Acid Rain and Atmospheric Pollution (ARAP)

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ABSTRACTS

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CONTENTS

Illicit psychotropic substances in the air: The state-of-art

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The occurrence of psychotropic substances (PSs) in the air is known since long time. Recently, attention was paid to illicit PS, with most investigations undertaken in Italy and Spain. In general, collection of illicit substances was performed through aspirating airborne particulates onto filters; afterwards, gas chromatography or high-performance liquid chromatography coupled with mass spectrometry were applied for the PS evaluation. Over twenty substances could be characterized simultaneously. Cocaine concentrations up to 17 ng m− 3 were observed in Latin America, while this substance was absent (< 0.003 ng m− 3) in Algiers (Algeria) and Pančevo (Serbia). Cannabinoids (comprising the psycho-active principle Δ9-tetrahydrocannabinol [THC]) were high in the winter and very low in the summer (up to 6 ng m− 3 and < 0.1 ng m− 3, respectively). Many other substances (e.g., heroin, ephedrine and drug by-products) occurred less frequently and at lesser extents (< 20 pg m− 3). In Rome (Italy), investigations were carried out in interiors of dwellings, schools, an office and a coffee bar, all sites resulting affected by drugs. Besides, solid phase microextraction methods were applied to detect ketamine and methamphetamine in interiors. The PS concentrations depended on substance, physical–chemical contour, and internal or external type of locations. Air monitoring allows detecting the drug consumption or preparation, because illicit substances prevail in sites frequented by abusers.

Keywords : Psychotropic substances; Indoor pollution; Airborne particulates; Cocaine; Δ9-Tetrahydrocannabinol; Amphetamines

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Modeling and evaluation of urban pollution events of atmospheric heavy metals from a large Cu-smelter

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Source: *Science of The Total Environment, Volume 539, 1 January 2016, Pages 17–25*

Metal smelting and processing are highly polluting activities that have a strong influence on the levels of heavy metals in air, soil, and crops. We employ an atmospheric transport and dispersion model to predict the pollution levels originated from the second largest Cu-smelter in Europe. The model predicts that the concentrations of copper (Cu), zinc (Zn), and arsenic (As) in an urban area close to the Cu-smelter can reach 170, 70, and 30 ng m− 3, respectively. The model captures all the observed urban pollution events, but the magnitude of the elemental concentrations is predicted to

be lower than that of the observed values; ~ 300, ~ 500, and ~ 100 ng m− 3 for Cu, Zn, and As, respectively. The comparison between model and observations showed an average correlation coefficient of 0.62 ± 0.13. The simulation shows that the transport of heavy metals reaches a peak in the afternoon over the urban area. The under-prediction in the peak is explained by the simulated stronger winds compared with monitoring data. The stronger simulated winds enhance the transport and dispersion of heavy metals to the regional area, diminishing the impact of pollution events in the urban area. This model, driven by high resolution meteorology (2 km in horizontal), predicts the hourly-interval evolutions of atmospheric heavy metal pollutions in the close by urban area of industrial hotspot.

Keywords : Heavy metal; Industrial emission; Particulate matter; HYSPLIT, Atmospheric model

Enhanced removal of petroleum hydrocarbons using a bioelectrochemical remediation system with pre-cultured anodes

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Source: *Science of The Total Environment ,Volume 539, 1 January 2016, Pages 61–69*

Bioelectrochemical remediation (BER) systems such as microbial fuel cells (MFCs) have recently emerged as a green technology for the effective remediation of petroleum hydrocarbon contaminants (PH) coupled with simultaneous energy recovery. Recent research has shown that biofilms previously enriched for substrate degrading bacteria resulted in excellent performance in terms of substrate removal and electricity generation but the effects on hydrocarbon contaminant degradation were not examined. Here we investigate the differences between enriched biofilm anodes and freshly inoculated new anodes in diesel fed single chamber mediatorless microbial fuel cells (DMFC) using various techniques for the enhancement of PH contaminant remediation with concomitant electricity generation. An anodophilic microbial consortium previously selected for over a year through continuous culturing with a diesel concentration of about 800 mg l− 1 and which now showed complete removal of this concentration of diesel within 30 days was compared to that of a freshly inoculated new anode MFC (showing 83.4% removal of diesel) with a simultaneous power generation of 90.81 mW/m2 and 15.04 mW/m2 respectively. The behaviour of pre-cultured anodes at a higher concentration of PH (8000 mg l− 1) was also investigated. Scanning electron microscopy observation revealed a thick biofilm covering the pre-cultured anodic electrode but not the anode from the freshly inoculated MFC. High resolution imaging showed the presence of thin 60 nm diametre pilus-like projections emanating from the cells. Anodic microbial community profiling confirmed that the selection for diesel degrading exoelectrogenic bacteria had occurred. Identification of a biodegradative gene (*alk*B) provided strong evidence of the catabolic pathway used for diesel degradation in the DMFCs.

Keywords **:** Bioelectrochemical remediation; Diesel-fed microbial fuel cell; Pre-cultured MFC; *Alk*B gene

Association between indoor air pollutant exposure and blood pressure and heart rate in subjects according to body mass index

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Source: *Science of The Total Environment* , Volume 539*, 1 January 2016, Pages 271–276*

This study investigates the effects of high body mass index (BMI) of subjects on individual who exhibited high cardiovascular disease indexes with blood pressure (BP) and heart rate (HR) when exposed to high levels of indoor air pollutants. We collected 115 office workers, and measured their systolic blood pressure (SBP), diastolic blood pressure (DBP) and HR at the end of the workday. The subjects were divided into three groups according to BMI: 18–24 (normal weight), 24–27 (overweight) and > 27 (obese). This study also measured the levels of carbon dioxide (CO2), total volatile organic compounds (TVOC), particulate matter with an aerodynamic diameter less than 2.5 μm (PM2.5), as well as the bacteria and fungi in the subjects' work-places. The pollutant effects were divided by median. Two-way analysis of variance (ANOVA) was used to analyze the health effects of indoor air pollution exposure according to BMI. Our study showed that higher levels of SBP, DBP and HR occurred in subjects who were overweight or obese as compared to those with normal weight. Moreover, there was higher level of SBP in subjects who were overweight or obese when they were exposed to higher levels of TVOC and fungi ($p < 0.05$). We also found higher value for DBP and HR with increasing BMI to be associated with exposure to higher TVOC levels. This study suggests that individuals with higher BMI have higher cardiovascular disease risk when they are exposed to poor indoor air quality (IAQ), and specifically in terms of TVOC.

Keywords : Indoor air quality; Obesity; Blood pressure; Heart rate

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Five-year trends of selected halogenated flame retardants in the atmosphere of Northeast China

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Source**:** *Science of The Total Environment , Volume 539, 1 January 2016, Pages 286–293*

This study collected 227 pairs of gas phase and particle phase air samples in a typical urban city of Northeast China from 2008 to 2013. Four alternative halogenated flame retardants for polybrominated diphenyl ethers (PBDEs) were analyzed, namely 2-ethylhexyl 2,3,4,5-tetrabromobenzoate (EHTBB), bis (2-ethylhexyl) tetrabromophthalate (BEHTBP), syn-dechlorane plus (syn-DP) and anti-dechlorane plus (anti-DP). The average concentrations for EHTBB and BEHTBP were 5.2 ± 20 and 30 ± 200 pg/m3, respectively, while for syn-DP and anti-DP were 1.9 ± 5.1 and 5.8 ± 18 pg/m3, respectively. Generally, they were frequently detected in the particle phase, and the gas/particle partitioning suggested they were the maximum partition chemicals. The fractional abundance of EHTBB (fEHTBB) and syn-DP (fsyn) were comparable with those in other studies. Strong local sources were identified based on the air parcel backward trajectories and the

potential source contribution function. The concentrations of these chemicals were significantly increased during this sampling campaign, possibly suggesting their increasing usages from 2008 to 2013 in China.

Keywords: Alternative halogenated flame retardants; Potential source contribution function;Temporal trend; Atmosphere; China

Comprehensive approach to understand the association between diurnal temperature range and mortality in East Asia

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Source: *Science of the Total Environment , Volume 539, 1 January 2016, Pages 313–321*

An adverse association between diurnal temperature range (DTR) and mortality has been suggested, but with variable relationships in different cities. Comprehensive approaches to understanding the health effects of DTR using multinational data are required. We investigated the association between DTR and cause-specific mortality in an age-specific population and assessed the dependency of the health effects of DTR on geographic and climatic factors. Poisson generalized linear regression analyses with allowances for over-dispersion were applied to daily DTR and cause-specific mortality data from 30 cities in China, Japan, Korea, and Taiwan between 1979 and 2010, adjusted for various climatic and environmental factors. City-specific effects of DTR were estimated and summarized for the overall effects using geographic and climatic determinants in a meta-analysis. For all-cause, circulatory, and respiratory mortality, the greatest city-specific effects per 1 °C DTR were found in Tianjin, China (1.80%; 95% confidence interval [CI]: 0.48, 3.14); Tangshan, China (2.25%; 95% CI: 0.65, 3.87); and Incheon, Korea (2.84%; 95% CI: 0.04, 5.73), respectively, and overall effects across 30 cities were 0.58% (95% CI: 0.44, 0.72), 0.81% (95% CI: 0.60, 1.03), and 0.90% (95% CI: 0.63, 1.18), respectively. Using quartile cutoff values for climatic (DTR, and mean temperature) and geographic (latitude, and longitude) characteristics, we divided the 30 cities into 4 different groups and conducted a meta-analysis within the groups using either a random or fixed effects model. Adverse effects of DTR were more pronounced for those aged ≥ 65 years and varied according to geographic, longitudinal (0.07%; 95% CI: 0.05, 0.10), and climatic characteristics and the scale of DTR (0.33%; 95% CI: 0.12, 0.55) for overall all-cause mortality. The DTR is a risk factor affecting human health, depending on geographic location and the temperature variation, with particular vulnerability in aged populations.

Keywords: Climatic determinant; Diurnal temperature range; Lag effect; Mortality; Temperature variatio

Mixing ratio and carbon isotopic composition investigation of atmospheric CO2 in Beijing, China

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Source: *Science of The Total Environment , Volume 539, 1 January 2016, Pages 322–330*

The stable isotope composition of atmospheric CO2 can be used as a tracer in the study of urban carbon cycles, which are affected by anthropogenic and biogenic CO2components. Continuous measurements of the mixing ratio and δ13C of atmospheric CO2 were conducted in Beijing from Nov. 15, 2012 to Mar. 8, 2014 including two heating seasons and a vegetative season. Both δ13C and the isotopic composition of source CO2(δ13CS) were depleted in the heating seasons and enriched in the vegetative season. The diurnal variations in the CO2 mixing ratio and δ13C contained two peaks in the heating season, which are due to the effects of morning rush hour traffic. Seasonal and diurnal patterns of the CO2 mixing ratio and δ13C were affected by anthropogenic emissions and biogenic activity. Assuming that the primary CO2 sources at night (22:00–04:00) were coal and natural gas combustion during heating seasons I and II, an isotopic mass balance analysis indicated that coal combustion had average contributions of 83.83 ± 14.11% and 86.84 ± 12.27% and that natural gas had average contributions of 16.17 ± 14.11% and 13.16 ± 12.27%, respectively. The δ13C of background CO2 in air was the main error source in the isotopic mass balance model. Both the mixing ratio and δ13C of atmospheric CO2 had significant linear relationships with the air quality index (AQI) and can be used to indicate local air pollution conditions. Energy structure optimization, for example, reducing coal consumption, will improve the local air conditions in Beijing.

Keywords : Isotope ratio infrared spectroscopy (IRIS); δ13C; Keeling plot; Atmospheric CO2 source

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Assessing public health burden associated with exposure to ambient black carbon in the United States

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Source**:** *Science of The Total Environment , Volume 539, 1 January 2016, Pages 515–525*

Black carbon (BC) is a significant component of fine particulate matter (PM2.5) air pollution, which has been linked to a series of adverse health effects, in particular premature mortality. Recent scientific research indicates that BC also plays an important role in climate change. Therefore, controlling black carbon emissions provides an opportunity for a double dividend. This study quantifies the national burden of mortality and morbidity attributable to exposure to ambient BC in the United States (US). We use GEOS–Chem, a global 3-D model of atmospheric composition to estimate the 2010 annual average BC levels at 0.5 × 0.667° resolution, and then re-grid to 12-km grid resolution across the continental US. Using PM2.5 mortality risk coefficient drawn from the American Cancer Society cohort study, the numbers of deaths due to BC exposure were estimated for each 12-km grid, and then aggregated to the county, state and national level. Given evidence that BC particles may pose a greater risk on human health than other components of PM2.5, we also

conducted sensitivity analysis using BC-specific risk coefficients drawn from recent literature. We estimated approximately 14,000 deaths to result from the 2010 BC levels, and hundreds of thousands of illness cases, ranging from hospitalizations and emergency department visits to minor respiratory symptoms. Sensitivity analysis indicates that the total BC-related mortality could be even significantly larger than the above mortality estimate. Our findings indicate that controlling BC emissions would have substantial benefits for public health in the US.

Keywords : Air pollution; Black carbon; Mortality; Public health burden

What are the most fire-dangerous atmospheric circulations in the Eastern-Mediterranean? Analysis of the synoptic wildfire climatology

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Source: *Science of The Total Environment , Volume 539, 1 January 2016, Pages 536–545*

Wildfire management is closely linked to robust forecasts of changes in wildfire risk related to meteorological conditions. This link can be bridged either through fire weather indices or through statistical techniques that directly relate atmospheric patterns to wildfire activity. In the present work the COST-733 classification schemes are applied in order to link wildfires in Greece with synoptic circulation patterns. The analysis reveals that the majority of wildfire events can be explained by a small number of specific synoptic circulations, hence reflecting the synoptic climatology of wildfires. All 8 classification schemes used, prove that the most fire-dangerous conditions in Greece are characterized by a combination of high atmospheric pressure systems located N to NW of Greece, coupled with lower pressures located over the very Eastern part of the Mediterranean, an atmospheric pressure pattern closely linked to the local Etesian winds over the Aegean Sea. During these events, the atmospheric pressure has been reported to be anomalously high, while anomalously low 500 hPa geopotential heights and negative total water column anomalies were also observed. Among the various classification schemes used, the 2 Principal Component Analysis-based classifications, namely the PCT and the PXE, as well as the Leader Algorithm classification LND proved to be the best options, in terms of being capable to isolate the vast amount of fire events in a small number of classes with increased frequency of occurrence. It is estimated that these 3 schemes, in combination with medium-range to seasonal climate forecasts, could be used by wildfire risk managers to provide increased wildfire prediction accuracy.

Keywords : Wildfires; Synoptic circulation; Classification methods; COST-733Site-specific criteria; Historic species composition; Criteria maximum concentration (CMC); Criteria continuous concentration (CCC); Water-effect ratio

Variance of indoor radon concentration: Major influencing factors

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Source: *Science of The Total Environment , Volume 541, 15 January 2016, Pages 155–160*

Variance of radon concentration in dwelling atmosphere is analysed with regard to geogenic and anthropogenic influencing factors. Analysis includes review of 81 national and regional indoor radon surveys with varying sampling pattern, sample size and duration of measurements and detailed consideration of two regional surveys (Sverdlovsk oblast, Russia and Niška Banja, Serbia). The analysis of the geometric standard deviation revealed that main factors influencing the dispersion of indoor radon concentration over the territory are as follows: area of territory, sample size, characteristics of measurements technique, the radon geogenic potential, building construction characteristics and living habits. As shown for Sverdlovsk oblast and Niška Banja town the dispersion as quantified by GSD is reduced by restricting to certain levels of control factors. Application of the developed approach to characterization of the world population radon exposure is discussed.

Keywords :Radon; Indoor; Survey; GSD

Regional contribution to PM1 pollution during winter haze in Yangtze River Delta, China

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Source: Science of The Total Environment , Volume 541, 15 January 2016, Pages 161–166

To quantify regional sources contributing to submicron particulate matter (PM1) pollution in haze episodes, on-line measurements combining two modeling methods, namely, positive matrix factorization (PMF) and backward Lagrangian particle dispersion modeling (LPDM), were conducted for the period of one month in urban Nanjing, a city located in the western part of Yangtze River Delta (YRD) region of China. Several multi-day haze episodes were observed in December 2013. Long-range transport of biomass burning from the southwestern YRD region largely contributed to PM1 pollution with more than 25% of total organics mass in a lasting heavy haze. The LPDM analysis indicates that regional transport is a main source contributing to secondary low-volatility production. The high-potential source regions of secondary low-volatility production are mainly located in areas to the northeast of the city. High aerosol pollution was mainly contributed by regional transport associated with northeastern air masses. Such regional transport on average accounts for 46% of total NR-PM1 with sulfate and aged low-volatility organics being the largest fractions (> 65%).

Keywords :Haze; Submicron particulate matter; Components; Regional source; Yangtze River Delta

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Real-Time Determination of Absorptivity of Ambient Particles in Urban Aerosol in Budapest, Hungary

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Source: *Volume 16, No. 1, January 2016, Pages 1-10 doi: 10.4209/aaqr.2015.05.0356*

The absorption properties of ambient urban aerosols as well as their size distributions were measured in real-time using a dual wavelength optical particle spectrometer (DWOPS). A photoacoustic spectrometer (PAS) was simultaneously used to directly measure light absorptivity of the particles in question along with an Aethalometer and a commercial optical particle counter (OPC). The sizing performance of the DWOPS was compared with the commercial OPC unit showing very good agreement. The data from DWOPS, PAS and Aethalometer instruments are presented and show the potential of the DWOPS technique for a direct and simple assessment of the absorbing part of the complex refractive index of ambient particles as a function of particle size.

Keywords: Atmospheric particles; Light absorption; Refractive index; Soot aerosol.

A Two-Stage Virtual Impactor for In-Stack Sampling of PM2.5 and PM10 in Flue Gas of Stationary Sources

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Source: *Volume 16, No. 1, January 2016, Pages 36-45 doi: 10.4209/aaqr.2015.06.0383*

Two ISO standard methods for in-stack sampling and measurement of PM2.5 and PM10 mass concentrations in flue gas from stationary sources were published in 2009 (ISO 23210, conventional cascade impactors) and 2012 (ISO 13271, virtual impactors). The performances of these two methods in terms of PM2.5 separation efficiency and the accuracy of measured mass concentration were compared at the same sampling point and conditions both using laboratory-scale model flue gas with different dust concentrations and using real flue gas sampled from a test facility for pulverized coal combustion. The virtual impactor showed very satisfactory performance for PM2.5 mass concentration and separation efficiency within the investigated range of mass concentrations and ambient conditions. The conventional cascade impactor method overestimated PM2.5 mass concentration by more than 25% due to particle bounce and re-entrainment of coarse

particles from the collection plates. During in-stack PM2.5 sampling from coal combustion flue gas with reactive components at high temperature, the use of greased plates with the conventional impactor caused overestimation of mass concentration, even when grease with high temperature endurance was used. The use of a quartz-fiber filters on the impaction plates reduced overestimation but particle bounce and re-entrainment still remained.

Keywords: PM2.5; PM10; Mass concentration; Stationary sources; Virtual impactor; Conventional impactor; Particle bounce.

A Key Indicator of Transboundary Particulate Matter Pollution Derived from Indonesian Peatland Fires in Malaysia

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Source: *Volume 16, No. 1, January 2016, Pages 69-78 doi: 10.4209/aaqr.2015.04.0215*

We characterized ambient total suspended particulates (TSP) based on ground-based samplings in Malaysia during non-haze days and haze ones affected by Indonesian peatland fires. Furthermore, a key indicator of Indonesian peatland fire was determined based on chemical characterization of TSP in Malaysia. TSP samples were chemically analyzed to determine organic carbon (OC), elemental carbon (EC), inorganic ions, and biomarkers (solvent-extractable organic compounds derived from biomass burning). Regarding OC and EC, concentrations of OC1 and OP (pyrolyzed OC) defined by IMPROVE_A protocol increased remarkably during the haze episodes. On the contrary, there were no significant differences in concentrations of OC4, EC, and EC fractions between the haze and non-haze samples. Regarding inorganic ions, sulfate and ammonium concentrations increased in strong haze days, however, it is difficult to use these compounds as indicators for Indonesian peatland fires in light haze days due to the partial overlapping of the variation ranges of sulfate and ammonium concentrations in non-haze days. Concentrations of many biomarkers derived from cellulose, hemicellulose, and lignin pyrolysis products were significantly increased during strong haze days but not during light haze days except p-hydroxybenzoic acid. We proposed the OP to OC4 ratio as a potential indicator of transboundary haze pollution from Indonesian peatland fires at the receptor sites even in light haze.

Keywords: Carbonaceous aerosols; Air pollution; Biomass burning.

Secondary PM2.5 in Zhengzhou, China: Chemical Species Based on Three Years of Observations

Jia Wang, Xiao Li, Wenkai Zhang, Nan Jiang, Ruiqin Zhang, Xiaoyan Tang

Source: *Volume 16, No. 1, January 2016, Pages 91-104) doi: 10.4209/aaqr.2015.01.0007*

The chemical properties and secondary components of PM2.5 were investigated in the city of Zhengzhou, China. Water-soluble ionic species (Na+, NH4+, K+, Mg2+, Ca2+, F–, Cl–, NO3– and SO42–) contents, carbonaceous components (organic carbon (OC) and elemental carbon (EC)) in PM2.5 were measured for three years. The EC tracer method was used to estimate the secondary organic carbon (SOC) content, and the Interagency Monitoring of Protected Visual Environments formula was used to estimate light extinction due to the chemical composition of PM2.5. The annual mean concentrations of PM2.5 were 186, 180 and 218 μg m–3 in 2011, 2012 and 2013, respectively. These concentrations were 5–6 times greater than the National Ambient Air Quality Standards of China (annual value of 35 μg m–3) and indicated the presence of severe PM2.5 pollution in Zhengzhou. Particulate organic matter (OM) contributed the most (18–26%) to the annual average PM2.5, followed by SO42– (14–19%), NO3– (10–11%), NH4+ (8–9%) and EC (3%). From 2011 to 2013, the contributions of OM and SO42– increased by 8% and 3%, respectively. The higher sulfur oxidation ratio indicated the formation of significant amounts of secondary inorganic aerosols (SIA), particularly during the summer and spring. Obvious SOC enrichment occurred during the winter and autumn. In addition, SIA and secondary organic aerosols accounted for 26–50% and 4–21% of the PM2.5 by mass, respectively. An investigation of the secondary species revealed that secondary aerosols played a dominant role in the total PM2.5 mass and the decrease in visibility. The secondary aerosols ((NH4)2SO4 + NH4NO3 + SOC) accounted for 80% of bext. The main secondary aerosols that led to poor visibility in Zhengzhou were (NH4)2SO4 and NH4NO3.

Keywords: PM2.5; Water-soluble ionic species; Carbonaceous components; Secondary aerosols; Light extinction coefficient.

