, 70.69°E, 30 m above mean sea level) a remote site in western India during October 2014 to August 2015. Spectral aerosol optical depth (AOD) measurements were performed using the CIMEL CE-318 automatic Sun/sky radiometer. The annual-averaged AOD₅₀₀ and Ångström exponent ($\alpha_{440-870}$) values are found to be 0.43 ± 0.26 and 0.69 ± 0.39 , respectively. On the seasonal basis, high AOD₅₀₀ of 0.45 ± 0.30 and 0.61 ± 0.34 along with low $\alpha_{440-870}$ of 0.41 ± 0.27 and 0.41 ± 0.35 during spring (March–May) and summer (June–August), respectively, suggest the dominance of coarse-mode aerosols, while significant contribution from anthropogenic sources is observed in autumn (AOD₅₀₀ = 0.47 ± 0.26 , $\alpha_{440-870}$ = 1.02 ± 0.27). The volume size distribution and the spectral single-scattering albedo also confirm the presence of coarse-mode aerosols during March–August. An overall dominance of a mixed type of aerosols (~ 56%) mostly from October to February is found via the AOD₅₀₀ vs $\alpha_{440-870}$ relationship, while marine aerosols contribute to ~ 18%. Spectral dependence of α and its second derivative (α') are also used for studying the aerosol modification processes. The average direct aerosol radiative forcing (DARF) computed via the SBDART model is estimated to range from -27.08 W m^{-2} to -10.74 W m^{-2} at the top of the atmosphere, from -52.21 W m^{-2} to -21.71 W m^{-2} at the surface and from 10.97 W m^{-2} to 26.54 W m^{-2} within the atmosphere. This atmospheric forcing translates into heating rates of $0.31\text{--}0.75 \text{ K day}^{-1}$. The aerosol properties and DARF are also examined for different trajectory clusters in order to identify the sources and to assess the influence of long-range transported aerosols over Desalpar.

Keywords : Aerosol physical-optical properties, Aerosol types, Radiative forcing, Sector influence, Western India

Tropospheric volatile organic compounds in China

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Source: Volume 574, 1 January 2017, Pages 1021-1043
<https://doi.org/10.1016/j.scitotenv.2016.09.116>

Photochemical smog, characterized by high concentrations of ozone (O₃) and fine particles (PM_{2.5}) in the atmosphere, has become one of the top environmental concerns in China. Volatile organic compounds (VOCs), one of the key precursors of O₃ and secondary organic aerosol (SOA) (an important component of PM_{2.5}), have a critical influence on atmospheric chemistry and subsequently affect regional and global climate. Thus, VOCs have been extensively studied in many

cities and regions in China, especially in the North China Plain, the Yangtze River Delta and the Pearl River Delta regions where photochemical smog pollution has become increasingly worse over recent decades. This paper reviews the main studies conducted in China on the characteristics and sources of VOCs, their relationship with O₃ and SOA, and their removal technology. This paper also provides an integrated literature review on the formulation and implementation of effective control strategies of VOCs and photochemical smog, as well as suggestions for future directions of VOCs study in China.

Keywords : Volatile organic compounds, Ozone, Photochemical smog, China

Air quality perception of pedestrians in an urban outdoor Mediterranean environment: A field survey approach

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Source: Volume 574, 1 January 2017, Pages 663-670

<https://doi.org/10.1016/j.scitotenv.2016.09.090>

Perception plays a significant role on people's response to preventive measures. In the view of public awareness, the aim of this study was to explore factors that affect air quality perception and to reveal its potential patterns. Air quality perception of individuals, in terms of dust and overall air quality, was examined in relation to air pollutants concentrations, meteorological variables, personal characteristics as well as their thermal sensation and health condition. The data used were obtained from environmental measurements, in situ and from stations, and questionnaire surveys conducted in an outdoor urban Mediterranean area, Athens, Greece. The participants were asked to report their air quality perception and thermal sensation based on predefined scales. A thermal index, Physiological Equivalent Temperature (PET), was estimated to obtain an objective measure of thermal sensation. Particulate matter (PM₁₀) and nitrogen oxide (NO) were associated with dust perception. Nitrogen oxides (NO_x) and carbon monoxide (CO) were associated to air quality perception. Age, area of residence, health symptoms and thermal sensation also affected the perception of air quality. Dusty or poor air quality conditions were more likely to be reported when pollutants' concentrations were increased. Younger people, participants residing in the city center, experiencing health symptoms or warm thermal sensation showed a trend towards reporting more unfavorable air quality conditions.