Contribution of Biomass Burning to Carbonaceous Aerosols in Mexico City during May 2013

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Source: *Volume 16, No. 1, January 2016, Pages 114-124 doi: 10.4209/aaqr.2015.01.0030*

During the springtime fire season, wildfires and agricultural burning represent a potentially large contribution to air quality degradation in the Mexico City Metropolitan Area (MCMA). PM10 filter samples were collected at six different stations in May 2013, the month with the maximum reported regional fire counts from 2002 to 2013. Two regimes were identified considering changes in predominant wind direction and precipitation patterns inside MCMA. The filter samples were analyzed for water-soluble organic carbon (WSOC) and the biomass burning tracers including levoglucosan (LEV) and water-soluble potassium (WSK+). LEV concentrations correlated positively with ambient concentrations of PM2.5 and PM10 (R2 = 0.61 and R2 = 0.46, respectively). Strong correlations were also found between WSOC and LEV (R2= 0.94) and between WSK+ and LEV (R2 = 0.75). PM2.5 accounted for 60% of the PM10 mass concentrations. Our speciated measurements accounted for 37% of the total PM10 mass concentration and ~60% of the PM2.5 mass concentrations; the missing mass was attributed to crustal material (soil or dust) and carbonaceous aerosols that were not segregated into the WSOC fraction. Average LEV/WSOC ratios ranged from 0.015 in the first, smokier and drier part of the month, to 0.006 during the rainier end of the measurement period. Using previously reported LEV/WSOC emissions ratios, the estimated biomass burning contributions to WSOC ranged from 7–23% assuming LEV is stable in the atmosphere, and 8–57% when accounting for LEV photochemical degradation in the atmosphere. Thus, our findings indicate that primary emissions from biomass burning sources represent significant contributions to ambient WSOC and PM in MCMA during the springtime fire season.

Keywords: Levoglucosan; Water-soluble organic carbon; Potassium; Air quality.

Local and Regional Contributions to Black Carbon Aerosols in a Mid-Sized City in Southern Brazil

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Source: *Volume 16, No. 1, January 2016, Pages 125-137 doi: 10.4209/aaqr.2015.06.0388*

Black carbon (BC) concentrations were monitored at three sites (suburban, street canyon and urban rooftop) in a mid-sized Brazilian city, from August 2014 to January 2015. The suburban site presented weak diurnal cycles, suggesting little influence of motorized traffic, but distinctively large 95th percentile concentrations, reaching values as large as 11.04 and 3.34 μg m−3 in the evenings of the dry and wet season (respectively), likely attributable to local waste burning. Moreover, higher BC concentrations at the suburban site were observed throughout the dry period, primarily caused by long-range transport (LRT) of smoke from the central part of Brazil and neighboring countries, carried by WNW, SSW and NNE winds. Local traffic was by far the most important source of BC in street canyon, with mean hourly peaks of 5.8 μg m−3 (at 7:00) and 4.6 μg m−3 (18:00), coinciding with rush hour periods. The rooftop data showed a mean peak of 1.4 μg m−3 at 7:00, reflective of traffic on a busy avenue adjacent to the site. Meteorological data clustered into groups of similar air temperature (Tair) and relative humidity (RH) showed that BC concentrations were highest (18.3

μg m−3) at the suburban site during the evenings of dry (RH ca. 20%) and hot days (maximum Tair ca. 30°C). Diurnal concentrations in the canyon and rooftop were linked to traffic patterns and showed no clear linkage to meteorological conditions. This study shows that the BC concentrations in the city are highly variable and that air quality diminishes considerably due to sporadic waste burning and LRT of biomass smoke, even in neighborhoods with little motor traffic. While air pollution due to transboundary smoke is more difficult to abate, these results suggest that targeting local backyard burning and traffic volume would lead to a depletion of BC concentrations in the city.

Keywords: Domestic waste burning; Traffic pollutants; Long-range transport; Air quality.

Surface Ozone Variability and Trend over Urban and Suburban Sites in Portugal

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The surface ozone time series is analyzed for the seasonal and inter-annual variations and the trend in the following three categories (a) monthly mean (b) 8 hr monthly mean and (c) daily maximum monthly mean measured at 8 (urban/suburban) sites in Portugal for the period 2000–2010. The inter-annual variation of the monthly mean surface ozone time series showed an year to year variation with the highest value in May 2009 (95 μg m−3) at Monte Chãos and the lowest in Jan 2002 (17 μg m−3) at Paio Pires. The trend analysis of (1) original surface ozone time series and of (2) deseasonalized surface ozone time series for all the three data set categories and for all the 8 sites was performed and found to be statistically significant at 6 sites for monthly mean and 8 hr monthly mean and at 5 sites for daily maximum monthly meanusing the non-parametric Mann-Kendall test. The analysis of original surface ozone time series showed a positive trend at 6 out of 8 sites, but the results were not statistically significant for most of the sites due to the presence of the annual cycle masking the actual trend values. However, the analysis of deseasonalized surface ozone time series showed a statistically significant increasing trend at 7 out of 8 sites with high Z values. The positive trends found in the deseasonalized surface ozone time series were in the range 0.44 up to 1.42 μg m−3 year−1 for the monthly mean surface ozone time series (7 stations), 0.33 up to 1.43 μg m−3 year−1 in the case of the 8 hr monthly mean surface ozone time series (7 stations) and 0.66 up to 1.55 μg m−3 year−1 in the daily maximum monthly mean surface ozone time series (6 stations).

Keywords: Surface Ozone; Ozone variability; Ozone trend; Deseasonalization.

The Influence of Spatial Variability of Critical Conversion Point (CCP) in Production of Ground Level Ozone in the Context of Tropical Climate

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Source: *Volume 16, No. 1, January 2016, Pages 153-165 doi: 10.4209/aaqr.2015.04.0225*

Critical conversion point (CCP) is a very crucial step in production of the ground level O3 chemistry. Thus, a multivariate analysis was applied on the dataset of nine selected locations in Malaysia from 1999 to 2010. It incorporated hierarchical agglomerative cluster analysis (HACA) to explore the spatial variability of CCP and principal component analysis (PCA) to determine the major sources of the air pollutants that influence ozone CCP. High variability in CCP was observed between the monitoring stations that occurred during critical conversion time (CCT) from 8:00 a.m. to 11:00 a.m. The HACA results grouped the nine monitoring stations into three different clusters, based on the characteristics of ozone concentrations during CCT period. Results of PCA for the three clusters showed that the contributions to O3 level variation during CCT by meteorological variables (UVB, temperature, relative humidity, and wind speed) are higher at 51.6%, 48.5%, and 33.3% than that of primary air pollutants (NO2, SO2, PM10) at 19.2%, 21.4%, and 15.2% for cluster 1, cluster 2, and cluster 3, respectively. Therefore, applying a targeted spatial control strategy for ground level O3 precursors during the CCT period is a crucial step.

Keywords: NO2 photolysis; NO titration; Critical conversion point; Multivariate analysis.

Neuro-Fuzzy Approach to Forecast NO2 Pollutants Addressed to Air Quality Dispersion Model over Delhi, India

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Source: *Volume 16, No. 1, January 2016, Pages 166-174 doi: 10.4209/aaqr.2015.04.0249*

Air pollution forecasting is the most important environmental issue in urban areas as it is useful to assess the effects of air pollutants on human health. It has been observed that the air pollution has been increased above the standard level in the urbanized area of Delhi and will be a major problem in the next few years. Therefore, the main objective of the present study is to develop the model that can forecast daily concentrations of air pollutions in one-day advance. In the present study, the artificial intelligence based Neuro-Fuzzy (NF) model has been proposed for air quality forecasting and the concentration of nitrogen dioxide (NO2) pollutant has been chosen for analysis. The available meteorological variables viz. temperature, pressure, relative humidity, wind speed and direction, visibility and the estimated concentrations through AERMOD. The application of

introducing AERMOD aims to improve the forecasting ability of model on the basis the emissions from anthropogenic sources. The training and validation have been made with the eight and two year's available seasonal daily data respectively. The evaluation of the model has been made by comparing its results with observed values as well as other statistical models like MLR and ANN, which reveals that the NF model is performing well and can be used for operational use.

Keywords: AERMOD; Artificial neural network; Delhi; Neuro-Fuzzy; NO2; Statistical measure.

Emissions from residential combustion considering end-uses and spatial constraints: Part II, emission reduction scenarios

Ekbordin Winijkul, Tami C. Bond

Source: *Atmospheric Environment,Volume 124, Part A, January 2016, Pages 1–11*

Cooking, heating, and other activities in the residential sector are major sources of indoor and outdoor air pollution, especially when solid fuels are used to provide energy. Because of their deleterious effects on the atmosphere and human health, multinational strategies to reduce emissions have been proposed. This study examines the effects of some possible policies, considering realistic factors that constrain mitigation: end-uses, spatial constraints involving proximity to forest or electricity, existing technology, and assumptions about user behavior. Reduction scenarios are applied to a year-2010, spatially distributed baseline of emissions of particulate matter, black carbon, organic carbon, nitrogen oxides, methane, non-methane hydrocarbons, carbon monoxide, and carbon dioxide. Scenarios explored are: (1) cleanest current stove, where we assume that existing technology in each land type is applied to burn existing fuels; (2) stove standards, where we assume that stoves are designed to meet performance standards; and (3) clean fuels, where users adopt the cleanest fuels plausible in each land type. We assume that people living in forest access areas continue to use wood regardless of available fuels, so the clean-fuels scenario leads to a reduction in emissions of 18–25%, depending on the pollutant, across the study region. Cleaner stoves preferentially affect land types with forest access, where about half of the fuel is used; emission reductions range from 25 to 82%, depending on the pollutant. If stove performance standards can be met, particulate matter emissions are reduced by 62% for the loosest standards and 95% for the tightest standards, and carbon monoxide is reduced by 40% and 62% for the loosest and tightest standards. Reductions in specific regions and countries depend on the existing fuel mixture and the population division among land types, and are explored for Latin America, Africa, East Asia, South Asia, and Southeast Asia.

Keywords: Residential emissions; Emission reduction scenarios; Emission standards; Fuel switching; Cookstoves

Seasonal variations in whole-ecosystem BVOC emissions from a subtropical bamboo plantation in China

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Source: *Atmospheric Environment, Volume 124, Part A, January 2016, Pages 12–21*

Isoprene and monoterpene emissions and environmental conditions were measured over a six month period in a Lei bamboo (Phyllostachys violascens) forest in a subtropical region in China. Isoprene and monoterpene emissions were measured using a relaxed eddy accumulation (REA) system on an above-canopy tower. From July to November of 2012, isoprene contributed 99.1% of terpenoid emissions. α-pinene, constituting 0.8% of total observed terpenoid emissions, was the only monoterpene for which a significant flux was detected. Emissions of the sesquiterpenes longifolene and α-cedrene were observed at very low rates. Isoprene and α-pinene emissions exhibited strong diurnal variations, with lower emissions in the morning and late evening, and the highest emissions around noon. BVOC peak emissions typically occurred a few hours after the noon PAR peak and coincided with the daily temperature peak. This behavior can be described reasonably well by the MEGANv2.1 biogenic emission model. During the campaign (i.e., from 7 July, 2012 to 19 Jan., 2013), the mean (and maximum) emission fluxes (mg m−2 h−1) were 0.95 (10.32) for isoprene, 0.010 (0.176) for α pinene, 0.001 (0.063) for longifolene, and 2.6 × 10−4 (0.009) for α-cedrene, respectively. During the winter season, when the ground was covered by organic mulch to increase soil temperature and to increase the yield of bamboo shoot, there was no evident impact on BVOC emissions. The observed seasonal variation followed the general behavior predicted by the MEGANv2.1 model, with lower emissions associated with cooler conditions, but the magnitude of the emission decrease was greater than expected indicating driving variables are missing from the model. Emission factors, representing the emission expected for a Leaf Area Index of 5 at a temperature of 30 °C and PAR of 1500 μmol m−2 s−1, during the peak growing season for this site were 0.008 mg m−2 h−1for α-pinene and 3.3 mg m−2 h−1 for isoprene. The isoprene emission factor is similar to the value (3.6 mg m−2 h−1) for this location in the MEGANv2.1 global biogenic emission model. A second bamboo plantation, containing Moso bamboo (Phyllostachys heterocycla), was investigated and found to have similar isoprene and monoterpene emission rates as Lei bamboo forest. The emission data obtained in this study are the first canopy-scale flux measurements reported for bamboo plantations and demonstrate the potential importance of bamboo isoprene emissions for regional ozone and organic aerosol production.

Keywords: Biogenic volatile organic compounds; Emission flux; Isoprene; Monoterpene; Emission model

Forecasting hourly PM2.5 in Santiago de Chile with emphasis on night episodes

Patricio Perez, , Ernesto Gramsch

Source: *Atmospheric Environment,Volume 124, Part A, January 2016, Pages 22–27*

We present the results of an hourly PM2.5 concentrations forecasting model in Santiago, Chile. The study concentrates on the comparison between model and observed values at the monitoring station with the highest concentrations (Cerro Navia station) for the time period between April and August, which is the season when high concentration episodes are frequent. The forecasting model is a feed forward neural network, The input variables are past values of hourly PM10 and PM2.5 concentrations measured at the city station with the highest values during episodes, concentrations from a neighboring station and some observed and forecasted meteorological variables. Training is performed with 2010 and 2011 data and the model is tested with 2012 values. Information is collected until 7 PM of the present day and percent error forecasting up to 15 h in advance, starting at 8 PM of the present day, is of the order of 30%. Accuracy of forecasting is significantly better than different forms of persistence and may be considered as a useful tool for anticipating episodes.

Keywords: Air quality forecasting; Particulate matter; PM2.5; Neural networks; Meteorology forecast

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On-road particle number measurements using a portable emission measurement system (PEMS)

Jens Gallusa, Ulf Kirchnera, Rainer Vogta, Christoph Börensena, Thorsten Bente

Source: *Atmospheric Environment, Volume 124, Part A, January 2016, Pages 37–45*

In this study the on-road particle number (PN) performance of a Euro-5 direct-injection (DI) gasoline passenger car was investigated. PN emissions were measured using the prototype of a portable emission measurement system (PEMS).

PN PEMS correlations with chassis dynamometer tests show a good agreement with a chassis dynamometer set-up down to emissions in the range of 1•1010 #/km. Parallel on-line soot measurements by a photo acoustic soot sensor (PASS) were applied as independent measurement technique and indicate a good on-road performance for the PN-PEMS. PN-to-soot ratios were 1.3•1012 #/mg, which was comparable for both test cell and on-road measurements.

During on-road trips different driving styles as well as different road types were investigated. Comparisons to the world harmonized light-duty test cycle (WLTC) 5.3 and to European field operational test (euroFOT) data indicate the PEMS trips to be representative for normal driving. Driving situations in varying traffic seem to be a major contributor to a high test-to-test variability of PN emissions. However, there is a trend to increasing PN emissions with more severe driving styles. A cold start effect is clearly visible for PN, especially at low ambient temperatures down to 8 °C.

Keywords: Portable emission measurement system (PEMS); Direct injection gasoline; Particulate emission; PN-to-soot ratio; Cold start effect

Downward transport of ozone rich air and implications for atmospheric chemistry in the Amazon rainforest

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Source: *Atmospheric Environment, Volume 124, Part A, January 2016, Pages 64–76*

From April 2014 to January 2015, ozone (O3) dynamics were investigated as part of GoAmazon 2014/5 project in the central Amazon rainforest of Brazil. Just above the forest canopy, maximum hourly O3 mixing ratios averaged 20 ppbv (parts per billion on a volume basis) during the June–September dry months and 15 ppbv during the wet months. Ozone levels occasionally exceeded 75 ppbv in response to influences from biomass burning and regional air pollution. Individual convective storms transported O3-rich air parcels from the mid-troposphere to the surface and abruptly enhanced the regional atmospheric boundary layer by as much as 25 ppbv. In contrast to the individual storms, days with multiple convective systems produced successive, cumulative ground-level O3 increases. The magnitude of O3 enhancements depended on the vertical distribution of O3 within storm downdrafts and origin of downdrafts in the troposphere. Ozone mixing ratios remained enhanced for > 2 h following the passage of storms, which enhanced chemical processing of rainforest-emitted isoprene and monoterpenes. Reactions of isoprene and monoterpenes with O3 are modeled to generate maximum hydroxyl radical formation rates of 6×106 radicals $cm{-}3s{-}1$. Therefore, one key conclusion of the present study is that downdrafts of convective storms are estimated to transport enough O3 to the surface to initiate a series of reactions that reduce the lifetimes of rainforest-emitted hydrocarbons.

Keywords: Isoprene; Monoterpenes; Air chemistry; Convection; Mesoscale convective storms

Chemical composition and source of fine and nanoparticles from recent direct injection gasoline passenger cars: Effects of fuel and ambient temperature

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Source: *Atmospheric Environment, Volume 124, Part A, January 2016, Pages 77–84*

Particle number, mass, and chemical compositions (i.e., elemental carbon (EC), organic carbon (OC), elements, ions, and organic species) of fine particles emitted from four of the recent direct injection spark ignition (DISI) gasoline passenger cars and a port fuel injection (PFI) gasoline passenger car were measured under Japanese official transient mode (JC08 mode). Total carbon (TC = EC + OC) dominated the particulate mass (90% on average). EC dominated the TC for both hot and cold start conditions. The EC/TC ratios were 0.72 for PFI and 0.88–1.0 (average = 0.92) for DISI vehicles. A size-resolved chemical analysis of a DISI car revealed that the major organic components were the C20–C28 hydrocarbons for both the accumulation-mode particles and nanoparticles. Contribution of engine oil was estimated to be 10–30% for organics and the sum of the measured elements. The remaining major fraction likely originated from gasoline fuel. Therefore, it is suggested that soot (EC) also mainly originated from the gasoline. In experiments using four fuels at three ambient temperatures, the emission factors of particulate mass were consistently higher with regular gasoline than with premium gasoline. This result suggest that the high content of less-volatile compounds in fuel increase particulate emissions. These results suggest that focusing on reducing fuel-derived EC in the production process of new cars would effectively reduce particulate emission from DISI cars.

Keywords: Soot; Engine oil; Emission factor; Organic species; Elemental carbon; Direct injection spark ignition car

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A systematic analysis of PM2.5 in Beijing and its sources from 2000 to 2012

Baolei Lva, Bin Zhanga, Yuqi Bai

Source: *Atmospheric Environment, Volume 124, Part B, January 2016, Pages 98–108*

Particulate matter with an aerodynamic diameter of 2.5 μm or less (PM2.5) is the main air pollutant in Beijing. To have a comprehensive understanding of concentrations, compositions and sources of PM2.5 in Beijing, recent studies reporting ground-based observations and source apportionment results dated from 2000 to 2012 in this typical large city of China are reviewed. Statistical methods were also used to better enable data comparison. During the last decade, annual average concentrations of PM2.5 have decreased and seasonal mean concentrations declined through autumn and winter. Generally, winter is the most polluted season and summer is the least polluted one. Seasonal variance of PM2.5 levels decreased. For diurnal variance, PM2.5 generally increases at night and decreases during the day. On average, organic matters, sulfate, nitrate and ammonium are the major compositions of PM2.5 in Beijing. Fractions of organic matters increased from 2000 to 2004, and decreased afterwards. Fractions of sulfate, nitrate and ammonium decreased in winter and remained largely unchanged in summer. Concentrations of organic carbon and elemental carbon were always higher in winter than in summer and they barely changed during the last decade. Concentrations of sulfate, nitrate and ammonium exhibited significant increasing trend in summer but in reverse in winter. On average they were higher in winter than in summer before 2005, and took a reverse after 2005. Receptor model results show that vehicle, dust, industry, biomass burning,

coal combustion and secondary products were major sources and they all increased except coal combustions and secondary products. The growth was decided both changing social and economic activities in Beijing, and most likely growing emissions in neighboring Hebei province. Explicit descriptions of the spatial variations of PM2.5 concentration, better methods to estimate secondary products and ensemble source apportionments models to reduce uncertainties would remain being open questions for future studies.

Keywords: PM2.5; Beijing; Carbonaceous materials; Inorganic ions; Source apportionments

Comparison of atmospheric nitrous acid during severe haze and clean periods in Beijing, China

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Source: *Atmospheric Environment, Volume 124, Part B, January 2016, Pages 199–206*

Continuous measurements were made in Beijing from 22 February to 2 March in 2014, including a severe haze period and a relatively clean period. The concentration of nitrous acid (HONO) ranged from 0.49 to 3.24 ppbv in the severe haze period and from 0.28 to 1.52 ppbv in the clean period. Daytime mean concentrations of SO2, NO, NO2, and NOxin the severe haze period were 58.7 ppbv, 23.2 ppbv, 48.0 ppbv, 71.2 ppbv, respectively, much higher than those in the clean period. The diurnal variations of HONO, NO, NO2, and NOx were weaker in the severe haze period, whereas O3 diurnal variations were opposite with other species in both periods. Moreover, we found better correlations between HONO and NO2 in the clean period than in the severe haze period. Besides, good correlations between HONO and PM2.5, and between PM2.5 and HONO/NO2 ratio were found when PM2.5 concentrations were <350 μg m−3. When PM2.5 concentrations were >350 μg m−3, HONO remained nearly invariable, with a value of 2.95 ppbv. Additionally, very good correlations between HONO and RH (R2 = 0.839), and between HONO/NO2 ratio and RH (R2 = 0.508) were found when RH was ≤65%. The calculated daytime average unknown HONO source rate was 1.85 ppbv h−1 in the severe haze period, higher than 1.26 ppbv h−1 in the clean period.

Keywords: Nitrous acid; Nitrogen oxides; PM2.5; Haze

Model assessment of atmospheric pollution control schemes for critical emission regions

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Source: *Atmospheric Environment, Volume 124, Part B, January 2016, Pages 367–377*

In recent years, the atmospheric environment in portions of China has become significantly degraded and the need for emission controls has become urgent. Because more international events are being planned, it is important to implement air quality assurance targeted at significant events held over specific periods of time. This study sets Yanqihu (YQH), Beijing, the location of the 2014 Beijing APEC (Asia–Pacific Economic Cooperation) summit, as the target region. By using the atmospheric inversion model FLEXPART, we determined the sensitive source zones that had the greatest impact on the air quality of the YQH region in November 2012. We then used the air-quality model Models-3/CMAQ and a high-resolution emissions inventory of the Beijing-Tianjian-Hebei region to establish emission reduction tests for the entire source area and for specific sensitive source zones. This was achieved by initiating emission reduction schemes at different ratios and different times. The results showed that initiating a moderate reduction of emissions days prior to a potential event is more beneficial to the air quality of Beijing than initiating a high-strength reduction campaign on the day of the event. The sensitive source zone of Beijing (BJ-Sens) accounts for 54.2% of the total source area of Beijing (BJ), but its reduction effect reaches 89%–100% of the total area, with a reduction efficiency 1.6–1.9 times greater than that of the entire area. The sensitive source zone of Huabei (HuaB-Sens.) only represents 17.6% of the total area of Huabei (HuaB), but its emission reduction effect reaches 59%–97% of the entire area, with a reduction efficiency 4.2–5.5 times greater than that of the total area. The earlier that emission reduction measures are implemented, the greater the effect they have on preventing the transmission of pollutants. In addition, expanding the controlling areas to sensitive provinces and cities around Beijing (HuaB-sens) can significantly accelerate the reduction effects compared to controlling measures only in the Beijing sensitive source zone (BJ-Sens). Therefore, when enacting emission reduction schemes, cooperating with surrounding provinces and cities, as well as narrowing the reduction scope to specific sensitive source zones prior to unfavorable meteorological conditions, can help reduce emissions control costs and improve the efficiency and maneuverability of emission reduction schemes.

Keywords: Critical emission regions; Sensitive source zones; Emission reduction schemes;Numerical model

Modeled deposition of fine particles in human airway in Beijing, China

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Source: *Atmospheric Environment,Volume 124, Part B, January 2016, Pages 387–395*

This study aims to simulate depositions of size-segregated particles in human airway in Beijing, China during seasons when fine particulate matter concentrations are high (December 2011 and April 2012). Particle size distributions (5.6–560 nm, electrical mobility diameter) near a major road in Beijing were measured by the TSI Fast Mobility Particle Sizer (FMPS). The information of size distributions provided by FMPS was applied in the Multiple-Path Particle Dosimetry model (MPPD) to quantify number and mass depositions of particles in human airway including extrathoracic (ET), tracheobronchial (TB), and pulmonary (PUL) regions of exposed Chinese in Beijing. Our results show

that under ambient conditions, particle number concentration (NC) deposition in PUL is the highest in the three major regions of human airway. The total particle NC deposition in human airway in winter is higher than that in spring, especially for ultrafine particles (1.8 times higher) while particle mass concentration (MC) deposition is higher in spring. Although particle MC in clean days are much lower than that in heavily polluted days, total particle NC deposition in human airway in clean days is comparable to that in heavily polluted days. NC deposition for nucleation mode particles (10–20 nm, aerodynamic diameter) in clean days is higher than that in heavily polluted days. MC deposition for accumulation mode particles (100–641 nm, aerodynamic diameter) in heavily polluted days is much higher than that in clean days, while that of nucleation mode is negligible. The temporal variation shows that the arithmetic mean and the median values of particle NC and MC depositions in the evening are both the highest, followed by morning and noon, and it is most likely due to increased contribution from traffic emissions.

Keywords: Fine particle; Ultrafine particle; Deposition; Human airway; MPPD model; Beijing

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Ozone reaction with interior building materials: Influence of diurnal ozone variation, temperature and humidity

Donghyun Rima, Elliott T. Gallb, Randy L. Maddalenad, William W. Nazaroff

Source: *Atmospheric Environment, Volume 125, Part A, January 2016, Pages 15–23*

Elevated tropospheric ozone concentrations are associated with increased morbidity and mortality. Indoor ozone chemistry affects human exposure to ozone and reaction products that also may adversely affect health and comfort. Reactive uptake of ozone has been characterized for many building materials; however, scant information is available on how diurnal variation of ambient ozone influences ozone reaction with indoor surfaces. The primary objective of this study is to investigate ozone-surface reactions in response to a diurnally varying ozone exposure for three common building materials: ceiling tile, painted drywall, and carpet tile. A secondary objective is to examine the effects of air temperature and humidity. A third goal is to explore how conditioning of materials in an occupied office building might influence subsequent ozone-surface reactions. Experiments were performed at bench-scale with inlet ozone concentrations varied to simulate daytime (ozone elevated) and nighttime (ozone-free in these experiments) periods. To simulate office conditions, experiments were conducted at two temperatures (22 °C and 28 °C) and three relative humidity values (25%, 50%, 75%). Effects of indoor surface exposures were examined by placing material samples in an occupied office and repeating bench-scale characterization after exposure periods of 1 and 2 months. Deposition velocities were observed to be highest during the initial hour of ozone exposure with slow decrease in the subsequent hours of simulated daytime conditions. Daily-average ozone reaction probabilities for fresh materials are in the respective ranges of (1.7–2.7) × 10−5, (2.8–4.7) × 10−5, and (3.0–4.5) × 10−5 for ceiling tile, painted drywall, and carpet tile. The reaction probability decreases by 7%–47% across the three test materials after two 8-h periods of ozone exposure. Measurements with the samples from an occupied office reveal that deposition

velocity can decrease or increase with time. Influence of temperature and humidity on ozone-surface reactivity was not strong.

Keywords: Deposition velocity; Reaction probability; Exposure; Surface aging; Regeneration

Mercury in the atmosphere and in rainwater at Cape Point, South Africa

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Source: *Atmospheric EnvironmentVolume 125, Part A, January 2016, Pages 24–32*

Mercury measurements were concurrently made in air (Gaseous Elemental Mercury, i.e. GEM) as well as in precipitation samples (Total mercury, i.e. TotHg) over a seven year period (2007–2013) at Cape Point, South Africa, during the rainy seasons (May–October). Eighty-five rain events, almost exclusively associated with cold fronts, have been identified of which 75% reached the Cape Point observatory directly across the Atlantic Ocean from the south, while 19% moved in to the measuring site via the Cape Town metropolitan region. In statistic terms the GEM, TotHg, CO and 222Rn levels within the urban-marine events do not differ from those seen in the marine rain episodes. Over the 2007–2013 period, the May till Oct averages for GEM ranged from 0.913 ng m−3 to 1.108 ng m−3, while TotHg concentrations ranged from 0.03 to 52.5 ng L−1 (overall average: 9.91 ng L−1). A positive correlation (R2 = 0.49, n = 7) has been found between the average annual (May till October) GEM concentrations in air and TotHg concentration in rainwater suggesting a close relationship between the two species. The wetter years are normally associated with higher GEM and TotHg levels. Both GEM and TotHg annual means correlate positively with total annual (May till October) rain depths. If one or two outlier years are removed from the data set, the R2 values increase from 0.23 to 0.10 for GEM and TotHg to 0.97 (n = 5) and 0.89 (n = 5), respectively. The relationship between annual mean GEM and annual precipitation depth also holds for the period 1996–2004 (R2 = 0.6, n = 8) when GEM was measured manually (low resolution data). A positive correlation was also seen between annual average GEM concentrations and the El Niño Southern Oscillation (ENSO) Index (SOI), for the 1996–2004 period (R2 = 0.7, n = 8). For the 2007–2013 periods this relationship was also positive but less pronounced. The relationship between annual precipitation depth and annual SOI suggests that the inter-annual variations of GEM (Hg0) concentration might be caused by large-scale meteorological processes.

Keywords: Gaseous elemental mercury; Atmospheric; Total mercury; Rainwater; Cape Point

Potential effects of using biodiesel in road-traffic on air quality over the Porto urban area, Portugal

Isabel Ribeiro, Alexandra Monteiro, Myriam Lopes

Source: *Atmospheric EnvironmentVolume 125, Part A, January 2016, Pages 78–91*

This work aims to assess the impacts of biodiesel blends use in road-traffic on air quality. In this frame, the air quality numerical modelling system WRF-EURAD was applied over Portugal and the Porto urban area, forced by two emission scenarios (including CO, NOx, PM10, PM2.5, NMVOC, formaldehyde, acetaldehyde, acrolein and benzene): a reference scenario, without biofuels, and a scenario where a B20 fuel (20% biodiesel/80% diesel, v/v) is used by the diesel vehicle fleet. Regarding carbonyl compounds, emission scenarios pointed out that B20 fuel can promote an increase of 20% on formaldehyde, acetaldehyde and acrolein emissions, leading to increments on equivalent ozone production. On the other hand, through the air quality modelling exercise, it was verified that the use of B20 helps in controlling air pollution, improving CO and NO2 concentrations in urban airshed in about 20% and 10%, respectively, taking into account a regional simulation grid. However, according to the urban scale simulation, NO2 levels can increase in about 1%, due to the use of B20, over the Porto urban area. For the remaining studied pollutants, namely PM10 and PM2.5, mean concentrations will be reduced all over the territory, however in a negligible amount of <1%.

Keywords: Biodiesel; Atmospheric pollutant emissions; Numerical modelling; Air quality

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Canopy stomatal uptake of NOX, SO2 and O3 by mature urban plantations based on sap flow measurement

Yanting Hu, Ping Zhao, Junfeng Niu, Zhenwei Sun, Liwei Zhu, Guangyan Ni

Source: *Atmospheric Environment ,Volume 125, Part A, January 2016, Pages 165–177*

Canopy stomatal uptake of NOX (NO, NO2), SO2 and O3 by three mature urban plantations (of Schima superba, Eucalyptus citriodora and Acacia auriculaeformis) were studied using the sap flow-based approach under free atmospheric conditions. The annual mean concentration for NO, NO2, SO2 and O3 were 18.2, 58.1, 12.8 and 42.4 μg m−3, respectively. The atmospheric concentration exhibited a spring or winter maximum for NO, NO2 and SO2, whereas the concentration maximum for O3 occurred in the autumn. Despite the daytime mean canopy stomatal conductance (GC) being positively related with the photosynthetically active radiation (PAR) and negatively with the vapour pressure deficit (VPD), the maximal daytime mean GC did not appear when the PAR was at its highest level or the VPD was at its lowest level because a positive correlation was noted between the daytime mean PAR and VPD ($P < 0.001$) under field conditions. The GC value was regulated by the cooperation of the PAR and VPD. When analysing the respective effect of the PAR or VPD on GC separately, a positive logarithmical correlation was noted between the daytime mean GC and PAR as the following equation:$Gc=a\times lnPAR-b$ ($P < 0.01$), and the daytime mean GC was negatively logarithmically

correlated with the VPD : Gc=Gsref−m×lnVPD($P < 0.001$). The daytime mean GC declined with decreases in the soil water content (SWC) under similar meteorological condition. Differences in the seasonal pattern of the canopy stomatal conductance and atmospheric concentrations led to a differentiated peak flux. The flux for NO, NO2 and SO2 exhibited a spring maximum, whereas the flux maximum for O3 appeared in the autumn or summer. The annual cumulative stomatal flux for NO, NO2, O3 and SO2 was 100.19 ± 3.76, 510.68 ± 24.78, 748.59 ± 52.81 and 151.98 ± 9.33 mg m−2 a−1, respectively. When we focus on the foliar uptake of trace gases, the effect of these gases on the vegetation in turn should be considered, particularly for regions with serious air pollution problems. These trace gases had not yet reached injury levels, except for NO2. Flux-based measurements were better suited for evaluating the risk of O3 damage to vegetation than the exposure-based method.

Keywords: Sap flow; Canopy stomatal conductance; Environmental factors; Stomatal uptake

Dry deposition of particulate matter at an urban forest, wetland and lake surface in Beijing

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Source: *Atmospheric Environment, Volume 125, Part A, January 2016, Pages 178–187*

The dry deposition of particular matters from atmosphere to ecosystems is an undesirable consequence of this pollution while the deposition process is also influenced by different land use types. In current study, concentration of fine particles, coarse particles and meteorological data were collected during the daytime in an artificial forest, wetland and a water surface in the Beijing Olympic Park. Dry deposition velocity, fluxes and vegetation collection were calculated by different models and the results were compared. The results show: (1) the deposition velocity onto the forest canopy was higher than which onto the wetland and the water surface and the velocity varied in different seasons; (2) the fine particles deposited most in the winter while the coarse particles was in the spring; (3) the vegetation collection rates of fine particles were lower than coarse particles, and the forest collected more PMs than the wetland plants.

Keywords: PM pollution; Dry deposition; Vegetation collection; Land use types; Seasons

Long-term trends of dust events over Tibetan Plateau during 1961–2010

Litai Kang, Jianping Huang, Siyu Chen, Xin Wang

Source: *Atmospheric Environment, Volume 125, Part A, January 2016, Pages 188–198*

In this study we analyzed dust events records of surface meteorological stations on Tibetan Plateau (TP) during 1961–2010 and provided the spatial and temporal distribution of dust events. The

occurrence of dust events has significantly decreased since the 1970s. We defined the Tibetan Plateau Dust Index (TPDI) for the most dust active periods, spring and winter, to characterize the large scale variability of dust events over TP. Mann–Kendall test suggested the decreasing trend was possibly an abrupt change in the 1990s. The decline of surface wind speeds could partly explain the decrease of dust events over TP. TPDI is positively correlated to the surface winds, with correlation coefficients of 0.42 for spring and 0.46 for winter, respectively. The averaged number days with strong winds (wind speed greater than 6.5 ms-1) for the 4 selected stations, which were chosen to define TPDI, are significantly correlated with TPDI for both spring (correlation coefficient = 0.69) and winter (correlation coefficient = 0.76) and also showed a deceasing trend. The upward trend of vegetation cover was indicated by the normalized difference vegetation index (NDVI), which can be attributed as another factor driving the decrease of dust events over TP. TPDI is negatively correlated to the NDVI, with correlation coefficients of −0.48 for spring and −0.29 for winter. Additionally, analysis of geopotential height fields and wind fields indicate an enhanced ridge in the north of TP and weakened westerly jet in the low-frequency years of dust events, which also drive the decline of dust events over TP.

Keywords: Tibetan Plateau; Dust event; Plateau dust index; Wind; NDVI

Secondary effects of urban heat island mitigation measures on air quality

Joachim Fallmann, Renate Forkel, Stefan Emeis

Source: *Atmospheric Environment, Volume 125, Part A, January 2016, Pages 199–211*

This study presents numerical simulations analysing the effect of urban heat island (UHI) mitigation measures on the chemical composition of the urban atmosphere. The mesoscale chemical transport model WRF-Chem is used to investigate the impact of urban greening and highly reflective surfaces on the concentrations of primary (CO, NO) as well as secondary pollutants (O3) inside the urban canopy. In order to account for the sub-grid scale heterogeneity of urban areas, a multi-layer urban canopy model is coupled to WRF-Chem. Using this canopy model at its full extend requires the introduction of several urban land use classes in WRF-Chem. The urban area of Stuttgart serves as a test bed for the modelling of a case scenario of the 2003 European Heat Wave. The selected mitigation measures are able to reduce the urban temperature by about 1 K and the mean ozone concentration by 5–8%. Model results however document also negative secondary effects on urban air quality, which are closely related to a decrease of vertical mixing in the urban boundary layer. An increase of primary pollutants NO and CO by 5–25% can be observed. In addition, highly reflective surfaces can increase peak ozone concentration by up to 12% due to a high intensity of reflected shortwave radiation accelerating photochemical reactions.

Keywords: Urban heat island mitigation; Air quality; WRF-Chem; Urban canopy model;Turbulence; Ozone photochemistry

Improving the modeling of road dust levels for Barcelona at urban scale and street level

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Source: *Atmospheric Environment, Volume 125, Part A, January 2016, Pages 231–242*

Road dust emission is an emerging issue in air quality due to the lack of remediation measures in contrast to vehicle exhaust emissions. The evidence of receptor modeling studies allows for quantifying impact on a few receptors, but the high cost of PM chemical speciation data and the questionable representativeness of single monitoring sites, limit considerably the development of population exposure estimates and epidemiologic studies based on georeferenced data. This study attempts to initiate and promote urban-scale dispersion modeling for road dust emissions, which will allow for a more robust estimate of population exposure and health outcomes. The TNO URBIS (URBan Information System) model was applied in the city of Barcelona, implementing a Gaussian line source and a street canyon dispersion model, together with new experimental estimates of road dust emission factors and algorithm to describe the time variability. Annual, daily and hourly road dust contributions were simulated and validated against observation of PM10, mineral dust and hourly PM2.5-10 concentrations. Results show that road dust contributed 9–15% to PM10 levels at background sites, and 23–44% at traffic sites. Highest contributions were modeled in the commercial/residential district where most of population live and work (Eixample) structured by 120 m wide square blocks, separated by roads with >10,000 vehicles per day. Street level contributions rise up to 20 μg/m3 (96% of roads) and an additional 3% of roads within 20–40 μg/m3. Hourly simulations of road dust contributions revealed to benefit from the implementation of the new emission module (Amato et al., 2012), able to describe the exponential recovery of road dust emission potential after rain events, when compared to common approach such as the use of constant emission factor or an ON/OFF approach. Correlation coefficients with observed data varied from 0.61, 0.58 and 0.43 for annual, daily and hourly means, respectively, revealing a clear improvement in terms of both spatial and temporal variability. However, more efforts need to be done in validating the model in different climatic scenarios and evaluating the seasonal variation of road dust emissions, due to droughts or Saharan dust events.

Keywords: URBIS; Emission factor; Rain; Temporal variation; Non-exhaust; Resuspension

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Greenhouse gas emissions, soil quality, and crop productivity from a mono-rice cultivation system as influenced by fallow season straw management

Wei Liu, Saddam Hussain, Lishu Wu, Ziguo Qin, Xiaokun Li, Jianwei Lu, Fahad Khan, Weidong Cao, Mingjian Geng

Source: *Environmental Science and Pollution Research, January 2016, Volume 23, Issue 1, pp 315–328*

Straw management during fallow season may influence crop productivity, soil quality, and greenhouse gas (GHG) emissions from rice field. A 3-year field experiment was carried out in central China to examine the influence of different fallow season straw management practices on rice yield, soil properties, and emissions of methane (CH4) and nitrous oxide (N2O) from a mono-rice cultivation system. The treatments comprised an unfertilized control (CK), inorganic fertilization (NPK), rice straw burning in situ (NPK + RSB), rice straw mulching (NPK + RSM), and rice straw strip mulching with green manuring (NPK + RSM + GM). The maximum rice yield, soil organic carbon, soil total nitrogen, and available potassium were observed in NPK + RSM + GM treatment. Compared with NPK, the NPK + RSM + GM recorded 9 % higher grain yield averaged across 3 years. However, NPK + RSM and NPK + RSB were statistically similar with NPK regarding grain yield. The NPK + RSM and NPK + RSM + GM recorded significantly higher CH4 emission during rice growing season as well as winter fallow; however, the response of N2O emissions was variable. The NPK + RSM and NPK + RSM + GM were statistically similar for annual cumulative CH4 and N2O emissions. The NPK + RSM + GM recorded 103 and 72 % higher straw-induced net economic benefits and soil organic carbon sequestration rate, and reduced net global warming potential by 27 % as compared with NPK + RSM. Considering the benefits of soil fertility, higher crop productivity, and environmental safety, the NPK + RSM + GM could be the most feasible and sustainable option for mono-rice cultivation system in central China.

Keywords: Global warming potential, Greenhouse gas emissions, Rice productivity, Soil organic carbon, Straw management, Winter fallow

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Spatial and temporal trends of ozone distribution in the Jizerské hory Mountains of the Czech Republic

Iva Hůnov, Petra Stoklasová, Jana Schovánková, Alena Kulasová

Source: *Environmental Science and Pollution Research January 2016, Volume 23, Issue 1, pp 377–387*

We present results of the 5-year monitoring of ambient O3 concentrations in a Central European medium altitude mountain forested area. O3 levels were measured at 11 sites between 714 and 1000 m a.s.l. in 2006–2010 vegetation seasons using Ogawa diffusive samplers. Our results reveal that O3 exposure in the Jizerské hory Mts. was relatively high and comparable with polluted sites in Southern Europe and in higher altitudes. O3

concentrations differed significantly between individual sites and in individual years. O3 concentrations showed clear dependence on altitude at sites with similar aspect. Its gradient for the entire 5-year period under review equaled 3.5 ppb/100 m of altitude, ranging between nearly 5 ppb/100 m of altitude in 2006 and nearly 3 ppb/100 m of altitude in 2010. O3 concentrations at the site with northern aspect were consistently significantly lower than at the site at similar altitude with southern aspect. O3 concentrations measured at the forest edge were consistently lower than those measured at the same site but at the forest clearing. It is evident that the macro-setting of the O3 monitoring site is crucial for obtaining reliable results with high representativeness for the area.

Keywords: Ambient ozone, Diffusive sampler, s Spatial and temporal variations

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Removal efficiency of particulate matters at different underlying surfaces in Beijing

Jiakai Liu, Lichun Mo. Lijuan Zhu, Yilian Yang, Jiatong Liu, Dongdong Qiu, Zhenming Zhang, Jinglan Liu

Source: *Environmental Science and Pollution Research, January 2016, Volume 23, Issue 1, pp 408–417*

Particulate matter (PM) pollution has been increasingly becoming serious in Beijing and has drawn the attention of the local government and general public. This study was conducted during early spring of 2013 and 2014 to monitor the concentration of PM at three different land surfaces (bare land, urban forest, and lake) in the Olympic Park in Beijing and to analyze its effect on the concentration of meteorological factors and the dry deposition onto different land cover types. The results showed that diurnal variation of PM concentrations at the three different land surfaces had no significant regulations, and sharp short-term increases in PM10 (particulate matter having an aerodynamic diameter <10 μm) occurred occasionally. The concentrations also differed from one land cover type to another at the same time, but the regulation was insignificant. The most important meteorological factor influencing the PM concentration is relative humidity; it is positively correlated with the PM concentration. While in the forests, the wind speed and irradiance also influenced the PM concentration by affecting the capture capacity of trees and dry deposition velocity. Other factors were not correlated with or influenced by the PM concentration. In addition, the hourly dry deposition in unit area (μg/m2) onto the three types of land surfaces and the removal efficiency based on the ratio of dry deposition and PM concentration were calculated. The results showed that the forest has the best removal capacity for both PM2.5 (particulate matter

having an aerodynamic diameter <2.5 μm) and PM10 because of the faster deposition velocity and relatively low resuspension rate. The lake's PM10 removal efficiency is higher than that of the bare land because of the relatively higher PM resuspension rates on the bare land. However, the PM2.5 removal efficiency is lower than that of the bare land because of the significantly lower dry deposition velocity.

Keywords: Particulate matter, Meteorological factors, Underlying surfaces, Dry deposition, Removal efficiency

The impact of PM2.5 on asthma emergency department visits: a systematic review and meta-analysis

Jingchun Fan, Shulan Li, Chunling Fan, Zhenggang Bai, Kehu Yang

Source: *Environmental Science and Pollution Research, January 2016, Volume 23, Issue 1, pp 843–850*

Although the relationship between asthma and exposure to fine particulate matter (PM2.5) has been frequently measured, reported conclusions have not been consistent. As emergency department (ED) visits are an effective way to estimate health outcomes for people with asthma and short-term exposure to PM2.5, this review systematically searched five databases without language or geographical restrictions from inception to January 13, 2015 to study the impact of PM2.5 on asthma ED visits. A random-effects model was used to calculate the pooled risk ratio (RR) and 95 % confidence intervals (CI). With respect to short-term effects, asthma ED visits increased at higher PM2.5 concentrations (RR 1.5 % per 10 μg/m3; 95 % CI 1.2–1.7 %), and children were more susceptible (3.6 % per 10 μg/m3; 95 % CI 1.8, 5.3 %) than adults (1.7, 95 % CI 0.7 %, 2.8 %) to increased PM2.5; the ED visits increased during the warm season by 3.7 % (95 % CI 0.5, 6.9 %) per 10 μg/m3 increase in PM2.5, which was higher than the corresponding increase during the cold season (2.6, 95 % CI 0.7–4.6 %). This demonstrates that ambient PM2.5 has an adverse impact on asthma ED visits after short-term exposure and that children are a high-risk population when PM2.5 concentrations are high, particularly in warm seasons, during which measures should be taken to prevent PM2.5.