Keywords : Air pollution, Questionnaires, Thermal sensation, Environmental perception, Health effects

Vertical distribution of ambient aerosol extinctive properties during haze and haze-free periods based on the Micro-Pulse Lidar observation in Shanghai

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Source: Volume 574, 1 January 2017, Pages 1502-1511

Ambient aerosols make a significant contribution to the environment and climate through their optical properties. In this study, the aerosol extinction coefficient and Aerosol optical depth (AOD) retrieved using the Fernald Method from the ground-based Micro-Pulse Lidar (MPL) were used to investigate the characteristics of aerosols during haze and haze-free periods in Shanghai. There were 216 haze days including 145 dry haze days, 39 damp haze days and 32 days of both dry and damp haze in Shanghai from March 2009 to February 2010. During the haze periods, aerosols were concentrated mainly below 600 m resulting in the most severe pollution layer in Shanghai. In contrast to the aerosol optical properties during haze-free periods, aerosol extinction coefficients and AOD were larger in the lower altitude (below 1 km) during haze periods. The lowest 1 km contributed 53–72% of the Aerosol optical depth (AOD) below 6 km for the haze periods and < 41% of that for the haze-free periods except summer. According to the analysis of influencing factors, although atmospheric convection was strong in summer which led to reduce the extinction, the highest occurrence of haze with relatively low aerosol extinction most of time was in summer, which resulted from the factors such as higher relative humidity, temperature and more solar radiation causing hygroscopic growth of particles and formation of secondary aerosols; in spring and autumn, there was less haze occurrences because the boundary layer was relatively higher, which allowed pollutants to diffuse more easily, but spring was the second most frequency season of haze due to frequent dust transport from the north; in winter high concentrations of particles and low boundary layer height were not beneficial to the diffusion of pollutants near the surface and caused haze occurrence rather high with high aerosol extinction.

Keywords : Micro-Pulse Lidar, Haze, Haze-free, Extinction, AOD, Influencing factors

Estimating ground-level PM_{2.5} concentrations in Beijing, China using aerosol optical depth and parameters of the temperature inversion layer

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Source: Volume 575, 1 January 2017, Pages 1219-1227

<https://doi.org/10.1016/j.scitotenv.2016.09.186>

The aerosol optical depth (AOD) is widely used to predict surface PM_{2.5} (particles with an aerodynamic diameter smaller than 2.5 μm) concentrations using regression methods, as a way to supplement observations from sparse ground PM_{2.5} monitoring networks. Several meteorological parameters, such as surface humidity, temperature and height of planetary boundary layer (HPBL), are usually combined with AOD to improve the accuracy of the regression model in predicting PM_{2.5} concentrations. In this paper, we investigate the role of the temperature inversion layer in the prediction of PM_{2.5} concentrations by the optimal subset regression method. The result indicates that the optimal subset regression model with the parameters of the depth and temperature difference of the inversion can significantly improve the accuracy of the predictions of surface PM_{2.5} concentrations, compared with the original regression model with one factor of AOD. The determination coefficient (R^2) increases from 0.51 to 0.63, and the Root Mean Square Error (RMSE) decreases from 40.38 to 35.45 $\mu\text{g}/\text{m}^3$. The optimal subset regressions were also built for each season. The temperature difference of the inversion is introduced into the autumn and winter

optimal subset regression, and the depth of the inversion is introduced into the spring optimal subset regression. The contribution of the inversion parameters to the regression model is affected by the different type of the temperature inversion layer present in each season.

Keywords : Aerosol optical depth, PM2.5, Temperature inversion, Optimal subset regression

A new-generation 3D ozone FACE (Free Air Controlled Exposure)

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Source: Volume 575, 1 January 2017, Pages 1407-1414
<https://doi.org/10.1016/j.scitotenv.2016.09.217>

To artificially simulate the impacts of ground-level ozone (O₃) on vegetation, ozone FACE (Free Air Controlled Exposure) systems are increasingly recommended. We describe here a new-generation, three-dimensional ozone FACE, with O₃ diffusion through laser-generated micro-holes, pre-mixing of air and O₃, O₃ generator with integral oxygen generator, continuous (day/night) exposure and full replication. Based on three O₃ levels and assumptions on the pre-industrial O₃ levels, we describe principles to calculate relative yield/biomass and estimate impacts even at lower-than-ambient O₃ levels. The case study is called FO₃X, and is at present the only ozone FACE in Mediterranean climate and one of the very few ozone FACEs investigating more than one stressor at a time. The results presented here will give further impulse to the research on O₃ impacts on vegetation all over the world.