Keywords: Fine particulate matter (PM2.5), Asthma Emergency department (ED), Short-term exposure, Systematic review, Meta-analysis

Forecasting PM10 in Algiers: efficacy of multilayer perceptron networks

Hamza Abderrahim, Mohammed Reda Chellali, Ahmed Hamou

Source: *Environmental Science and Pollution Research, January 2016, Volume 23, Issue 2, pp 1634–1641*

Air quality forecasting system has acquired high importance in atmospheric pollution due to its negative impacts on the environment and human health. The artificial neural network is one of the most common soft computing methods that can be pragmatic for carving such complex problem. In this paper, we used a multilayer perceptron neural network to forecast the daily averaged concentration of the respirable suspended particulates with aerodynamic diameter of not more than 10 μm (PM10) in Algiers, Algeria. The data for training and testing the network are based on the data sampled from 2002 to 2006 collected by SAMASAFIA network center at El Hamma station. The meteorological data, air temperature, relative humidity, and wind speed, are used as inputs network parameters in the formation of model. The training patterns used correspond to 41 days data. The performance of the developed models was evaluated on the basis index of agreement and other statistical parameters. It was seen that the overall performance of model with 15 neurons is better than the ones with 5 and 10 neurons. The results of multilayer network with as few as one hidden layer and 15 neurons were quite reasonable than the ones with 5 and 10 neurons. Finally, an error around 9 % has been reached.

Keywords: Pollution, Neural network, Multilayer perceptron, PM10

Economic development and multiple air pollutant emissions from the industrial sector

Hidemichi Fujii, Shunsuke Managi

Source: *Environmental Science and Pollution Research, February 2016, Volume 23, Issue 3, pp 2802–2812*

This study analyzed the relationship between economic growth and emissions of eight environmental air pollutants (carbon dioxide (CO2), methane (CH4), nitrous oxide (N2O), nitrogen oxide (NOx), sulfur oxide (SOx), carbon monoxide (CO), non-methane volatile organic compound (NMVOC), and ammonia (NH3)) in 39 countries from 1995 to 2009. We tested an environmental Kuznets curve (EKC) hypothesis for 16 individual industry sectors and for the total industrial sector. The results clarified that at least ten individual industries do not have an EKC relationship in eight air pollutants even though this relationship was observed in the country and total industrial sector level data. We found that the key industries that dictated the EKC relationship in the country and the total industrial sector existed in CO2, N2O, CO, and NMVOC emissions. Finally, the EKC turning point and the relationship between economic development and trends of air pollutant

emissions differ among industries according to the pollution substances. These results suggest inducing new environmental policy design such as the sectoral crediting mechanism, which focuses on the industrial characteristics of emissions.

Keywords: Environmental Kuznets curve, Air pollution Industrial sector , Key industry , Sectoral crediting mechanism, Industrial characteristics

Ambient PM2.5, Black Carbon, and Particle Size-Resolved Number Concentrations and the Ångström Exponent Value of Aerosols during the Firework Display at the Lantern Festival in Southern Taiwan

Chi-Chi Lin, Li-Sing Yang, Yu-Hsiang Cheng

Source: *Volume 16, No. 2, February 2016, Pages 373-387 doi: 10.4209/aaqr.2015.09.0569*

The Yanshui Beehive Firework Festival is a traditional folk activity in Southern Taiwan held during the Lantern Festival, and it is the third largest folk celebration in the world. During this festival, more than 200 firecracker towers with hundreds of thousands of firecrackers are ignited, posing a risk to public health because of an abrupt increase in particulate matter concentrations within a short period. In this study, real-time variations of PM2.5 (particles with an aerodynamic diameter less than or equal to 2.5 μm), black carbon (BC), and particle number concentrations were monitored before and during the firework display to understand the effect of the firework display on the short-term air quality. The hourly average concentrations of PM2.5, BC, total particle number, and ultrafine particle number during the firework display (episode period) were 146.9 μg m−3, 2639 ng m−3, 3.37 × 104 # cm−3, and 1.18 × 104 # cm−3, respectively. These values were 6.9, 2.3, 5.9, and 3.7 times greater than those during the same period on reference days (nonepisode period), respectively. The measured ultraviolet BC (UVBC) and BC concentrations indicated that aerosols were bound with ultraviolet-absorbing organic compounds, which were abundant, during the episode period. BC aerosols during the episode originated from vehicular traffic and firecracker burning, and the absorption Ångström exponent value was 1.4. The particle number size distribution during the episode period showed a major accumulation mode and a minor Aitken mode of 180 and 63 nm, respectively. This particle number size distribution pattern was considerably different from that in the nonepisode period. During the episode period, particle coagulation played a crucial role in removing particles in the nucleation and Aitken modes in the ambient air at high particle number concentrations.

Keywords: Firework display; PM2.5; BC; Particle number; Ångström exponent value.

Emission Characteristics and Concentrations of Gaseous Pollutants in Environmental Moxa Smoke

Jian Huang, Yu-Hai Huang, Min Yee Lim, Jing-Ying Zhang, Bai-Xiao Zhao, Xiao-Bin Jin, Jin-Liang Zhang

Source: Volume 16, No. 2, February 2016, Pages 398-404 doi:10.4209/aaqr.2015.01.0027

The burning of moxa floss in moxibustion constitutes a major anthropogenic source of many gaseous pollutants, which has been associated with many different negative environmental health effects. The aim of the present study is to systemically study the concentration of gaseous pollutants emitted from different types of moxa floss combustion and present key information in abbreviated tabular form to assist in the assessment of air quality in moxibustion clinics and contribute to the safety evaluation of moxibustion. Sampling was divided into pre-combustion, combustion and post-combustion phases. The pollutants determined were carbon monoxide (CO), carbon dioxide (CO2), sulphur dioxide (SO2) and nitrogen dioxide (NO2) produced by burning three types of moxa floss samples. The average post-combustion concentrations for CO, CO2 and NO2 in moxibustion clinics were 9.333 ppm, 0.138% and 10.556 μg m–3, respectively. SO2 was below detectable limit. NO2 concentration decreased during post-combustion, possibly as a result of reactions from moxa floss combustion. The levels of target gaseous pollutants from 4 g of moxa floss combustion were not produced in quantities that exceeded present international air quality standards and occupational exposure limits. Data from our study is important for the recognition and control of occupational and non-occupational gaseous exposure and for the assessment of air quality in moxibustion clinics by professional authorities.

Keywords: Moxibustion; Gaseous pollutants; Moxa smoke.

Diagnosis of Photochemical Ozone Production Rates and Limiting Factors in Continental Outflow Air Masses Reaching Fukue Island, Japan: Ozone-Control Implications

Yugo Kanaya, Hiroshi Tanimoto, Yoko Yokouchi, Fumikazu Taketani, Yuichi Komazaki, Hitoshi Irie, Hisahiro Takashima, Xiaole Pan, Susumu Nozoe, Satoshi Inomata

Source: *Volume 16, No. 2, February 2016, Pages 430-441 doi:10.4209/aaqr.2015.04.0220*

Asian continental outflow air masses reach western Japan in the springtime, carrying high levels of ozone produced over the Asian continent, and facilitating in-situ production. In this study, in-situ production was highlighted; the rate and limiting factors of net ozone production were diagnosed at Fukue Island, a remote island west of Japan, on 17 days during May–June 2009, when the continental outflow air mass arrived, using an observation-based modeling approach. The average ozone production was estimated to be 6.8 ppb per day. Information on the chemical status of the arriving air mass is important, because it affects how further ozone production in the air mass occurs after precursor addition from Japanese domestic emissions. The main limiting factor of ozone production for such air masses was usually nitrogen oxides (NOx), suggesting that domestic NOx emission control is important in reducing further ozone production. Volatile organic compounds (VOCs) also increased the ozone production rate, and occasionally (14% of time) became the dominant controlling factor. This analysis implies that the VOC reduction legislation recently enacted by the Japanese government should be effective. VOC-limited conditions occurred particularly when the air mass traveled within 6–8 h, via the Korean Peninsula. The uncertainty in the radical chemistry mechanism governing ozone production had a non-negligible impact, but the main conclusion relevant to policy was not altered. When chain termination was augmented by $HO_2 + NO/NO_2$ reactions in the presence of H_2O and by heterogeneous loss of HO_2 on aerosol particle surfaces, as recently verified or hypothesized, the daily ozone production rate decreased by up to 24%, and the fraction of hours when the VOC-limited condition occurred varied from 14% to 13–26%.

Keywords: In-situ ozone photochemistry; East Asia; Policy implications; Observation-based model.

Residential Biomass Burning Emissions over Northwestern Himalayan Region of India: Chemical Characterization and Budget Estimation

Mohit Saxena, Sudhir Kumar Sharma, Nidhi Tomar, Humaira Ghayas, Avirup Sen, Rohtash Singh Garhwal, Naresh Chandra Gupta, Tuhin Kumar Mandal

Source: *Volume 16, No. 3, March 2016, Pages 504-518 doi: 10.4209/aaqr.2015.04.0237*

In the present study, we have determined the emission factors (EF) and estimated the emission of particulate matter (PM), organic carbon (OC), elemental carbon (EC), polycyclic aromatic hydrocarbons (PAHs), water soluble inorganic constituents (WSIC) and trace gases such as SO_2, NO and NO_2 from the combustion of biomass fuels (FW: fuel wood and DC: dung cake) used in rural sectors for cooking over Himachal Pradesh (HP), representing the Northwestern Himalayan region of India. The average EFs of PM estimated from FW and DC were 3.44 ± 2.38 and 11.43 ± 1.13 g kg^{-1}, respectively. OC and EC emission ranged from 0.106 to 3.55 g kg^{-1} and 0.07 to 0.90 g kg^{-1}, respectively for variety of biomass fuels. Total emission of PAHs from DC (44.37 mg kg^{-1}) and FW (43.25 mg kg^{-1}) noted in this study was almost similar. Similarly, the average EFs of NOx from FW and DC were 0.59 ± 0.49 g kg^{-1} and 0.34 ± 0.18 g kg^{-1}, respectively. FWs have comparatively higher SO_2 emission (average: 0.43 ± 0.38 g kg^{-1}) than from DC (average: 0.23 ± 0.15 g kg^{-1}).

Among anionic inorganic constituents emitted from FW, maximum EF was noted for Cl– (0.30 ± 0.26 g kg–1). Similarly for cations, highest EF was noted of K+ (0.20 ± 0.09 g kg–1). Ca2+ and Na+ were the major cationic species identified in plumes of DC burning. Utilizing total annual consumption of biomass fuels and EFs of particulates and trace gases determined in the present study over HP and in the past study (Saud et al., 2011, 2012) over Uttarakhand, budget estimates of PM, OC, EC, TC, PAHs, SO2 and NOx have been determined over the Northwestern Himalayan region. Total annual emission estimated over Northwestern Himalayan region are as: PM (18.32 ± 9.53 Gg), OC (4.38 ± 2.31 Gg), EC (1.39 ± 0.55 Gg) and trace gases (SO2: 1.47 ± 1.0 Gg; NOx: 1.77 ± 1.31 Gg).

Keywords: Particulate matter; Trace gases; Emission factor; Budget estimation; Northwestern Himalayan region.

Biomass Combustion a Dominant Source of Carbonaceous Aerosols in the Ambient Environment of Western Himalayas

Ajay Kumar, Arun K. Attri

Source: *Volume 16, No. 3, March 2016, Pages 519-529 doi: 10.4209/aaqr.2015.05.0284*

Use of biomass combustion as primary energy source emit substantial amounts of carbonaceous aerosols (CA) in the Himalayan environment. Any understanding regarding the impact of CA on human health and climate requires a reliable estimation of compositional variability of CA associated carbon forms: Elemental carbon (EC), Organic carbon (OC), and Light absorbing organic carbon (LAOC). This investigation spanning over 14 months was undertaken in the rural part of the Western Himalayas to estimate temporal variability in the ambient aerosol load (PM10, PM2.5), CA associated carbon forms. All CA associated carbon forms were part of PM2.5 size fraction, their significantly high concentrations in winter corresponded with the high biomass combustion. Source apportionment of CA done on the basis of Char-EC/Soot-EC estimates showed that > 90% of the EC was Char-EC contributed by biomass and coal combustion in winter. Estimates of K+ (tracer for biomass combustion) showed a strong association with CA associated carbon forms. The estimated values of CA associated carbon forms during winter matched with the reported values of emission factors for biomass burning. Both the mass and composition of ambient aerosol were predominantly contributed by biomass combustion in the region.

Keywords: Biomass combustion; Carbonaceous aerosol; Elemental carbon; Organic carbon; Western Himalayas.

Chemical Characterization of PM1 at a Regional Background Site in the Western Mediterranean

Nuria Galindo, Eduardo Yubero, Jose F. Nicolás, Javier Crespo, Rubén Soler

Source: *Volume 16, No. 3, March 2016, Pages 530-541 doi: 10.4209/aaqr.2015.05.0302*

From February 2011 to September 2012, PM1 samples were collected at the regional background station of Mt. Aitana, located near the eastern coast of the Iberian Peninsula at 1558 m a.s.l. Samples were subsequently analyzed to determine the major chemical composition (elemental and organic carbon, secondary inorganic ions and oxalate). The seasonal patterns of the concentrations of PM1 and its main components and the influence of long-range transport of dust from the Sahara desert were studied in this work. PM1 was mainly composed of organic matter and ammonium sulfate, while EC and nitrate were minor components. Concentrations ranged from 3.4 μg m−3 in winter to 5.8 μg m−3 during summer. This seasonal cycle is typical of high mountain sites, which are generally above the planetary boundary layer during winter time. All the analyzed components exhibited the same seasonal pattern except nitrate, which showed minimum values in summer. This is most likely the result of the decomposition of NH_4NO_3 favored by the higher summer temperatures. Due to the close proximity to the African continent, PM1 levels significantly increased during Saharan dust intrusions. The concentrations of sulfate were 35% higher during dust events since the formation of secondary ammonium sulfate is favored by heterogeneous reactions on the surface of mineral particles.

Keywords: PM1; High mountain; Western Mediterranean; Secondary ions; OC; EC.

Increased PM Concentrations during a Combined Wildfire and Saharan Dust Event Observed at High-Altitude Sonnblick Observatory, Austria

Gerhard Schauer, Anne Kasper-Giebl, Griša Močnik

Source: *Volume 16, No. 3, March 2016, Pages 542-554 doi: 10.4209/aaqr.2015.05.0337*

A period of increased particulate matter concentrations was observed at the high-altitude Sonnblick Observatory in August 2013. Trajectory analysis, wildfire maps and the evaluation of aerosol measurements revealed a combined and sometimes alternating influence of long-range transport of Saharan dust and emissions of wildfires. The occurrence of Saharan dust was confirmed by an increase of coarse particle number concentration and a negative exponent of the single scattering albedo wavelength dependence, determined by Nephelometer and Aethalometer measurements. During time periods less influenced by Saharan dust, number concentration of

accumulation mode particles increased and a marked correlation of aerosol mass concentrations and CO mixing ratios was observed. By analyzing the wavelength dependence of the absorption coefficients determined with a seven wavelength Aethalometer, the influence of the two aerosol sources was decoupled. Therefore, absorption exponents of 3 and 1.3 were assumed for Saharan dust and wildfires, respectively. Mass concentrations of particulate matter caused by Saharan dust and wildfire emissions were estimated, with the contribution of Saharan dust to overall particulate matter mass ranging from 5% to 80%.

Keywords: Saharan dust; Wildfires; Background aerosols; Long range transport; Absorption exponent.

Non-Methane Volatile Organic Compounds in the Background Atmospheres of a Southern European Mountain Site (Mt. Cimone, Italy): Annual and Seasonal Variability

Eleonora Lo Vullo, Francesco Furlani, Jgor Arduini, Umberto Giostra, Paolo Cristofanelli, Martin L. Williams, Michela Maione

Source: *Volume 16, No. 3, March 2016, Pages 581-592 doi: 10.4209/aaqr.2015.05.0364*

Since January 2010 continuous high-frequency in situ measurements of a range of anthropogenic Non-Methane Volatile Organic Compounds (NMVOCs) has been carried out at the World Meteorological Organisation Global Atmospheric Watch observatory at Mt. Cimone, on the highest peak of the Italian Northern Apennines, at the border between the Po Valley and the Mediterranean Basin. Five-year (2010–2014) time series of eleven NMVOCs, including aromatic and aliphatic species, have been analysed in order to derive average mixing ratios and detect annual and seasonal variability. Recent studies conducted in Europe, mainly in urban areas, have reported a decrease in atmospheric NMVOCs. Here we investigate how the decline in emissions, due to the implementation of air pollution policies, is reflected in the annual variability of NMVOC mixing ratios measured at a regional background location. Analysis of temporal trends for well-mixed conditions showed statistically significant decreases in ethyne, n-pentane and ethyl-benzene, while no significant trends were found for propane, butanes, i-pentane, toluene and xylenes. The seasonal variability of NMVOCs has been studied showing clear seasonal cycles for longer lived compounds and cycles with smaller seasonal amplitudes for shorter- lived species. We used the propane time series to describe the seasonal cycle and to verify to what extent the mixing ratios of propane have been depleted by OH oxidation. We found that, during the summer, different transport times to the receptors and different source distribution are the main responsible for the relatively low integrated OH concentrations at Mt. Cimone.

Keywords: Volatile hydrocarbons; Air quality; Hydroxyl radical; Long-term observations.

Long Range Transport and Wet Deposition Fluxes of Major Chemical Species in Snow at Gulmarg in North Western Himalayas (India)

Bablu Kumar, Sudha Singh, Gyan Prakash Gupta, Farooq Ahmad Lone, Umesh Chandra Kulshrestha

Source: *Volume 16, No. 3, March 2016, Pages 606-617 doi: 10.4209/aaqr.2015.01.0056*

The study reports snow chemistry and long range transport of pollutants at Gulmarg in north-western Himalayan region of India during winters of 2012–2013. The pH of snowmelt varied between 5.16 and 7.68 with an average of 5.90. The frequency distribution of pH of snowmelt showed that the maximum number of samples (31%) had pH between 6.81 and 7.20. However, 12% samples were observed to be acidic (below 5.6). Scavenging ratios (SR) values suggested that crustal components (Ca2+, Mg2+) are efficiently removed by snow. The study site has significant influence of non-marine sources. Wet deposition contributed 34, 27, 45, 71, 8 and 13 meq m–2 fluxes of nssSO42–, NO3–, NH4+, nssCa2+, nssMg2+ and nssK+ respectively. Both local emissions as well as long range transport (LRT) of pollutants were found to be the sources of these ionic species. Backward airmass trajectory calculations showed that this site received airmasses from six major sectors i.e., i) North Atlantic Ocean origin (NAO), ii) African origin (Af), iii) Middle East origin (ME), iv) European origin (Eu), v) Western India origin (InW), vi) Pakistan origin (Pk). The highest average pH (7.58) of the snowfall was noticed during InW airmasses which had the lowest ratios of nssSO42–/nssCa2+ and NO3–/nssCa2+. Very high pH has been observed in precipitation samples at Indian sites due to buffering of acidic components by atmospheric dust rich in CaCO3. The lowest pH (4.94) was noticed for ME airmasses which had the highest nssSO42–/nssCa2+ and NO3–/nssCa2+ ratios. Data of present study was compared with a study reported almost three decades ago. We noticed a drastic increase in the concentrations of anthropogenic components such as nssSO42– (114%), NO3– (109%) and NH4+ (90%). This is probably due to increase in LRT of pollutants as well as local activities during past three decades.

Keywords: Snow chemistry; Himalayan region; Acidic depositions; Wet fluxes; Airmass trajectories.

A Review of More than 20 Years of Aerosol Observation at the High Altitude Research Station Jungfraujoch, Switzerland (3580 m asl)

Nicolas Bukowiecki, Ernest Weingartner, Martin Gysel, Martine Collaud Coen, Paul Zieger, Erik Herrmann, Martin Steinbacher, Heinz W. Gäggeler, Urs Baltensperger

Source: *Volume 16, No. 3, March 2016, Pages 764-788 doi: 10.4209/aaqr.2015.05.0305*

Among the worldwide existing long-term aerosol monitoring sites, the Jungfraujoch (JFJ) belongs to the category where both free tropospheric (FT) conditions and influence from planetary boundary layer (PBL) injections can be observed. Thus, it is possible to characterize free tropospheric aerosol as well as the effects of vertical transport of more polluted air from the PBL. This paper summarizes the current knowledge of the key properties for the JFJ aerosol, gained from the large number of in-situ studies from more than 20 years of aerosol measurements at the site. This includes physical, chemical and optical aerosol properties as well as aerosol-cloud interactions and cloud characteristics. It is illustrated that the aerosol size distribution and the aerosol chemical composition are fairly constant in time due to the long distance from aerosol sources, and that many climate relevant aerosol properties can be derived due to this behavior.

Keywords: Mountain site; Aerosol physical properties; Aerosol optical properties; Aerosol chemical properties; Aerosol-cloud interactions.

Atmospheric Aerosol Elements over the Inland Tibetan Plateau: Concentration, Seasonality, and Transport

Shichang Kang, Pengfei Chen, Chaoliu Li, Bin Liu, Zhiyuan Cong

Source: *Volume 16, No. 3, March 2016, Pages 789-800 doi: 10.4209/aaqr.2015.05.0307*

Between November 2005 and November 2007, weekly total suspended particle samples were collected at the Nam Co station in the inland Tibetan Plateau (TP). Through inductively coupled plasma mass spectrometry, twenty-nine elements were analyzed and their sources and fluxes were investigated. Mean elemental concentrations were lower than those at the edge of the TP. Some elements, such as Cr, Ni, Cd, and Pb, exhibited high enrichment factors (Cr: 22; Ni: 17; Cd: 23; and Pb: 9), indicating possible anthropogenic influence in this remote region, particularly during the pre-monsoon and monsoon seasons. In addition, an empirical orthogonal function analysis revealed the dominance of crustal-origin elements, rather than anthropogenic elements, in the aerosol. Furthermore, backward air mass trajectories demonstrated that the Nam Co region was mainly influenced by air masses from Central and South Asia. Accordingly, because of dust storms from

Central Asia and within the TP, crustal element concentrations, such as of Al, were higher during winter and pre-monsoon seasons than during the monsoon season. By contrast, anthropogenic elements, such as Cr and Cd, were relatively higher during the pre-monsoon and monsoon seasons because of pollutants transported from South Asia, where atmospheric brown clouds are concentrated and biomass combustion is prevalent. Dry deposition of aerosols dominated in the Nam Co region, particularly during the non-monsoon period, which is useful to interpret the elemental records in the TP ice cores and lake sediments.

Keywords: Elements; Aerosol; Atmospheric transport; Deposition flux; Nam Co.

Seasonal and Diurnal Variation of Formaldehyde and its Meteorological Drivers at the GAW Site Zugspitze

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Source: *Volume 16, No. 3, March 2016, Pages 801-815 doi: 10.4209/aaqr.2015.05.0334*

Continuous formaldehyde measurements were performed at the high-altitude GAW site Environmental Research Station Schneefernerhaus for more than one year. This unique dataset was analyzed for daily and seasonal variation and for the influence of large-scale synoptic conditions and air-mass origin on the observed concentrations. The average daily course exhibited maxima in the afternoon and minima at night, however differing between seasons. The general strong seasonal variation with average values for winter, spring, summer, and fall of 0.350, 0.529, 0.986, 0.429 ppbv, respectively, could be well explained by secondary production following photochemical activity. The large variability of formaldehyde mixing ratios within the seasons was shown to be influenced by different factors in this complex topography such as mixing of air masses from the planetary boundary layer and the free troposphere, advection of differently aged air from various source regions, and local meteorological conditions. An analysis of the impact of large-scale weather types, cyclonality, and flow directions revealed that the cleanest air masses were advected from westerly directions in particular under cyclonic conditions while southerly cyclonic and northerly/northwesterly anticyclonic conditions led to the highest formaldehyde levels.

Keywords: Formaldehyde; Remote site; Trajectories; Large-scale weather types.

Aerosol Measurements at South Pole: Climatology and Impact of Local Contamination

Patrick Sheridan, Elisabeth Andrews, Lauren Schmeisser, Brian Vasel, John Ogren

Source: *Volume 16, No. 3, March 2016, Pages 855-872 doi: 10.4209/aaqr.2015.05.0358*

The Atmospheric Research Observatory (ARO), part of the National Science Foundation's (NSF's) Amundsen-Scott South Pole Station, is located at one of the cleanest and most remote sites on earth. NOAA has been making atmospheric baseline measurements at South Pole since the mid-1970's. The pristine conditions and high elevation make the South Pole a desirable location for many types of research projects and since the early 2000's there have been multiple construction projects to accommodate both a major station renovation and additional research activities and their personnel. The larger population and increased human activity at the station, located in such close proximity to the global baseline measurements conducted at the ARO, calls into question the potential effects of local contamination of the long-term background measurements. In this work, the long-term wind and aerosol climatologies were updated and analyzed for trends. Winds blow toward the ARO from the Clean Air Sector ~88% of the time and while there is some year-to-year variability in this number, the long-term wind speed and direction measurements at South Pole have not changed appreciably in the last 35 years. Several human activity markers including station population, aircraft flights and fuel usage were used as surrogates for local aerosol emissions; peak human activity (and thus likely local emissions) occurred in the 2006 and 2007 austral summer seasons. The long-term aerosol measurements at ARO do not peak during these seasons, suggesting that the quality control procedures in place to identify and exclude continuous sources of local contamination are working and that the NSF's sector management plan for the Clean Air Sector is effective. No significant trends over time were observed in particle number concentration, aerosol light scattering coefficient, or any aerosol parameter except scattering Ångström exponent, which showed a drop of ~0.02 yr−1 over the 36-year record. The effect of discrete local contamination events in the Clean Air Sector is discussed using one well-documented example.

Keywords: Aerosol monitoring; Clean Air Sector; Wind sector screening.

Atmospheric Chemistry Measurements at Whiteface Mountain, NY: Ozone and Reactive Trace Gases

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Source: *Volume 16, No. 3, March 2016, Pages 873-884 doi: 10.4209/aaqr.2015.05.0376*

Measurements of ozone and reactive trace gases spanning four decades at the Whiteface Mountain summit observatory are presented. Ozone (O3) measurements began in the mid-1970's, and acid rain and O3 precursor gas measurements became routine in the late 1980's and early 1990's. Measurements at the lower altitude lodge level have also been performed routinely since about 2000. The 40-year O3record shows up and down fluctuations through the 1980's, a relatively stable period into the early 2000's, and indications of a decreasing trend over the past ten years. Sulfur dioxide (SO2) and carbon monoxide (CO) trends are clearly decreasing over the roughly 25-year period of measurements at the summit observatory. Oxides of nitrogen (NOy and NO2) show rather more complicated trends, increasing to a maximum in the mid-2000's, and decreasing sharply until 2011 with slight increases in concentration since then. Wind rose analysis shows the greatest contribution to high concentrations of precursor gases are from the west, southwest, and southern sectors, with SO2 and oxides of nitrogen having the most sharply defined high pollution sectors. Seasonal variations of trace gas concentrations at the summit and lodge levels are also examined. Ozone concentrations are highest in the spring months at both locations, and higher at the summit than the lodge. In contrast precursor gases (SO2 and NOx) show highest concentrations in winter months with the lodge consistently higher than the summit.

Keywords: Air quality; Air pollution; Trend analysis; Seasonal variation; Pollution roses.

Nature and Sources of Ionic Species in Precipitation across the Indo-Gangetic Plains, India

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Source: *Volume 16, No. 4, April 2016, Pages 943-957 doi: 10.4209/aaqr.2015.06.0423*

The spatial distribution of rainwater chemistry over the densely-populated and highly polluted Indo-Gangetic Plains (IGP) was investigated using samples (total = 687) collected during three consecutive summer monsoon seasons from 2009 to 2011. The concentrations of secondary ionic species (SO_4^{2-} and NO_3^-) were measured along with the other major ions (F^-, Cl^-, Na^+, K^+, Ca^{2+}, Mg^{2+} and NH_4^+) and pH and specific conductivity. The weighted mean pH (± std) and conductivity of rainwater were 5.73 (± 0.17) and 31.6 (± 31.0) μS cm^{-1}, respectively. Approximately 16% of rainwater samples were acidic (pH < 5.61) with a mean pH = 5.38 of acid rain and rest of them were more alkaline (pH > 5.61) (mean pH = 6.34 for the more basic samples). Specific conductivity was ~39% lower (20.6 μS cm^{-1}) for the acidic rain compared to the more basic (33.6 μS cm^{-1}) samples. The mean sum of all of the measured ions is 351.6 ± 130.1 μeq L^{-1} with the highest contributions being Ca^{2+} (30%) and SO_4^{2-} (15%). Mean [SO_4^{2-}] (52 μeq L^{-1}) and [NO_3^-] (29 μeq L^{-1}) were approximately five and ten times higher, respectively, compared to background hemispheric values.

Secondary ions had the highest deposition fluxes (SO_4^{2-}, 25.2 kg ha^{-1} y^{-1} and NO_3^-: 18.3 kg ha^{-1} y^{-1}). The mean ratio of $H^+/(NO_3^- + SO_4^{2-})$ was 0.02 indicating ~98% of the acidity was neutralized. Ca^{2+}, (57%), Mg^2 (25%), NH_4^+ (15%) and K^+ (4%) were important neutralizing species. Positive Matrix Factorization (PMF) was applied to the deposition fluxes. Five factors were identified and identified as ammonia neutralized, sea salt, soil, biomass burning, and calcium neutralized.

Keywords: Rainwater chemistry; Ion balance; Positive matrix factorization; Specific conductivity; Acid rain.

Improving the Representation of Cross-Boundary Transport of Anthropogenic Pollution in East Asia Using Radon-222

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Source: *Volume 16, No. 4, April 2016, Pages 958-976 doi: 10.4209/aaqr.2015.08.0522*

We report on 10 years of hourly atmospheric radon, CO, and SO_2 observations at Gosan Station, Korea. An improved radon detector was installed during this period and performance of the detectors is compared. A technique is developed whereby the distribution of radon concentrations from a fetch region can be used to select air masses that have consistently been in direct contact with land-based emissions, and have been least diluted *en route* to the measurement site. Hourly radon concentrations are used to demonstrate and characterise contamination of remote-fetch pollution observations by local emissions at this key WMO GAW site, and a seasonally-varying 5-hour diurnal sampling window is proposed for days on which diurnal cycles are evident to minimise these effects. The seasonal variability in mixing depth and "background" pollutant concentrations are characterised. Based on a subset of observations most representative of the important regional fetch areas for this site, and least affected by local emissions, seasonal estimates of CO and SO_2 in air masses originating from South China, North China, Korea and Japan are compared across the decade of observations.

Keywords: ^{222}Rn; Fetch analysis; Local effects; Model benchmarking; Sampling window.

Satellite and Ground Observations of Severe Air Pollution Episodes in the winter of 2013 in Beijing, China

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Source: *Volume 16, No. 4, April 2016, Pages 977-989 doi: 10.4209/aaqr.2015.01.0057*

Beginning in early January 2013, Beijing experienced multiple prolonged and severe smog events that were characterized by very high levels of $PM_{2.5}$, with peak daily $PM_{2.5}$ over 400 μg m^{-3}. With $PM_{2.5}$ concentration contours created from ground observations and satellite remote sensing data, we describe the spatial and temporal characteristics of these episodes and further investigated the factors that contributed to these episodes. Our results indicated that these smog episodes affected a much larger geographic region, far beyond Beijing metropolitan area, corresponding to a total area of ~550,000 km^2 and ~180 million people. The extremely cold weather in December 2012 and regional pollution transport were likely the main causes of these severe PM pollutions. In addition to aggressive emission control measures for Beijing, coordinated regional policy must be put in place to achieve more blue-sky days. Although the configuration of the current ground monitoring network may be sufficient to record $PM_{2.5}$ levels in urban centers, these monitors alone cannot fully characterize the spatial pattern and track the transport of air pollution on a regional scale. Satellite remote sensing data can provide valuable information to fill the gaps left by ground monitors to create a more comprehensive picture of $PM_{2.5}$.

Keywords: Smog; MODIS; Aerosol optical depth; $PM_{2.5}$; HYSPLIT.

Seasonal Variation of Selected Metals in Particulate Matter at an Industrial City Kota, India

Manju Meena, Bharat Singh Meena, Uttra Chandrawat, Ashu Rani

Source: *Volume 16, No. 4, April 2016, Pages 990-999 doi: 10.4209/aaqr.2015.02.0074*

This study investigates seasonal variation in concentration of some heavy metals in suspended and respirable particulate matter (SPM and RPM) collected from five different zones situated in Kota city during both summer (March, April, May and October) and winter (January, February, November and December) seasons of 2011–2012. Mean concentrations of anthropogenic origin metals (Pb, Zn, Cu and Cd) were higher in winter and lower in summer with their relative abundance in order: Zn > Pb > Cu > Cd while reverse trend was observed for crustal origin metals (Ca, Mg and Fe) at all zones. Meteorological conditions such as temperature, relative humidity, wind velocity and wind direction during winter and summer were found affecting the metals concentration trends in different seasons. Wind roses indicate that the zones lying in predominant

North-east wind direction from point source (KSTPS) in winter (25.74%) and summer (15.31%) faced higher metal burden following zone 1, which is suffering most owing to its closest location to the source. Statistical analysis by Pearson's correlations, enrichment factor and principal component analysis indicates that coal based Thermal Power Plant is the major source of heavy metals besides other industrial activities in the study area. It is to be noted that because of higher residence time, significant concentration of Pb is found at all the zones in the city which, probably, has its origin in earlier vehicular exhaust as well.

Keywords: Seasonal variation; Suspended and respirable particulate matter; Pearson's correlations; Enrichment factor; Principal component analysis.

Profile of Atmospheric PAHs in Rawalpindi, Lahore and Gujranwala Districts of Punjab Province (Pakistan)

Atif Kamal, Jabir Hussain Syed, Jun Li, Gan Zhang, Adeel Mahmood, Riffat Naseem Malik

Source: *Volume 16, No. 4, April 2016, Pages 1010-1021 doi: 10.4209/aaqr.2015.01.0016*

In this study, polyurethane foam passive air samplers (PUF-PAS) were deployed to evaluate the atmospheric concentration levels and ambient exposure of polycyclic aromatic hydrocarbons (PAHs) in Gujranwala, Lahore and Rawalpindi districts of the Punjab Province (Pakistan). The PAHs were extracted from the PUFs disks using Soxhlet extraction assembly, and were further concentrated using rotary evaporator, purified on a column, packed with alumina/silica, and eluted with a solution of dichloromethane:hexane (1:1 v:v). The PAHs quantification was carried out gas-chromatograph equipped with a mass-spectrometer (GC-MS). Regression scatter plots and molecular diagnostic ratios were used to identify and characterize the emission of PAH species from different sources. Among all detected PAHs, a high concentration of naphthalene (Naph) was observed in Lahore (327 pg m^{-3}) and Rawalpindi (316 pg m^{-3}) cities followed by phenanthrene, banzo(a)pyrene, pyrene, benzo(b)florenthene and benzo(k)flourenthene. Our findings revealed that the low molecular weight (LM)-PAHs in Rawalpindi and Gujranwala cities could have possibly originated from a local petroleum refinery and vehicular emissions respectively, whereas the high molecular weight (HM)-PAHs observed in Wazirabad, could be largely related to both biomass and traffic emissions. Results also showed that ~88 percent of the atmospheric PAHs could be attributed to the wood combustions (R^2 = 0.88), out of which more than 50 percent of wood combustion were possibly with the brick kiln sector (R^2 = 0.53).

Keywords: Environmental pollution; ILCR; Exposure; Pakistan; PAHs

Investigating the Health Effects of Exposure to Criteria Pollutants Using AirQ2.2.3 in Shiraz, Iran

Aezam Mohammad, Abooalfazl Azhdarpoor, Abbas Shahsavani, Hamidreza Tabatabaee

Source: *Volume 16, No. 4, April 2016, Pages 1035-1043 doi: 10.4209/aaqr.2015.07.0434*

The quality of air is one of the main environmental issues related to human health. The aim of this study was to evaluate the effects of air pollution on the health of residents in Shiraz, as one of the major cities in Southern Iran, with population of 1500000 people. In this study, AirQ2.2.3 model developed by the WHO European Centre for Environment and Health was used. Daily concentration of particulate matter less than 10 microns in diameter (PM_{10}), sulfur dioxide, nitrogen dioxide and maximum 8-hour average ozone concentration were used to evaluate the health effects of human exposure to these pollutants. The total number of excess deaths, Cardiovascular Disease (CVD) mortality, mortality from respiratory diseases, hospital admissions for CVD and hospital admissions for respiratory disease (RD) were calculated. In 2012 and 2013, hospital admissions for respiratory disease for the WHO baseline incidence of PM10 were respectively 54.6% and 38.6% of the total hospital admissions for respiratory disease. That was the highest short-term health effects on 1500000 Shirazi residents. The assessments carried out indicated the possibility that CVD mortality can play a major role in mortality due to PM_{10}, SO_2 and O_3 pollutants. Overall, the results showed that health effects resulting from exposure to pollutants are directly related to their concentration. Therefore, immediate action to prevent pollution and reduce emissions from various sources, such as transport and energy production industries, is required to reduce the concentration of pollutants in Shiraz.

Keywords: AirQ; Particulate matter; Sulfur dioxide; Nitrogen dioxide; Ozone; Mortality.

Identification of Long-Range Transport Pathways and Potential Sources of PM10 in Tibetan Plateau Uplift Area: Case Study of Xining, China in 2014

Yujie Xin, Guochen Wang, Li Chen

Source: *Volume 16, No. 4, April 2016, Pages 1044-1054 doi: 10.4209/aaqr.2015.05.0296*

The aim of the study is to identify long-range transport pathways that may have an important influence on PM10 levels in plateau uplift area, namely Xining in northwestern China. Cluster

analysis was applied to identify the main trajectory groups in horizontal direction and 3D cluster analysis was employed to identify the origins and distributions of major trajectory groups in vertical direction. Potential Source Contribution Function (PSCF) and Concentration-weighted Trajectory (CWT) were applied to identify the major potential source areas (PSA). Based on the temporal and spatial distribution of backward trajectories, four major trajectory pathways were clustered. The results indicated that Xining was easily affected by inland trajectories in four seasons but there were obvious results that different trajectories have dissimilar influences on the mean PM_{10} concentrations. In horizontal direction, the long-range transport pathways were obvious in spring and winter while a few of long-range transport pathways could be found in summer and autumn. Because wind mainly came from north or west in spring and winter and it was very strong, which had a big influence on the transportation of transport pathways while wind in summer and autumn had a small impact on the transportation of transport pathways. In vertical direction, in the 700 hPa barometric altitude (3000 m) above, air masses in winter and spring with long transport pathways were the most important back-trajectories which had a great influence on Xining city. In summer and autumn, Xining was mainly influenced by airflow distributed below 700 hPa barometric altitude. In spring and winter, eastern Xinjiang, border areas between Gansu and Inner Mongolia and southern Tibet in China with the highest Weight Potential Source Contribution Function (WPSCF) and Weight Concentration-weighted Trajectory (WCWT) values were the dominant potential sources, which demonstrated the contribution from sources outside of Xining were significant. In summer and autumn, WPSCF values outside of Xining were no more than 0.5 (most of them were less than 0.3) and WCWT values were almost lower than 100 μg m^{-3} in those two seasons, which suggested that there were no main important PSA in those two seasons. Furthermore, the study also revealed that Tibet in China was one of the potential sources of PM_{10} in Xining.

Keywords: HYSPLIT; Cluster analysis; PSCF; CWT; PM_{10}; Xining.

An Improved Method for Monitoring Fine Particulate Matter Mass Concentrations via Satellite Remote Sensing

Hamed Karimian, Qi Li, Chengcai Li, Lingyan Jin, Junxiang Fan, Ying Li

Source: *Volume 16, No. 4, April 2016, Pages 1081-1092 doi: 10.4209/aaqr.2015.06.0424*

Ground level monitoring of Particulate Matter (PM) is limited by spatial coverage and resolution, in spite of possessing high temporal resolution and accuracy. Atmospheric Aerosol Optical Depth (AOD), a product of space-borne remote sensing, has shown significant potential for estimating ground level PM concentrations. Several approaches have been used to improve the correlation between AOD-PM by providing corrections for the aerosol vertical profile and ground level

humidity. However, the effects of the vertical profile of humidity and aerosol size on the AOD-PM relationship requires further study. In this paper, we propose a method for developing an AOD-$PM_{2.5}$ relationship by retrieving the vertical profile of relative humidity via ground observation data and aerosol size distribution in Beijing. Moreover, a series of Hanel growth coefficients (γ) are applied to determine the specific value, which maximizes the correlation. The results show that applying our proposed method can improve the correlation from R = 0.610 to R = 0.707 for Terra and R = 0.707 to 0.752 for Aqua. The best correlations were obtained for γ = 1.2 and 1.3 for Terra and Aqua, respectively. A good correlation (R = 0.8) between ground based and MODIS based $PM_{2.5}$ measurements, together with employing MODIS to predict true air pollution levels (65% accuracy), suggests that the vertical profile of RH derived via ground level observation and aerosol size should be considered and applied to models in future studies, which utilize satellite data for air pollution monitoring and controlling.

Keywords: Vertical relative humidity; Hygroscopic effect; $PM_{2.5}$; Aerosol Optical Depth; Boundary layer.

Airborne Measurements of High Pollutant Concentration Events in the Free Troposphere over the West Coast of South Korea between 1997 and 2011

Greem Lee, Hye-Ryun Oh, Chang-Hoi Ho, Jinwon Kim, Chang-Keun Song, Lim-Seok Chang, Jae-Bum Lee, Seungmin Lee

Source: *Volume 16, No. 5, May 2016, Pages 1118-1130 doi: 10.4209/aaqr.2015.06.0407*

Aircrafts enable the direct measurement of chemical components in the free troposphere (FT). This study employed airborne measurements to examine the occurrences of high concentrations of SO2 and NOx in the FT over the coastal region west of the Seoul metropolitan area, South Korea. The data from a long-term (1997–2011) airborne measurement campaign were used to determine the meteorological conditions favorable for carrying these pollutants into the Seoul area. The back trajectory analyses of 21 instances of high FT pollutant concentration events showed ascending patterns from the major pollutant sources, mainly the industrial complexes in eastern China, in 9 instances and passing patterns in 12 instances. In the ascending instances, developing low-pressure systems over the source regions provide favorable conditions to uplift air pollutants from the surface into the FT. In the passing instances, an anomalous low-pressure system near the surface prevented airflows from descending into the boundary layer and upper-level anticyclonic systems helped to keep the ascending airflows in the FT. This study proposes the basic mechanisms for

predicting air quality in the Seoul area, considering that air pollutants in the FT often entrain into the boundary layer to increase local concentrations.

Keywords: Seoul; Aircraft measurement; Free troposphere; SO_2; NO_x.

What Causes Observed Fog Trends: Air Quality or Climate Change?

Otto Klemm, Neng-Huei Lin

Source: *Volume 16, No. 5, May 2016, Pages 1131-1142 doi: 10.4209/aaqr.2015.05.0353*

Fog is a situation when the visual range, which is the horizontal visibility, is reduced to less than 1000 m near the Earth's surface by the presence of cloud droplets. Fog trend analyses are reported in the literature for hundreds of stations worldwide, the majority of which showing a considerable reduction of fog. Although fog is often associated with conditions at which cloud condensation nuclei had been activated at rH (relative humidity) > 100% and rapid growth had lead to the formation of fog droplets, this study focusses on urban air masses and conditions when rH is just below 100%. Mie scattering analysis shows that fog can form under such conditions and the reduction of the visual range is mainly caused by submicron aerosol particles which grow to diameters around 1 μm through deliquescence. The liquid water content itself is poorly correlated with the visual range. Assuming equilibrium conditions, both an increase of the air temperature and a reduction of the aerosol particle concentration lead to reductions of fog. In our example case, the increment for a temperature increase by 0.1°C had about the same effect as the reduction of aerosol concentrations by 10%. Care must be taken in projecting this result to actual conditions because the system is non-linear. However, physical evidence is presented which confirms that both climate change and an improvement of air quality are mechanisms that can contribute to the reduction of fog.

Keywords: Fog trends; Air quality; Air pollution; Visibility; Visual range.

Pollution Characteristic of Atmospheric Carbonyls during One Haze Event in Nanning, South China

Song-Jun Guo, Mei Chen

Source: *Volume 16, No. 5, May 2016, Pages 1143-1151 doi: 10.4209/aaqr.2015.01.0047*

This study was the first to investigate pollution characteristics of atmospheric carbonyls in one haze event (July 17–22, 2012) in Nanning, south China. It was found that (1) acetaldehyde (29.74 ± 6.06 μg m^{-3}), formaldehyde (11.20 ± 1.72 μg m^{-3}), and acetone (9.18 ± 11.22 μg m^{-3}) were the most abundant in haze days, (2) concentrations and O_3 formation potentials of ambient carbonyls in haze days were significantly higher than those on normal days, and (3) visibility and wind speed in haze days were lower than those on normal days, indicating that haze days represented favorable pollution conditions for carbonyls. Diurnal variations of ambient carbonyls in haze days showed a pattern of two peaks occurring in two traffic rush-hour periods due to positive traffic emissions. Average concentration ratio of formaldehyde/acetaldehyde (C_1/C_2) in haze days (0.39 ± 0.10) was slightly lower than that (0.87 ± 0.23) on normal days, and the ratios in two traffic rush-hour periods were close to those in non rush-hour periods, likely implying that traffic emissions might not be a major source for ambient carbonyls. Correlation among formaldehyde, acetaldehyde, acetone, and total carbonyls was good (R^2 = 0.49–0.85) in haze days and excellent (R^2 = 0.80–0.98) on normal days, indicating that the sources of ambient carbonyls in haze days were more complex compared to normal days.