Keywords : Surface ozone, Tropospheric ozone, Ozone exposure, Forests, Crops, Vegetation

Assessment of PM2.5 sources and their corresponding level of uncertainty in a coastal urban area using EPA PMF 5.0 enhanced diagnostics

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Source: Volume 574, 1 January 2017, Pages 155-164
<https://doi.org/10.1016/j.scitotenv.2016.09.047>

Datasets that include only the PM elemental composition and no other important constituents such as ions and OC, should be treated carefully when used for source apportionment. This work is demonstrating how a source apportionment study utilizing PMF 5.0 enhanced diagnostic tools can achieve an improved solution with documented levels of uncertainty for such a dataset. The uncertainty of the solution is rarely reported in source apportionment studies or it is reported partially. Reporting the uncertainty of the solution is very important especially in the case of small datasets. PM2.5 samples collected in Patras during the year 2011 were used. The concentrations of

22 elements ($Z = 11\text{--}33$) were determined using PIXE. Source apportionment analysis revealed that PM_{2.5} emission sources were biomass burning (11%), sea salt (8%), shipping emissions (10%), vehicle emissions (33%), mineral dust (2%) and secondary sulfates (33%) while unaccounted mass was 3%. Although Patras city center is located in a very close proximity to the city's harbor, the contribution of shipping originating emissions was never before quantified. As rotational stability is hard to be achieved when a small dataset is used the rotational stability of the solution was thoroughly evaluated. A number of constraints were applied to the solution in order to reduce rotational ambiguity.

Keywords : Source apportionment, PMF 5.0, PM_{2.5}, PMF uncertainty

Quantifying the relationship between visibility degradation and PM_{2.5} constituents at a suburban site in Hong Kong: Differentiating contributions from hydrophilic and hydrophobic organic compounds

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*Source: Volume 575, 1 January 2017, Pages 1571-1581
<https://doi.org/10.1016/j.scitotenv.2016.10.082>*

Fine particulate matter (PM) is capable of scattering and absorbing light and is the main culprit of visibility degradation. Major PM chemical components have been characterized for their light absorption and scattering efficiency, but separating the organic components has yet to be fully parameterized with light extinction coefficients. In this study, light extinction data and PM_{2.5} chemical composition data were monitored at a suburban site in Hong Kong over a two-year period (2013–2014). Using the IMPROVE formula to reconstruct the light scattering coefficient under-estimates the measured scattering coefficient (slope = 0.85), but explains the data variability well (R^2 : 0.92). A multilinear regression analysis using the 'local' PM_{2.5} composition data and measured extinction coefficients was performed to empirically establish mass scattering and absorption efficiencies (i.e., MSE and MAE) for the different PM_{2.5} components. During this process, the stepwise separation of organic matter (OM) was performed according to water solubility: water soluble organic carbon (WSOC) and water insoluble organic carbon (WISOC); then according to water affinity: hydrophilic carbon (HPI) and hydrophobic carbon (HPO), the latter being the sum of humic-like substance carbon (HULISc) and WISOC. The localized formulas predict the measured extinction coefficients (i.e., σ_{sp} and σ_{ap}) very well (slope = 0.99 for both). The results showed that the dry MSE of ammonium sulfate and ammonium nitrate were comparable with those used in the IMPROVE equation while MSE for OC is noticeably larger in the localized formula (13.1 vs. 7.2 m² g⁻¹). Splitting the OM into different fractions revealed the MSE for hydrophilic carbon (16.1 m² g⁻¹) was distinctly higher than for hydrophobic carbon, including HULIS (11.0 m² g⁻¹) and WISOC (12.8 m² g⁻¹). Regression analysis of light absorption against EC and OC indicates that absorption is not fully accounted for considering only EC. OC also contributes to light absorption.

Keywords : Visibility, Light extinction, PM chemical composition, Organic matter, Hydrophilicity



जहाँ है हरियाली ।
वहाँ है खुशहाली ॥

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