Keywords: Pollution characteristic; Haze event; Carbonyls; Nanning

Continuous Measurement of Ambient Aerosol Liquid Water Content in Beijing

Oscar A. Fajardo,Jingkun Jiang, Jiming Hao

Source: *Volume 16, No. 5, May 2016, Pages 1152-1164 doi: 10.4209/aaqr.2015.10.0579*

The amount of liquid water present on ambient aerosol particles influences its radiative properties, the partitioning of gas-phase organic and inorganic compounds to the condensed phase, and the assessment of particle health effects upon inhalation. An improved home-made Dry-Ambient Aerosol Size Spectrometer (DAASS) was implemented and deployed at a sampling site in Beijing to measure water content and volumetric growth factor of fine ambient aerosol during clean and polluted days. Aerosol chemical composition was characterized by an aerosol chemical speciation monitor (ACSM). Meteorological conditions were also assessed to trace origin of the air masses arriving at the site. A thermodynamic model, ISORROPIA II, was used to predict aerosol water content based on inorganic compositions measured by ACSM. It was then compared with the aerosol water content measured by DAASS. During the two-week sampling period, an aerosol efflorescence behavior was observed around 50% relative humidity (RH). Aerosol at the sample location was predominantly alkaline, with an average acidity ratio of 1.3 during the polluted days but very variable during the clean days. This fact coupled with generally low ambient RH resulted in

low amounts of liquid water detected during the campaign. When RH reached its maximum of 87% and aerosol appeared to be more acidic, the maximum amount of aerosol water content measured was 1.3 μg m^{-3} (44.9% wt of DAASS measured ambient aerosol). Measured aerosol water content and model predictions seem to show a better agreement during the more polluted days, but during the clean days aerosol organic fraction appears to have an important contribution in the water content. We argue that the organic-associated water is comparable with that associated with inorganic compositions during the clean periods at low ambient RH conditions.

Keywords: Urban aerosol; Aerosol water content; Beijing pollution; Aerosol acidity.

Seasonal Variations in Water Soluble Inorganic Ions, OC and EC in PM_{10} and $PM_{>10}$Aerosols over Delhi: Influence of Sources and Meteorological Factors

Pawan Kumar, Sudesh Yadav

Source: *Volume 16, No. 5, May 2016, Pages 1165-1178 doi: 10.4209/aaqr.2015.07.0472*

The PM_{10} and particles of greater than 10 μm aerodynamic diameter (hence forth referred as $PM_{>10}$) collected over Delhi exceeded the National Ambient Air Quality Standards (100 μg m^{-3}) with an annual average of 215 μg m^{-3} and 495 μg m^{-3}, respectively. The water soluble inorganic ions (WSII) were higher throughout the year in PM_{10} (13.5%) than in $PM_{>10}$ (5%). SO_4^{2-} dominated over NO_3^- ions and contributed 54% to the total WSII in PM_{10}. NO_3^- and Ca^{2+} were dominant ions in $PM_{>10}$. NH_4^+ during winter and autumn, and Ca^{2+} in summer PM_{10}samples were the major acid neutralizing species. The WSII showed monthly and seasonal changes. The total carbon (TC = EC + OC) constituted 8.8%–47.8% of total PM_{10} mass, and OC and EC varied from 9.91 to 37.06 μg m^{-3} and 5.42 to 22.23 μg m^{-3} during the year long sampling period. The SOC contributed more to OC in summers and the char (EC1) dominated over soot (EC2 + EC3) in EC fractions throughout the year. In summer samples, secondary OC (SOC) contributed 81% to OC and the OC/EC ratio was 3.61 suggesting the possible role of mineral dust and high photochemical activity in SOC production. For the eight different fractions, three dominant sources were identified, coal combustion, biomass burning, and motor vehicle exhaust for OC1, OC2 and OC3; vehicle exhaust for OC4, OP, EC2 and EC3; and biomass and coal combustion for EC1. The seasonal changes in the WSII and carbon concentrations in PM_{10} and $PM_{>10}$ are attributed to both the sources and the meteorological conditions in and around the study area.

Keywords: Soluble ions; Carbon fractions; Processes; Air quality.

Source Apportionment of Fine and Coarse Particulate Matter in Industrial Areas of Kaduna, Northern Nigeria

Sunday A. Orogade, Kayode O. Owoade, Philip K. Hopke, Donatus B. Adie, Abubakar Ismail, Charles A. Okuofu

Source: *Volume 16, No. 5, May 2016, Pages 1179-1190 doi: 10.4209/aaqr.2015.11.0636*

This study was conducted to investigate the sources of fine and coarse airborne particulate matter in an urban environment in Nigeria. A total of 278 samples were collected over a twelve-month period from two industrial areas (Kudenda agricultural processing AP and Refinery industries NNPC) in Kaduna, Northern Nigeria for $PM_{2.5}$ and $PM_{2.5–10}$ on nuclepore polycarbonate filters using a Gent sampler. Elemental concentrations and black carbon analyses were performed using X-Ray fluorescence (XRF) and optical transmissometry respectively. The annual average concentrations for $PM_{2.5}$ at each site (Kudenda and NNPC) were 135.7 μg m^{-3} and 37.2 μg m^{-3} and for $PM_{2.5–10}$, concentrations were 269.2 μg m^{-3} and 97.4 μg m^{-3}, respectively. These values exceeded the Nigerian Annual National Ambient Air Quality Standard (NAAQS) of 15 μg m^{-3} for $PM_{2.5}$ and 60 μg m^{-3} for PM_{10}. Positive matrix factorization (PMF) was used to identify sources and quantify their contribution to pollutants at the sampling sites in one of the most industrialized cities in Nigeria. Four sources were resolved for both $PM_{2.5}$ and $PM_{2.5–10}$ and were identified as: Residual oil 49% (17.16 ± 0.04 μg m^{-3}), Soil 29% (10.36 ± 0.26 μg m^{-3}), Continental dust 18% (6.20 ± 0.18 μg m^{-3}), and Motor vehicles emissions 4% (1.56 ± 0.02 μg m^{-3}) for $PM_{2.5}$ while that for $PM_{2.5–10}$ were Soil 50% (27.37 ± 1.03 μg m^{-3}), Continental dust 21% (11.55 ± 0.26 μg m^{-3}), Vehicular emissions 18% (9.87 ± 0.03 μg m^{-3}), and Petrochemical 11% (6.23 ± 0.02 μg m^{-3}). About 82% and 79% were attributed to anthropogenic sources for both fine and coarse samples, respectively. Continental dust was associated with northwesterly and northerly regional transport. Residual oil combustion was the predominant fine PM source and was attributed to fuel oil combustion for power generation and process energy within the local industrial areas. Although transported continental dust is an important source, the majority of the airborne PM in this industrial area was the result of local emissions.

Keywords: $PM_{2.5}$; $PM_{2.5–10}$; XRF; Black carbon; Source apportionment; Nigeria.

Application of WRF-Chem Model to Simulate PM_{10} Concentration over Bogota

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Source: *Volume 16, No. 5, May 2016, Pages 1206-1221 doi: 10.4209/aaqr.2015.05.0318*

The online meteorological and chemical transport Weather Research and Forecasting with Chemistry (WRF-Chem) model is implemented over Bogota and validated against ground-based observations for meteorological variables and PM_{10} concentrations. The simulated average temperature shows a very small positive bias. The relative humidity and wind speed are also overpredicted for the selected simulations. The 24-h average PM_{10} concentration is underpredicted compared to the overall ground observations. Overall, the present case study shows that the WRF-Chem model has an acceptable performance for meteorological variables as well as PM_{10} concentration over Bogota. This study provides a general overview of WRF-Chem simulations and can serve as a reference for future air quality modelling exercises for PM_{10} over Bogota.

Keywords: WRF-Chem; PM_{10}; Air quality modeling; Bogota.

Emissions Study and Estimation of Carbon Dioxide Production from Jatropha Curcas Oil Biodiesel

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The goal of this study was to analyse the combustion characteristics and emissions of Jatropha curcas biodiesel (JCB) when run in a diesel engine. Jatropha curcas oil was used to produce Jatropha curcas biodiesel (JCB) through a transesterification process. The major fuel properties of JCB, including the acid value, kinematic viscosity, flash point, gross heating value, and iodine value, were determined and compared with that of soybean biodiesel (SBM), sunflower seed biodiesel (SFM), mackerel fish oil biodiesel (MB), and premium diesel (D). JCB had a higher density, acid value, kinematic viscosity, iodine value and flash point, but a lower gross heating value, than D. JCB was then used to analyze combustion characteristics, CO, CO_2, NO, NO_x, SO_2, and particulate matter (PM), under varied engine speeds and varied engine loads. The experimental results show CO_2 concentration increased with increasing engine loads for all fuels. Engine trials on D exhibited better combustion efficiency at lower engine loads (0 kW–4 kW) but engine trials on JCB exhibited better combustion efficiency for higher engine loads (5 kW– 8 kW). JCB emitted more NO and NO_x on a loaded engine. Engine trials on JCB emitted higher PM concentration when the engine was

not loaded, while engine trials on MB produced higher PM concentration when the engine was loaded. The estimated CO_2 emissions for JCB, MB, and D are 9221.3, 9617.2, and 10185.0 g (gal fuel)$^{-1}$, respectively.

Keywords: Jatropha curcas biodiesel; Biodiesel emisssions; CO_2 emission.

Source Apportionment of Urban Background Particulate Matter in Birmingham, United Kingdom Using a Mass Closure Model

Adewale M. Taiwo

Source: *Volume 16, No. 5, May 2016, Pages 1244-1252 doi: 10.4209/aaqr.2015.09.0537*

Particulate matter (PM) collected during the summer period of 2011 at the urban background of Elms Road Observatory Site (EROS) in Birmingham, United Kingdom was studied and apportioned using the mass closure model. Particulate matter samples were analysed for Cu, Zn, Fe, Ni, Mn, Cl^-, NO_3^-, SO_4^{2-}, PO_4^{3-}, $C_2O_4^{2-}$, Na^+, NH_4^+, K^+, Ca^{2+}, OC and EC using the standard procedures. Results showed mean mass concentrations of 5.42 ± 1.7 μg m^{-3} for $PM_{2.5}$ and 5.41 ± 0.27 μg m^{-3} for $PM_{10-2.5}$. Organic carbon (OC, 26%) formed the major component of $PM_{2.5}$ followed by sulphate (25%) and ammonium (12%). In the coarse PM fraction, ammonium, chloride and OC constituted 17, 15 and 14%, respectively of mass concentration. Metal concentrations in both PM fractions were less than 2% of the observed mass. The mass closure model applied to apportion $PM_{2.5}$ and $PM_{10-2.5}$ chemical species was able to identify four components namely: carbonaceous (45 and 19%, respectively), sea salt (7 and 25%), secondary aerosol (48 and 13%) and minerals (13 and 12%). The mass closure model was unable to explain all the chemical components in the coarse PM category leaving the unidentified mass as 31%. On the other hand, the model overestimated the mass of $PM_{2.5}$ by 14%. The study showed secondary aerosol and carbonaceous species as the dominating sources of PM pollution in the study area.

Keywords: Particulate matter; Urban background; Mass closure model; Emission sources.

Impact of Grid Resolution on Aerosol Predictions: A Case Study over Italy

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Source: *Volume 16, No. 5, May 2016, Pages 1253-1267 doi: 10.4209/aaqr.2015.02.0058*

This study investigates the effect of grid resolution on the particulate matter (PM_{10}, $PM_{2.5}$) mass concentrations and its chemical composition simulated with the AMS-MINNI modelling system. The air pollution was simulated over Italy with grid resolutions of 20 and 4 km, for a whole year. The gridded emissions were produced performing speciation and space-time disaggregation of aggregated inventory data, using both land use information and anthropogenic activity-based profiles. Often, the fine grid simulations, based on high resolution gridded emissions, improved the agreement between model and measurements. In particular, the use of a fine grid improved predictions of primary species such as elemental carbon (EC), PM_{10} and $PM_{2.5}$ mainly at urban stations. An improvement of predicted PM components and mass concentration at high altitudes sites was also observed, especially during winter. However, a general overestimation of nitrate (NO_3^-) and of secondary inorganic species, more evident at night than during the day, was increased by employing a finer grid. Organic carbon (OC) was more affected by the grid resolution than the other species. At urban and kerbside stations, the use of a finer grid resulted in an overestimation of primary organic carbon aerosol (POC) but had a negligible effect on secondary organic carbon aerosol (SOC). The overestimation of carbonaceous aerosol (defined as the sum of EC, POC and SOC), at an urban station, opposite to general underestimation of this component by air quality (AQ) models, indicates that the anthropogenic emissions can contribute as much as organic model formulation at the success of simulation in reproducing experimental data.

The modelling results obtained under stable meteorological conditions characterised by weak winds, which are often encountered in the Po Valley, did not improve substantially by the increase of the modelling system resolution.

Keywords: Horizontal grid resolution; Aerosol chemical composition; PM_{10}; $PM_{2.5}$; Air quality modelling.

Satellite-Based Estimates of Aerosol Washout and Recovery over India during Monsoon

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Source: *Volume 16, No. 5, May 2016, Pages 1302-1314 doi: 10.4209/aaqr.2015.01.0018*

Large aerosol optical depth (AOD) observed over the Indian subcontinent during the monsoon season in the satellite data challenges the common notion of aerosol washout by monsoon rain. Here, we examined recovery of aerosol field after washout by monsoon rain over various rainfall homogeneous zones of India in view of the duration of rainfall, recovery time and source strength. Mean (± 1 standard deviation) seasonal aerosol optical depth, AOD is highest over the central northeast 1 (0.74 ± 0.22) followed by central northeast 2 (0.60 ± 0.11), northwest (0.61 ± 0.15), west-central (0.54 ± 0.13), northeast (0.29 ± 0.08), peninsular India (0.39 ± 0.07) and hilly region (0.33 ± 0.08) in the monsoon season. Post-washout aerosol recovery in India is not a linear function to the recovery period relative to the two successive satellite overpasses. Fastest recovery is observed in the central northeast region dominated by anthropogenic emission. In general, washout is more for 9-hour spell than 3-hour spell, but not spatially uniform over the various rainfall homogeneous zones. In central northeast region it is observed that updraft plays an important role in post precipitation aerosol build up whereas in dust-dominated northwest India, monsoon rainfall (whenever occurs) suppresses dust emission because of the increased soil moisture and therefore inhibits the recovery. The number of grids where washout outweighs recovery during the monsoon season for a 3-hour rainfall increases by 5.6% with an increase in rain rate from < 2 mm day^{-1} to > 4 mm day^{-1}, while the corresponding increase for a 9-hour rainfall event is 2.8%. AOD reduces in 'cloudy-sky' condition relative to 'clear-sky' condition because aerosols are scavenged by cloud drops as the clouds grow vertically during the monsoon. Quantitatively, AOD decreases by 16% per 100 hPa increase in cloud base height.

Keywords: AOD; Precipitation; Recovery time; Indian monsoon; Homogeneous zones.

Origin Identification of Carbonaceous Aerosol Particles by Carbon Isotope Ratio Analysis

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Source: *Volume 16, No. 6, June 2016, Pages 1356-1365 doi: 10.4209/aaqr.2015.07.0443*

We applied stable carbon isotope ratio ($^{13}C/^{12}C$) and radiocarbon (^{14}C) analysis for the quantification of three main aerosol sources (coal, biomass and liquid fossil fuel derived aerosol emissions). Submicron aerosol samples (PM_1) were collected from 27th October, 2014 to 19thJanuary, 2015 at a suburban site of Vilnius city (Lithuania). To determine fossil and non-fossil contributions to submicron carbonaceous aerosol particles, ^{14}C measurements of total carbon (TC) were performed using single stage accelerator mass spectrometer (SSAMS, NEC, USA). The concentrations of TC and $\delta^{13}C$ in PM_1 fraction were measured using elemental analyzer interfaced to isotope ratio mass spectrometer (EA-IRMS). The TC concentration during measurement period ranged from 1.3 to 9.6 μg m^{-3}. The variation of TC concentrations can be explained by the influence of long-range transport and dispersion properties of the boundary layer (mixed layer depth).We found that biomass-derived aerosol sources are prevailing in Vilnius during wintertime and ranged from 57% to 84% of total carbonaceous aerosol fraction. Applying isotope mass balance calculations the traffic emissions were estimated to be 15 ± 7% and coal combustion made up 14 ± 9% in PM_1. To provide better information about the pollution sources, the carbon isotope analysis along air mass transport pattern was performed. Our results demonstrated that the high contribution to PM_1 from coal burning (up to 40%) was observed for air masses transported from highly industrialized Western Europe regions. Combination of stable carbon isotope ratio with the radiocarbon data allow to distinguish coal from liquid fossil fuel in the aerosol particle emissions.

Keywords: Aerosol particles; Stable carbon isotopic ratio; Radiocarbon analysis.

Source Identification of Acid Rain Arising over Northeast China: Observed Evidence and Model Simulation

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Source: *Volume 16, No. 6, June 2016, Pages 1366-1377 doi: 10.4209/aaqr.2015.05.0294*

Acid rain has already been the environment issue for the past century and is still concerned in Southern and Eastern China. However, Northeastern China, considered in the past as non-acid rain area, is reported high frequency of acid rain occurred recently. Through a measurement and model simulation analysis with linear regression technique, factor speciation, backward trajectory, source-tracing and fraction sampling methods, this study aims to investigate the causes of the acid rain frequently occurred in the two Northeastern cities (i.e., Dandong and Dalian) and to identify the contributing sources of the chemical ions in precipitation. The annual averaged pH_{vwa} of 2007 ranged within 4.15–4.27 and 4.5–5.15 over Dandong and Dalian, which suggested the similar acidity with Southern China. The precipitation acidity in Dandong was found to be sulfur dominant in winter-spring ($nssSO_4^{2-}/NO_3^-$ = 13.1) than summer-autumn ($nssSO_4^{2-}/NO_3^-$ = 1.05), whereas in

Dalian, estimated ratio nssSO_4^{2-}/NO_3^- was about 2.35 all over the year, reflecting the strong impact of local NO_x emissions from a much more intense mobile traffic in comparison with Dandong. The findings also revealed a typical physico-chemical condition with constant strong influence of regional transport over Dandong and Dalian particularly in summer. Besides, analysis of washout and rainout mechanisms showed that the long range transport in-cloud (rainout) was more pronounced in Dandong (with minor local emissions) than Dalian (with more significant local emissions). This study indicates that acid rain pollution in Northeastern China requires much effort, not only in local emission abatement, but also in regional trans-boundary pollution control.

Keywords: Acid rain; Northeast China; Precipitation chemistry; Scavenge; Source identification.

Variation of Urban Atmospheric Ammonia Pollution and its Relation with $PM_{2.5}$ Chemical Property in Winter of Beijing, China

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Source: *Volume 16, No. 6, June 2016, Pages 1378-1389 doi: 10.4209/aaqr.2015.12.0699*

To understand the air pollution problem in megacities such as Beijing, field measurement investigating the variation of NH_3 and its association with $PM_{2.5}$ chemical property was conducted from 25 November to 24 December 2013. The results indicated that the daily concentration of wintertime NH_3 tended to be high on the days with relatively high temperatures and low wind speeds. Affected by the synoptic condition, NH_3 concentration showed a bimodal diurnal variation pattern, which tended to peak at around 09:00 and 22:00 of the day. As the sole precursor for NH_4^+, NH_3 exerted a significant impact on the ion chemistry of $PM_{2.5}$ through enhancing the nighttime NH_4Cl formation and promoting both homogeneous and heterogeneous formation of NO_3^-. During heavy pollution episodes with $PM_{2.5}$concentrations over 200 μg m^{-3}, the NH_3 levels and NH_4^+/NH_3 ratios grew simultaneously with the increase of $PM_{2.5}$ levels, indicating that NH_3 is one of the key reasons for heavy pollution events. Revealed by the features of measured ionic species in $PM_{2.5}$, in conjunction with the acidity analysis using thermodynamic model, our results suggested that NH_3 was frequently sufficient in wintertime atmosphere of urban Beijing and the fine particulates were neutralized nearly fully by NH_3.

Keywords: Ammonia; Aerosol chemistry; urban atmosphere; $PM_{2.5}$ pollution.

Multi-Method Observation and Numerical Simulation of a $PM_{2.5}$ Pollution Episode in Beijing in October, 2014

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Source: *Volume 16, No. 6, June 2016, Pages 1403-1415 doi: 10.4209/aaqr.2015.09.0532*

Multi-method observation and numerical simulation were applied to analyze a $PM_{2.5}$ pollution episode in Beijing in October, 2014. The results of vertical observation showed that surface-level backscatter signal and extinction coefficient increased during the episode, suggesting that air pollutants accumulated near the ground. The main meteorological factors during this episode could be described as calm wind, high relative humidity and low surface pressure. The evolution of $PM_{2.5}$ concentrations in this episode was divided into four stages, including two-steps type concentration climbing stages (P1 and P2), high concentration maintenance stage (P3) and rapid cleanup stage (P4). Analysis on ground-based observation, satellite remote sensing and atmospheric general circulation showed that regional transport, including crop residue burning, was the main incentive of this pollution episode. Subsequently, local pollutants emission and regional transport maintained and aggravated the episode under unfavorable meteorological conditions. Temporal variation of OX was in close agreement with that of $PM_{2.5}$ and the concentration peaks of OX occurred few hours before those of $PM_{2.5}$, which indicated that strong atmospheric oxidation could promote the formation of secondary $PM_{2.5}$. The results of numerical simulation showed that during 8–10 October, the average contribution of regional transport to $PM_{2.5}$ in the five sites exceeded 50%.

Keywords: Beijing; $PM_{2.5}$; Chemical compositions; Numerical simulation; Regional transport; Crop residue burning.

Characterisation of Absorbing Aerosols Using Ground and Satellite Data at an Urban Location, Hyderabad

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Source: *Volume 16, No. 6, June 2016, Pages 1427-1440 doi: 10.4209/aaqr.2014.09.0220*

In the present study we have attempted to characterize aerosols using their optical properties over a tropical urban location of Hyderabad, India. We have analyzed three years of in-situ data on aerosol absorption from Aethalometer and scattering from Nephelometer measurements. Satellite based absorption measurements from ozone monitoring instrument, absorbing aerosol index are also analyzed to investigate the role of long range transport of dust. Further, the Cloud–Aerosol Lidar Pathfinder Satellite Observations (CALIPSO) data is used to study the vertical extent of aerosol particles as well as their sphericity using its particulate depolarization ratio. The study revealed that irrespective of seasonal variation, local anthropogenic fossil fuel aerosols form the predominant aerosol type over this site. Biomass/dust aerosols in their pure form are not present during the study period; however the spread of frequency distribution of scattering Angstrom exponent and absorption Angstrom exponent suggested their possible existence in mixed condition with local anthropogenic aerosols. The analysis of columnar aerosol absorption data during pre-monsoon period showed the dominance of UV absorbing dust aerosols in the study region. CALIPSO data analysis over study area showed that majority aerosols are confined within 2 km from the surface during winter while in pre-monsoon particles are distributed throughout the profile (~6 km) with extinction coefficient varying between 0.1–0.2 km^{-1}. As the season shift from winter to pre-monsoon a change in sphericity of particle is observed. Cluster mean trajectory analysis revealed that during pre-monsoon majority of air mass movements (~68%) are from western side passing through dust source region like Persian Gulf and Thar Desert before entering into Indian region. During post-monsoon (~70%) and winter (~65%), majority of the air masses are coming from north-west and north-east side of the study area where biomass burning is quite frequent during this period

Keywords: Absorption coefficient; Angstrom exponent; Black carbon; Aerosol index and particulate depolarization ratio.

Personal Exposure and Dose of Inhaled Ambient Particulate Matter Bound Metals in Five European Cities

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Source: *Volume 16, No. 6, June 2016, Pages 1452-1463 doi: 10.4209/aaqr.2015.09.0536*

The objective of the current study is the determination of the personal exposure and dose of ambient particulate matter-bound metals in human tissues at five European cities. The accumulation in human body of lead (Pb), arsenic (As) and cadmium (Cd), in five European cities (Athens, Seville, Rome, Frankfurt and Zabrze) was calculated using an exposure and dose assessment model, ExDoM, and a pharmacokinetic model, PBPK. The study subjects are adult Caucasian non-smoker males. It was calculated that the highest dose of particulate matter is received from a resident of Seville, due to the higher ambient PM_{10} levels in the city compared to

the other sites. First, the current study showed that the European Union thresholds of particle-bound Pb, Cd and As concentrations were not exceeded in the cities under study. As regard the dose of Pb and As the higher dose is calculated for Athens and Seville, respectively. The highest dose of Cd is found at Zabrze, due to the high industrial activity in the city. It was calculated that after one day of exposure, the highest accumulation of Pb occurred in blood, muscles and bones. Furthermore, the highest deposition of Cd occurred in the lungs and intestines and for As in the lung and muscles. The heavy metals intake, calculated in this study, was very low in comparison with the recommended WHO levels for heavy metals intake from all types of exposure (inhalation, ingestion).

Keywords: Human exposure; Human metal dose; Exposure modelling; PBPK model; Respiratory tract model.

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Evaluating Dust Particle Transport Performance within Urban Street Canyons with Different Building Heights

Dan Mei, Qihong Deng, Meng Wen, Zhi Fang

Source: *Volume 16, No. 6, June 2016, Pages 1483-1496 doi: 10.4209/aaqr.2015.07.0436*

In developing cities, buildings with different heights obstruct the diffusion of pollutants. In this study, dust particles transported within urban street canyons were simulated using Reynolds-averaged Navier–Stokes (RANS) method, and air-solid two-phase flow fields were obtained on a regional scale. Four typical street building models were used in this study: (a) low-rise buildings (H/b = 1), (b) step-up building arrangements (H/b = 1, 2, 3, 4, 5), (c) step-down building arrangement (H/b = 5, 4, 3, 2, 1), and (d) high-rise buildings (H/b = 5). The particle volume fraction distribution in the four models reflected the basic properties of particle transportation in the canyons. Vortices were observed on the roofs of street canyons, which prevented particles from being transmitted into the canyons, and the vortex regions were characterized as low particle concentration. To evaluate the dust particle transport performance in the models, three indices, namely particle transport efficiency, suspension fraction and suspension density, were defined. These concepts were based on the particle number concentrations, which were obtained using the Lagrange approach. The high-rise building model (H/b = 5) demonstrated the lowest transport efficiency among all four models, and it also had the lowest suspension density in the street canyons. This implied that the high-rise buildings hindered the particles from being transporting further and that the particles could not enter the deep canyons easily. In the step-down building arrangement model (H/b = 5, 4, 3, 2, 1), the particle concentration level and suspension density in the canyons were lower than those observed in the step-up building arrangement model (H/b = 1, 2, 3, 4, 5), indicating that a step-down building height arrangement is appropriate for creating

construction plans for developing cities.Finally, the variation of the streamwise air velocity, vertical velocity and fluctuating velocity on the roof of canyons along the x direction were separately examined. Notably, the fluctuating velocity was the dominant mechanism of the particle suspension in the canyons.

Keywords: Building arrangement; Particle number concentration; Transport efficiency; Suspension fraction; Fluctuating velocity.

Comparison of Two Receptor Model Techniques for the Size Fractionated Particulate Matter Source Apportionment

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Source: *Volume 16, No. 6, June 2016, Pages 1497-1508 doi: 10.4209/aaqr.2015.06.0416*

The goal of the study is to investigate elemental concentrations and source signatures of particulate matter at a selected receptor site in an urban area Navi Mumbai which is one of the hot spots of India with heavy industrialization. To achieve the objective, particulate matter samples were collected from 2008–2010 using Gent sampler with the automatic dichotomous size segregation unit. It was observed that the three year average concentrations of coarse (($PM_{10-2.5\mu m}$) and fine ($PM_{2.5\mu m}$) fractions of PM were 89.92 (μg m^{-3}) and 42.25 (μg m^{-3}) respectively which were higher than the national standards prescribed by Central Pollution Control Board (CPCB), India. Subsequent elemental analysis of air filters using INAA and EDXRF showed marginally higher levels of anthropogenic derived elements. Furthermore, in the present study potentially contributing sources of coarse and fine PM were identified using two different receptor model techniques Factor Analysis (FA) and Positive Matrix Factorization (PMF). Six possible contributing sources of coarse fraction and seven probable sources of fine PM were identified by both the techniques. Further, sources identified by the receptor techniques and the comparability between the two techniques were also evaluated.

Keywords: Course; Fine; Receptor model; FA-MLR; PMF.

Small-Scale Study of Siberian Biomass Burning: II. Smoke Hygroscopicity

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Source: *Volume 16, No. 7, July 2016, Pages 1558-1568 doi: 10.4209/aaqr.2015.11.0648*

A lack of understanding about the impact of Siberian wildfire emissions on the environment necessitates the characterization of biomass burning aerosol hygroscopicity. Flaming fires of typical Siberian biomass (pine and debris) were simulated during small-scale combustion experiments in a Large Aerosol Chamber (LAC). Analyses of individual particles with respect to morphology and elemental composition allows the separation of freshly-produced smoke into five fractions with the elemental carbon, chain soot agglomerates, irregular internally mixed soot, and distinct irregular minerals of fly ash containing S, Ca, Al, and Si. Aging in a dark chamber leads to an appearance of the fraction with inorganic inclusions such as KCl and $CaCl_2$. Categorization of fresh-emitted and aged particles on hydrophobic, hydrophilic, and hygroscopic ones is performed. The criteria for categorization are extended from fossil fuel high-temperature combustion, based on a concept of water uptake by soot particles and utilization of a number of reference soots with known oxygen content and mixtures with sulfates and other inorganic salts. We show how the hydration properties of emitted smoke particles and inorganic inclusions can increase the initial level of smoke hygroscopicity.

Keywords: Biomass burning; Siberian wildfires; Smoke aerosol; Elemental composition; Fractionation; Water absorption; Hygroscopicity.

Seasonal Variation, Sources and Transport of Aerosols at Lijiang, Southeast Tibetan Plateau

Ningning Zhang, Junji Cao, Rujin Huang, Yuanqing He, Qiyuan Wang, Chongshu Zhu

Source: *Volume 16, No. 7, July 2016, Pages 1579-1590 doi: 10.4209/aaqr.2015.07.0470*

Aerosol samples were collected during pre-monsoon, monsoon and post-monsoon periods in 2009 in Lijiang, a tourism city located on the southeast Tibetan Plateau, southwest China. To determine the seasonal variation and sources of aerosol species, main elements and water soluble ions were analyzed. The results showed that crustal elements (Si, K, Ca, Ti and Fe) were the main elements with an enrichment whose enrichment factor (EF) value were lower than 10, with the large value (except Ca) occurring during the pre-monsoon period. The EF values of S, Cl, Zn, As, Br, Sb, Pb, and

Cu were higher than those of the crustal elements and the large concentration appeared during the monsoon period. Ca^{2+} and SO_4^{2-} were the dominant cation and anion, respectively. The greatest value of total ionic concentration was found during the monsoon period, mainly because of the high concentration of SO_4^{2-} and NH_4^+. Using the positive matrix factorization model, it was found that the main sources of species were from crustal source, transport from south Asia and eastern China, local vehicle emissions and sea salt. Further results indicated that the pollutants mixed with dust, anthropogenic pollutants and biomass burning emissions can be transported to Lijiang from south Asia and Southeast Asia during the pre-monsoon period. In addition, pollutants rich in SO_4^{2-} and heavy metal from the Sichuan Basin and eastern Yunnan Province can also be occasionally transported to Lijiang during the monsoon period. The seasonal differences in chemical composition and transportation pathway may have important implications for regional climate change.

Keywords: Aerosol; Chemical composition; Sources identification; Transport pathway; Lijiang.

Observations of New Particle Formation, Subsequent Growth and Shrinkage during summertime in Beijing

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Source: *Volume 16, No. 7, July 2016, Pages 1591-1602 doi: 10.4209/aaqr.2015.07.0480*

Size-resolved aerosol number concentrations (10 nm–10 μm in diameter) were measured at an urban site in Beijing during summertime of 2008. Case studies of new particle formation (NPF) are presented in this work. The measured mean particle formation rate was 2.37 cm^{-3} s^{-1}, which varied from 1.5 to 3.8 cm^{-3} s^{-1}, with growth rates ranging from 3.2 to 10.6 nm h^{-1}. NPF was observed under low number concentration of preexisting particles as well as under relatively high number concentration of preexisting particles. It was found that condensation contributed mainly and preferentially to particles growth, however, coagulation would contribute a lot when formation rate of new particles was sufficiently high. The variation of concentration of nucleation mode particles was found to be coincident with sulfur dioxide, indicating that NPF could occur under relatively high number concentration of preexisting particles if sufficient concentration of gas-phase H_2SO_4 existed in the atmosphere. Grown particles were also observed to shrink from 61.1 nm to 15.4 nm at a shrinkage rate of 16.6 nm h^{-1}, accompanied by a reduction of the particle number concentration. The shrinkage rate was higher than those reported in recent studies, probably due to particle shrinkage occurred during summertime in Beijing with higher temperature and lower RH compared to those observed in other regions, thus enhanced particle shrinkage.

Keywords: Particle formation; Growth; Particle shrinkage; Beijing.

Emission Regulations Altered the Concentrations, Origin, and Formation of Carbonaceous Aerosols in the Tokyo Metropolitan Area

Takuma Miyakawa, Yugo Kanaya, Yuichi Komazaki, Takao Miyoshi, Hideki Nara, Akinori Takami, Nobuhiro Moteki, Makoto Koike, Yutaka Kondo

Source: *Volume 16, No. 7, July 2016, Pages 1603-1614 doi: 10.4209/aaqr.2015.11.0624*

To investigate the effects of the regulations of diesel and non-methane hydrocarbon (NMHCs) emissions in the Tokyo metropolitan area (TMA) on the characteristics of carbonaceous aerosols (organic carbon (OC) and elemental carbon (EC)), we conducted field observations to characterize carbonaceous aerosols in the TMA in the summer of 2004 and 2014 (the end of July–middle of August). Following the enforcement of diesel emission regulations, EC concentrations showed a four-fold decrease from 2004 to 2014. However, OC concentrations showed no significant decrease in the last decade. Multiple chemical analyses revealed the differences in the impacts of the contribution of oxygenated fraction, biogenic NMHCs on OC, and the secondary organic aerosol–Ozone relationship between 2004 and 2014. Further investigations into the emission inventory for recent years, especially in terms of precursor gases, are needed for better prediction of OC in the TMA using chemical transport models.

Keywords: Emission regulations; Carbonaceous aerosols; SOA; Biogenic carbons.

Chemical characteristics of rainwater in Sichuan basin, a case study of Ya'an

Yun-Chun Li, Meng Zhang, Man Shu, Steven Sai Hang Ho, Zi-Fang Liu, Xian-Xiang Wang, Xiao-Qing Zhao

Source: *Environmental Science and Pollution Research,July 2016, Volume 23, Issue 13, pp 13088–13099*

Rainwater chemistry was investigated at a semi-rural site in Ya'an, Sichuan basin with rain samples collected from May 2013 to July 2014. The rainwater pH values ranged from 3.25 to 6.86, with an annual volume-weighted mean (VWM) of 4.38, and the acid rain frequency was 74 %. Such severe

acidification, 15 % of the total events showed a pH below 4.0, attributed to the deficiency of Ca2+, significant anthropogenic pollution contribution, and rainy pattern to this area. The annual VWM of total ions concentration was 477.19 μeq/L. NH4+ was the most abundant ionic species, followed by SO42−, NO3−, Ca2+, Cl−, Na+, K+, Mg2+, and F− in a descending order. The total ionic concentrations presented a seasonal trend of lower values in autumn and summer but higher ones in winter and spring. Based on enrichment factor, correlation analysis and principle component analysis, three factors were identified: factor 1 (NH4+, SO42−, NO3−, K+, and Cl−, 47.45 % of the total variance) related to anthropogenic sources (coal/fuel combustion, biomass burning and agriculture), factor 2 (Ca2+, Mg2+, Na+, and Cl−, 34.01 % of the total variance) associated with natural sources, and factor 3 (H+, 11.78 % of the total variance) related to free acidity. Back trajectory analysis indicates that the rainwater chemistry in Ya'an was mainly affected by regional air masses from Sichuan basin. Long-range transported air masses from southwest with heavy anthropogenic pollution increased the total ion concentration and acidity of rainwater. Considering its special topography, anthropogenic emissions from regional and long-range transport (especially from southwest) must be controlled effectively to improve the acid rain condition of non-urban areas in Sichuan basin.

Keywords: Acid rain, Chemical composition, Sources identification, Sichuan basin

Size-Segregated Characteristics of Carbonaceous Aerosols over the Northwestern Indo-Gangetic Plain: Year Round Temporal Behavior

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Source: *Volume 16, No. 7, July 2016, Pages 1615-1624 doi: 10.4209/aaqr.2016.01.0023*

Size-segregated aerosol samples ($PM_{<0.95}$, $PM_{0.95–1.5}$, $PM_{1.5–3.0}$, $PM_{3.0–7.2}$ and $PM_{>7.2}$) were collected over Patiala (30.33°N, 76.40°E; 250 m amsl), a semi-urban city located in northwestern Indo-Gangetic Plain (IGP) during October, 2012 to September, 2013. These samples were analyzed for carbonaceous aerosols (organic carbon (OC), elemental carbon (EC) and water-soluble organic carbon (WSOC)) to study their temporal variation, prevailing emission source (s) and secondary formation processes. Annual average of total suspended particulates (TSP) concentration, estimated by adding the aerosol concentrations in different size ranges, was found to be 199 ± 82 μg m^{-3}, varying from 88–387 μg m^{-3} with majority of particulate mass found in submicron size ($PM_{<0.95}$). OC3.0 and WSOC3.0 ranged from 5.4 to 70 μg m^{-3} (23 ± 14 μg m^{-3}) and 2.5 to 37 μg m^{-3} (12 ± 8.7 μg m^{-3}), respectively over an annual cycle. Highest mass fraction of OC (> 75%) and WSOC (> 80%) was observed in submicron size, suggesting that OC mainly comes from combustion and/or secondary source (s). It has been observed that almost half of OC is secondary. On the other hand, climate forcing agent EC in $PM_{<0.95}$ varied from 1.1 to 9.8 μg m^{-3} (4.8 ± 2.2 μg m^{-3}). High mass

ratios of OC/EC (~1.5–7.2) and WSOC/OC (~0.33–0.68) indicate the relative dominance of biomass burning emission over the study region. Total carbonaceous aerosols account for ~10–59% of submicron particulates mass ($PM_{<0.95}$), indicating that fine particulates are enriched with carbonaceous species. These results have implications to regional climate model development and validation.

Keywords: Organic carbon; Water-soluble organic carbon; Elemental carbon; Secondary organic aerosols; Biomass burning emissions.

Characteristics of Haze Pollution Episodes and Analysis of a Typical Winter Haze Process in Shanghai

Mengfei Zhao, Guangli Xiu, Ting Qiao, Yulan Li, Jianzhen Yu

Source: *Volume 16, No. 7, July 2016, Pages 1625-1637 doi: 10.4209/aaqr.2016.01.0049*

One-year field campaign was conducted from July 2013 to August 2014 at the site of East China University of Science and Technology (ECUST) in urban Shanghai, and mass concentrations and chemical compositions of $PM_{2.5}$ were measured. Gaseous pollutants (SO_2, NO_2) and meteorological parameters (wind speed, wind direction, pressure, temperature and relative humidity) were simultaneously obtained. In this study, $PM_{2.5}$ mass balances on haze and non-haze days were reconstructed and the sum of secondary inorganic aerosols (SIA) and organic matter (OM) accounted for over 80%. The fraction of nitrate in SIA was much higher on haze days than that on non-haze days, while the corresponding fraction of ammonium was lower, implying that the variations of the sources and formation processes of SIA on haze days. In theory, sulfate and nitrate might be almost fully neutralized by ammonium. Moreover, the sulfur oxidation ratio (SOR) values were much higher than the nitrogen oxidation ratio (NOR) values, indicating the greater oxidation capacity of SO_2 would occur. On haze days, the high NOR values could be explained by the relatively low temperature, the high NO_2 concentration and the potential dominant gas-phase reaction. As for secondary organic aerosol (SOA), the formation processes were usually associated with nitrate formation. In winter, haze pollution episodes occurred more frequently than those in other seasons, associated with the different features of wind speed, wind direction and 72-h backward trajectory. In addition, one case from 17 November 2013 to 4 January 2014 was selected to investigate the formation mechanism of haze pollution episodes. The key factors that affected the haze formation might be the local stable synoptic conditions including weak surface wind, surface temperature inversion and high relative humidity, the long-range transportations from the Northwest and the large amounts of emissions from local sources.

Keywords: Haze pollution episode; $PM_{2.5}$; Secondary inorganic aerosols (SIA); Secondary organic aerosols (SOA); Meteorological conditions.

$PM_{2.5}$ Chemical Composition Analysis in Different Functional Subdivisions in Tangshan, China

Wei Wen, Shuiyuan Cheng, Lei Liu, Xufeng Chen, Xiaoqi Wang, Gang Wang, Song Li

Source: *Volume 16, No. 7, July 2016, Pages 1651-1664 doi: 10.4209/aaqr.2015.09.0559*

In this study, the 24-h $PM_{2.5}$ (i.e., the fine particles with aerodynamic diameter of ≤ 2.5 μm) samples were collected at two different functional subdivisions in the city of Tangshan during the period of July 2012 to April 2013. The months of July, October, January, and April were chosen to represent four different typical seasons. The total PM2.5 mass was measured. $PM_{2.5}$ samples were used for the analysis of inorganic elements, ions, Organic Carbon (OC) and Elemental Carbon (EC). $PM_{2.5}$ concentration in the industrial subdivision were generally higher than those in the residential subdivision. The annual mean $PM_{2.5}$ concentrations were 196 μg m^{-3} for the industrial subdivision and 116 μg m^{-3} for the residential subdivision. The coefficients of divergence (CD) calculated for spring, summer, autumn and winter were 0.67, 0.35, 0.65, and 0.33, respectively. The Enrichment Factor Method (EFM) was used to help determine the original sources of these inorganic elements. Secondary water-soluble ions (SO_4^{2-}, NO_3^-, and NH_4^+) were the major water soluble ions in the $PM_{2.5}$ of Tangshan, and they are present in the atmosphere as $(NH_4)_2SO_4$ and NH_4NO_3. The Sulfur Oxidation Ratio (SOR) and Nitrogen Oxidation Ratio (NOR) show that the precursor conversion ratio was highest in summer. At both subdivisions, winter had the highest monthly average OM concentration while summer had the lowest.

Keywords: $PM_{2.5}$ pollution; Chemical composition; Residential subdivision; Industrial subdivision.

Atmospheric Bulk Deposition of PAHs over Brahmaputra Valley: Characteristics and Influence of Meteorology

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Source: *Volume 16, No. 7, July 2016, Pages 1675-1689 doi: 10.4209/aaqr.2016.02.0060*

Bulk atmospheric deposition of Polycyclic Aromatic Hydrocarbons (PAHs) in Guwahati city of the Brahmaputra Valley have been characterised for a period of one year. The ∑PAHs (USEPA's priority 16) and benzo(s)pyrene (BaP) concentrations in the collected bulk deposit ranged between 2.2 and 1035 ng mL^{-1}, and BDL and 5.6 ng mL^{-1} respectively. Greater deposition of PAHs was observed during the dry season and the deposition of low molecular weight PAHs (LMWPAHs) were particularly high. The study revealed explicit effect of the prevailing meteorology on the bulk atmospheric deposition characteristics of PAHs. Greater number of inversion days and lowering of inversion height during the dry season were found to enhance deposition of PAHs. Diagnostic ratios indicated pyrogenic origin of the PAHs derived from combustion of diesel, coal and wood combustion. The deposition of LMWPAHs was greatly influenced by temperature. The diurnal variation of relative humidity, and wind pattern were found to affect deposition of PAHs.

Keywords: PAHs; Atmospheric bulk deposition; Deposition mass flux; Meteorology.

Removal Efficiency of Bimodal PM2.5 and PM10 by Electret Respirators and Mechanical Engine Intake Filters

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Source: *Volume 16, No. 7, July 2016, Pages 1722-1729 doi: 10.4209/aaqr.2015.08.0494*

As China is receiving an economic boom, PM (particulate matter) pollutions not only have become a serious regional problem but also frequently impacted its neighboring counties, e.g., Korea and Japan. In addition to its adverse effects on human health, the on- and off-road engines operated in ambient can also be affected. In this study, a simple system for generating simulated ambient bimodal PMs comprising fine ($PM_{2.5}$), coarse ($PM_{2.5–10}$) particles was developed for evaluating the initial efficiency of seven respirator and four engine intake filters. In addition to the size fractional efficiency curves for each filter media determined from the SMPS (scanning mobility particle sizer) and APS (aerodynamic particle sizer), both number and mass based efficiency of these filters for $PM_{2.5}$, $PM_{2.5–10}$ and PM_{10} were also obtained to evaluate their performances against ambient PM pollutions. Data showed that the engine intake filters had a low efficiency for both mass and number based $PM_{2.5}$, which was only about 25–30%. However, there was a large difference between their number and mass based PM_{10}efficiency. The former was much lower than the latter because these filters are with high efficiency only for coarse particles. Besides, the most of particles in number is resided in the fine size range while the mass is in coarse size range. For the respirator filtration tests, results showed that most of them can effectively remove both $PM_{2.5}$ and PM_{10}, in which the mass efficiency was always higher than that of number. The $PM_{2.5}$ number efficiency

results showed there are three out of seven respirator filters are with N-95 rated level, in which the efficiency of their most penetrating particle size is higher than 95%. The current simple experimental system could be applied to examine different purpose filters which protect human health and outdoor engines against ambient $PM_{2.5}$ and PM_{10}.

Keywords: $PM_{2.5}$ in China; $PM_{2.5}$ health effect; PM_{10}; Bimodal ambient PM; Electret respirator; Mechanical engine intake filter; Long range transport.

Research into Haze Removal Method Based on Diffusion and Relative Motion

Lian-Ze Wang, Hong-Tao Niu, Ning-Ning Peng, Xiong Shen

Source: *Volume 16, No. 7, July 2016, Pages 1757-1763 doi: 10.4209/aaqr.2015.11.0650*

A method for purifying polluted air in large open space was proposed and verified based on concentration gradient diffusion theory and relative motion principle of purifying device to polluted air. As to the sources of particle emitting, the purifying device works as a sink no matter it is moving or not. Both indoor and outdoor experiments have been made. In the indoor experiment, severe haze environment was simulated by fuming. The purifying device was made of metal wire and carbon cloth. The fixed device took 2.7, 6.2 and 13 minutes to reduce the particle concentration in the 7 m farthest corner of the laboratory to 50%, 20% and 0 respectively. By moving the device at 1 m s^{-1}, the concentration could be reduced to 50% in merely 15 seconds. In outdoor experiment, 23.5% decrease of haze concentration was measured.

Keywords: Concentration gradient; Diffusion; Relative motion; Haze removal.

Atmospheric PM2.5 and Depositions of Polychlorinated Dibenzo-p-dioxins and Dibenzofurans in Kaohsiung Area, Southern Taiwan

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Source: *Volume 16, No. 7, July 2016, Pages 1775-1791 doi: 10.4209/aaqr.2016.04.0168*

Kaohsiung County, in southern Taiwan, has both highly industrial and rural areas. In this study, the characteristics of PCDD/Fs in the ambient air of Kaohsiung (urban), Meinong (rural) and Xiaogang (heavy industrial zone) in 2014 and 2015 were modeled based on the prevailing meteorological conditions and the measured ambient air concentrations of $PM_{2.5}$, PM_{10} and TSP. The yearly average $PM_{2.5}$concentrations in the ambient air of the three areas were in the range of 23 to 31 μg m^{-3}, all above the National Air Quality Standard of Taiwan (15 μg m^{-3}). The simulated average concentrations of PCDD/Fs in the whole of Kaohsiung area in terms of toxicity equivalent were in the range of 0.034–0.053 pg WHO_{2005}-TEQ m^{-3}. The average total deposition fluxes of total-PCDD/Fs-WHO_{2005}-TEQ ranged between 155.4 and 276.6 pg WHO_{2005}-TEQ m^{-2} $month^{-1}$ with 1,2,3,7,8-PeCCD and 2,3,4,7,8-PeCDF being the dominant congeners in terms of PCDD/Fs WHO_{2005}-TEQ. Xiaogang area with highly industrial activities had the highest concentrations of $PM_{2.5}$ and PCDD/Fs and corresponding total-PCDD/Fs WHO_{2005}-TEQ deposition fluxes, while the Meinong in the rural site recorded the lowest. Average dry deposition velocities of total PCDD/Fs WHO_{2005}-TEQ for both 2014 and 2015 were 0.162, 0.148 and 0.161 cm s^{-1} for Kaohsiung, Meinong and Xiaogang, respectively, while the average scavenging ratios of total PCDD/Fs WHO_{2005}-TEQ were 6232, 4701 and 6802, for Kaohsiung, Meinong and Xiaogang, respectively. The information provided in this work is useful for both further studies and environmental control strategies concerning atmospheric aerosols and dioxins.

Keywords: PCDD/Fs; $PM_{2.5}$; PM_{10}; Dry deposition; Wet deposition.

Characterization of Background Aerosol Properties during a Wintertime Smog Episode

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Source: *Volume 16, No. 8, August 2016, Pages 1793-1804doi: 10.4209/aaqr.2015.04.0205*

The aim of this paper is to study the wintertime physical properties of atmospheric aerosol particles on the basis of data observed at the K-puszta regional background station in Hungary. In Hungary wintertime smog episodes are linked to strong stable air (high pressure blocking events) with thermal inversion. These atmospheric conditions are frequently formed during winter months (November–February) due to the special geographical location of the country. The formation of smog events is highly probable in cases of thermal inversion periods sustaining for at least 4 days. We discuss in the paper the role of high-pressure blocking events in aerosol properties in terms of PM_{10} concentrations, aerosol size distributions, new particle formation and optical properties. We found that high-pressure blocking events have significant impacts on the size distribution and particle formation processes. At K-puszta the aerosol is in highly aged state with size distribution dominated by the accumulation mode. This is further supported by the optical properties, e.g., by

high scattering Ångstrom exponent and by relatively weak absorption. The most significant effect of extreme episodes is manifested in the changes in PM_{10} concentrations and, consequently, in aerosol optical properties. The PM_{10} concentrations, scattering coefficients and absorption coefficients considerably increase to extreme values that are characteristic of a heavily polluted atmosphere rather than rural air. Our results indicate that in winter, the air quality at K-puszta is often influenced by regional air pollution as shown by spatial distribution of PM_{10} concentration. It is found that PM_{10} had almost the same concentration in regional background air and in different types of urban environments. The special meteorological conditions and the role of regional-scale transport can explain why local abatements in cities cannot lead to significant improvement of the air quality during smog events.

Keywords: PM_{10}; Aerosol size distribution; Particle formation; Scattering and absorption; Air pollution episode.

Quantifying the impact of PM2.5 and associated heavy metals on respiratory health of children near metallurgical facilities

Daniel Dune, Stefania Iordache, Hai-Ying Liu, Trond Bøhler, Alin Pohoata, Cristiana Radulescu

Source: *Environmental Science and Pollution Research, August 2016, Volume 23, Issue 15, pp 15395–15406*

The aim of this study was to link the concentrations of particulate matter with an aerodynamic diameter below 2.5 μm (PM2.5) and associated heavy metals with occurrence of wheezing and hospitalizations due to wheezing in 111 children who live near metallurgical plants in Targoviste City, Romania. A group of 72 children with high levels of immunoglobulin E (IgE) and eosinophils, as well as frequent wheezing episodes, was geolocated on digital thematic maps. Monitoring campaigns and medical assessments were performed over two consecutive years (2013–2014). The multiannual average concentrations of PM2.5 ranged from 4.6 to 22.5 μg m−3, up to a maximum value of 102 μg m−3. Significant correlations ($p < 0.01$) were observed between the locations of the children with respiratory issues and the PM2.5 multiannual average ($r = 0.985$) and PM2.5 maximum ($r = 0.813$). Fe, Ni, Cd, and Cr were the main marker elements of the emissions from steel production and metal-working facilities in the Targoviste area. The results support the hypothesis that increased PM2.5 levels directly influence wheezing symptom and asthma attacks in the analyzed group. IgE, eosinophils, and wheezing episodes may be considered key indicators with which to evaluate the adverse effects of PM2.5 air pollution on children's health.

Keywords: Fine particulates, Environmental mapping system, Wheezing, Immunoglobulin E, Eosinophil count

Seasonal Chemical Characteristics of Atmospheric Aerosol Particles and its Light Extinction Coefficients over Pune, India

Atar Singh Pipal, Suresh Tiwari, P. Gursumeeran Satsangi

Source: *Volume 16, No. 8, August 2016, Pages 1805-1819 doi: 10.4209/aaqr.2015.08.0529*

The present study has been conducted to characterize atmospheric aerosol particles in terms of carbonaceous species and ionic constituents for a yearlong period at Pune, India. This study provides the evidence for the ionic chemistry, secondary aerosols formation, temporal variability and its climatic effect in the atmosphere. The average concentrations of $PM_{2.5}$ and PM_{10} were 109.6 ± 23.2 and 166.9 ± 4 μg m^{-3}, respectively, by far exceeding National Ambient Air Quality (NAAQ) and World Health Organization (WHO) standards. Seasonal analyses indicated that $PM_{2.5}$ and PM_{10} mass concentrations were higher in the post-monsoon followed by the winter season and lower during the monsoon period. The average concentrations of organic carbon (OC) and elemental carbon (EC) were 31.3 ± 7.4 and 4.2 ± 2.4 μg m^{-3} for $PM_{2.5}$, while, 34.2 ± 6.2 and 5.0 ± 2.3 μg m^{-3} for PM_{10}, respectively. OC and EC data splits into seasons and their mass loadings were in the order of post-monsoon > monsoon > winter > summer for OC and for EC, it was as winter > post-monsoon > summer > monsoon. The overall chemical analysis revealed that particulate matter (PM) consist higher concentrations of OC followed by cations and the lowest one is EC. The ionic composition analysis indicated that cations were the abundant parts of PM in comparison to anions and Na+ and SO_4^{2-} were at a higher concentration amongst all the ionic species. The estimated light extinction coefficient (b_{ext}) of the aerosol particle was 291.2 ± 55.3 Mm^{-1} during the study period. Further apportionment of particle extinction coefficient was estimated and the contributions of light scattering coefficient by particles (*bsp*) were OC (45%), $(NH_4)_2SO_4$ (17%), NH_4NO_3 (8%) and coarse mass (12%), while, the contribution of light absorption coefficient by particle (*bap*) was 18% (EC). This indicates that in the present study the abundance of aerosol particles are more scattering in nature in comparison to absorption. The average value of Aerosol optical depth (AOD) was 0.46 and their positive correlation with anions and relative humidity (RH) showing same properties, while, in the case of EC, it showed contrast nature with respect to climate effect. Trajectory analysis indicated that the air masses appear as a result of long-range transportation during summer and monsoon period while during the winter and post-monsoon seasons local manmade activities showed dominant influence.

Keywords: Ionic chemistry; OC and EC; Light extinction; $(NH_4)_2SO_4$ and NH_4NO_3; AOD; Sources.

Temporal Assessment of NO2 Pollution Levels in Urban Centers of Pakistan by Employing Ground-Based and Satellite Observations

Muhammad Fahim Khokhar, Hadiqa Mehdi, Zain Abbas, Zeeshan Javed

Source: *Volume 16, No. 8, August 2016, Pages 1854-1867 doi: 10.4209/aaqr.2015.08.0518*

This study presents the assessment of nitrogen dioxide (NO_2) pollution within the twin cities of Islamabad and Rawalpindi by using car MAX-DOAS (Multi Axis-Differential Optical Absorption Spectroscopy) instrument during two different scenarios of with and without the availability of CNG (compressed natural gas) fuel. Information perceived from this study can be used to get an idea about recent air quality conditions of twin cities and its repercussions on ecological and human health. International air quality monitoring field campaigns were conducted during November 2012 and December 2013. Results showed different concentrations of NO_2 (68.2, 74.25 and 93.65 ppb) at various locations of Islamabad and Rawalpindi cities, exceeded the Pak-NEQS levels of 42.5 ppb. High NO_2 concentrations can be attributed to emissions from an oil refinery, traffic congestion and solid waste dump site along IJP road, and due to non-availability of CNG during December 2013. Results compared with OMI satellite observations exhibited that NO_2 columns from OMI observations are largely underestimated.

Keywords: CNG fuel; Car MAX-DOAS; Satellite validation; Air pollution; Pak-NEQS.

Statistical Downscaling of Air Dispersion Model Using Neural Network for Delhi

Anikender Kumar, Pramila Goyal

Source: *Volume 16, No. 8, August 2016, Pages 1879-1892 doi: 10.4209/aaqr.2015.06.0384*

Statistical downscaling methods are used to extract high resolution information from coarse resolution models. The accuracy of a modelling system in analyzing the issues of either continuous or accidental release in the atmosphere is important especially when adverse health effects are expected to be found. Forecasting of air quality levels are commonly performed with either deterministic or statistical. In this study, statistical downscaling approach is investigated for hourly PM_{10} (particulate matter with aerodynamic diameter < 10 μm) pollutant for Delhi. The statistical downscaling is used on air dispersion model using neural network technique. The air dispersion model is based on analytical solution of advection diffusion equation in Neumann boundary condition for a bounded domain. Power laws are assumed for height dependent wind speed; and

downwind and vertical eddy diffusivities are considered as an explicit function of downwind distance and vertical height. The predicted concentration of dispersion model with meteorological variables is used as input parameters to the neural network. It is found that performance of both air dispersion model and "pure" statistical models is inferior to that of the statistical downscaled model. In particular the root mean squares error (RMSE) of the deterministic model is reduced by at least 35% and 45% for hourly and rush hours particulate matter concentrations respectively using statistical downscaling. In addition, the results with statistical downscaled method show that the errors of the forecasts are reduced by at least 30% for stable and unstable-neutral atmospheric conditions.

Keywords: Statistical downscaling; Air dispersion model; Neural network; PM_{10}; Delhi.

Characteristics of Persistent Organic Pollutant Emissions from a Diesel-Engine Generator Fueled Using Blends of Waste Cooking Oil-Based Biodiesel and Fossil Diesel

Jen-Hsiung Tsai, Shui-Jen Chen, Kuo-Lin Huang, Guo-Ping Chang-Chien, Wen-Yinn Lin, Chien-Wei Feng, Jin-Yuan Syu, Ho-Tsang Huang

Source: *Volume 16, No. 8, August 2016, Pages 2048-2058 doi: 10.4209/aaqr.2016.06.0257*

This study elucidates the characteristics of persistent organic pollutants (POPs), including polychlorinated dibenzo-p-dioxins and dibenzofurans (PCDD/Fs), polychlorinated biphenyl (PCBs), polybrominated dibenzo-p-dioxins and polybrominated dibenzofurans (PBDD/Fs) and polybrominated diphenyl ethers (PBDEs), that are emitted from a generator (non-road diesel engine) that is fueled with a blend of waste cooking oil biodiesels (WCO-biodiesels). Experimental results reveal that the mass concentrations of PCDD/Fs, PCBs, PBDD/Fs and PBDEs from the diesel generator that is fueled with petroleum diesel (D100) under loads of 1.5 kW and 3.0 kW were 583–875 pg Nm^{-3}, 580–810 pg Nm^{-3}, 982–1408 pg Nm^{-3}, and 134–216 ng Nm^{-3}, respectively, while the toxicity concentrations of PCDD/Fs, PCBs, and PBDD/Fs were 33.2–58.6, 2.72–3.11, and 1.54–2.30 pg WHO_{2005}-TEQ Nm^{-3}, respectively. W20 and W40 emitted 15–65% and 23–85% less of these POPs by mass (and 25–47% and 46–91% by toxicity), respectively. Among the tested fuel blends, the W40 exhibited the greatest emission factor reductions of mass and toxicity of these four POPs, despite of the load. Therefore, WCO-biodiesels can feasibly be used as an alternative generator fuel, favoring the recycling/reuse of waste oils and mitigating hazards to human health and environment.

Keywords: Persistent organic pollutant; PCDD/F; PBDE; Waste cooking oil; Biodiesel.

Characteristics and Sources of Black Carbon in Atmospheric Dustfall Particles from Huangshi, China

Changlin Zhan, Jiaquan Zhang, Junji Cao, Yongming Han, Ping Wang, Jingru Zheng, Ruizhen Yao, Hongxia Liu, Hua Li, Wensheng Xiao

Source: *Volume 16, No. 9, September 2016, Pages 2096-2106 doi:10.4209/aaqr.2015.09.0562*

Concentrations of carbonaceous particles in atmospheric dustfall particles in Huangshi, an industrial city in central China, were determined using a thermal-optical reflectance method. The black carbon (BC) contents in ninety-five dustfall samples ranged from 4.3 to 64.9 g $kg^{-1}$with an average of 17.0 g kg^{-1}. These values were higher than those in world background soils and demonstrated serious contamination of the environment in this city. Overall, BC accounted for 17.6–71.3% (mean: 42.0%) of the organic carbon (OC), and BC and OC were positively correlated (r^2 = 0.90). Average char and soot contents were 8.01 g kg^{-1} and 8.65 g kg^{-1}, respectively, and char/soot ratios ranged from 0.28 to 1.97 with an average of 1.01. All the measured carbonaceous species positively correlated with each another, suggesting their common sources. BC, char, and soot showed large spatial distribution variability, with high levels of BC adjacent to the presumed emission sources, such as a power plant and railway line. Analyses of BC/OC and char/soot ratios indicate major impacts from fossil fuel combustion, especially motor vehicle emissions and coal combustion. Industrial dusts related to coal use appear to be the major contributor to BC in dustfall, and this is likely related to the extensive industrial activities in the city, including metal smelting.

Keywords: Black carbon; Dustfall; Char; Soot; Sources.

Process Contributions to Secondary Inorganic Aerosols during Typical Pollution Episodes over the Pearl River Delta Region, China

Zhijiong Huang, Jiamin Ou, Junyu Zheng, Zibing Yuan, Shasha Yin, Duohong Chen, Haobo Tan

Source: *Volume 16, No. 9, September 2016, Pages 2129-2144 doi:10.4209/aaqr.2015.12.0668*

The Integrated Process Rate (IPR) analysis embedded in CAMx model was used to quantify contributions from different atmospheric processes to the formations and accumulations of ambient $PM_{2.5}$ and the secondary inorganic aerosol (SIA) during two typical particulate pollution episodes in different seasons in the Pearl River Delta (PRD) region. Process analysis results indicated that primary fine particle emissions were the major sources of high ambient $PM_{2.5}$ in urban areas with intensive anthropogenic activities. Aerosol process and advection transport were another two major processes contributing to the increasing $PM_{2.5}$ and SIA over the PRD region. Regarding formation of SIA species, elevations of nitrate and ammonium at Guangzhou (urban), Heshan (rural) and Panyu (suburban) sites were largely associated with aerosol process, while those at Huizhou (urban) site were dominated by advection process, but elevated sulfate concentrations at these four sites were all dominated by advection process. The difference can be attributed to spatial variations of SO_2, NO_x and NH_3 emissions, site locations and meteorological conditions. Advection, aerosol chemistry, deposition and vertical diffusion were important pathways to remove SIA at these four sites. Within the hours with most growing $PM_{2.5}$ concentrations, aerosol process was the most important contributor to the formation of new SIA throughout the entire planetary boundary layer.

Keywords: Secondary inorganic aerosol; $PM_{2.5}$; PRD; CAMx; Process analysis.

Effects and mechanism of acid rain on plant chloroplast ATP synthase

Jingwen Sun, Huiqing Hu, Yueli Li, Lihong Wang, Qing Zhou, Xiaohua Huang

Source: *Environmental Science and Pollution Research, September 2016, Volume 23, Issue 18, pp 18296–18306*

Acid rain can directly or indirectly affect plant physiological functions, especially photosynthesis. The enzyme ATP synthase is the key in photosynthetic energy conversion, and thus, it affects plant photosynthesis. To clarify the mechanism by which acid rain affects photosynthesis, we studied the effects of acid rain on plant growth, photosynthesis, chloroplast ATP synthase activity and gene expression, chloroplast ultrastructure, intracellular H+ level, and water content of rice seedlings. Acid rain at pH 4.5 remained the chloroplast structure unchanged but increased the expression of six chloroplast ATP synthase subunits, promoted chloroplast ATP synthase activity, and increased photosynthesis and plant growth. Acid rain at pH 4.0 or less decreased leaf water content, destroyed chloroplast structure, inhibited the expression of six chloroplast ATP synthase subunits, decreased chloroplast ATP synthase activity, and reduced photosynthesis and plant growth. In conclusion, acid rain affected the chloroplast ultrastructure, chloroplast ATPase transcription and activity, and Pn by changing the acidity in the cells, and thus influencing the plant growth and development. Finally, the effects of simulated acid rain on the test indices were found to be dose-dependent.

Keywords: Acid rain, Rice leaves, Chloroplast, ATP synthase, Gene expression

Prevalence of Freshly Generated Particles during Pollution Episodes in Santiago de Chile

Ernesto Gramsch, Felipe Reyes, Yeanice Vásquez, Pedro Oyola, María A. Rubio

Source: *Volume 16, No. 9, September 2016, Pages 2172-2185 doi:10.4209/aaqr.2015.12.0691*

A winter campaign was carried out in Santiago de Chile the year 2012 in two urban sites that can be considered representative for most of the city in order to characterize formation of primary and secondary $PM_{1.0}$ during episodes. One site is located in the campus of the University of Santiago and measurements were carried out with an Aerosol Chemical Speciation Monitor and a black carbon monitor. Another site is located in a large park, about 2 km south-east of the first site, measurements of CO, NO_x, SO_2, O_3 were done in this site. A noticeable increase in most of the primary components of $PM_{1.0}$ (black carbon and organics) and primary gases (CO and NO) was observed during days in which the average $PM_{1.0}$ concentration was higher than 50 μg m^{-3} (episode). A small increase or no change was observed in the secondary pollutants (NH_4, NO_3, SO_4 and NO_2) at night during these episodes. Positive Matrix Factorization was used to extract four components from the ACSM data: hydrocarbon-like organic aerosol (HOA), biomass burning OA (BBOA), low volatility oxygenated OA (LV-OOA) and semi-volatile oxygenated OA (SV-OOA). The freshly generated components (HOA and BBOA) showed a clear increase at night during episodes, while the aged fraction of organic aerosol (LV-OOA and SV-OOA) showed a smaller increase or a decrease at night during episodes. Correlation of HOA and BBOA components with primary pollutants was also high, indicating that freshly created aerosols (HOA, BBOA and BC) are in large part responsible for the increase in pollution at night during episodes in Santiago de Chile.

Keywords: Pollution episode; Primary particle; Secondary organic aerosol (SOA).

Control of Bioaerosols in Indoor Environment by Filter Coated with Nanosilicate Platelet Supported Silver Nanohybrid (AgNPs/NSP)

Jiung-Wen Chen, Grace Whei-May Lee, Kai-Jie Chen, Shin-Hao Yang

Source: *Volume 16, No. 9, September 2016, Pages 2198-2207 doi:10.4209/aaqr.2016.06.0224*

Currently silver nanoparticles (AgNP)-modified filter are widely used to inactivate airborne microbes in indoor environment. However, AgNP is extremely small and thus will penetrate cells membranes to cause cytotoxicity. AgNPs/NSP has been proven to be less cytotoxic to human body. In this study, it was the first time that AgNPs/NSP was used to develop a new antimicrobial air filter with low cytotoxicity. The AgNPs/NSP filter was made by dip-coating of filter with AgNPs/NSP and acrylic resin solution and three different amount of silver on filter were obtained including 12.6, 31.5 and 63 ppm. The filtration efficiency and the antimicrobial activity of AgNP/NSP filter were evaluated by bioaerosols including *Escherichia coli* and *Candida famata* in testing chamber and HVAC simulation system under 30% and 70% relative humidity (RH). The results showed that filtration efficiency of AgNPs/NSP-modified filter increased by about 13 to 20% compared to unmodified filter for *E. coli* but remained almost the same for *C. famata*. The antimicrobial efficiency of AgNPs/NSP modified filter of 63 ppm was 95.1% for *E. coli* at RH of 30%. In addition, 91% of antimicrobial efficiency for *C. famata* was found at RH of 70%. On the other hand, the antimicrobial efficiency of yeast for AgNPs/NSP-modified filter was 97.8% and 86.4% for RH of 30% and 70% respectively when yeast just started to contact with filter in HVAC system. The results suggest that AgNP/NSP-modified air filter can effectively inactivate microorganisms retained on. Therefore, emission of bioaerosols from air filter can be avoided in order to improve the air cleaning technology in indoor environment.

Keywords: Silver nanoparticles; Filtration; Antimicrobial activity; Airborne microbes; Antimicrobial filter.

Upgrading the Estimation of Daily PM10 Concentrations Utilizing Prediction Variables Reflecting Atmospheric Processes

Konstantinos Dimitriou

Source: *Volume 16, No. 9, September 2016, Pages 2245-2254 doi:10.4209/aaqr.2016.05.0214*

This paper formulates a Multiple Linear Regression Model (MLRM), for the estimation of daily PM_{10} concentrations in background urban areas. 24-hour backward air mass trajectories, NO_2 concentrations and gridded (1° × 1° resolution) Aerosol Optical Depth (AOD) observations from MODIS were used in order to compose the model's predictor variables. As a supplement to local combustion/non-combustion contributions, the suggested method intends to comprise and quantify the effect that transboundary PM sources and wind dispersion have, on particulate air pollution levels. The proposed technique was implemented at a background sampling site in Birmingham (United Kingdom) and the results were compared with the outcome of a Simple Linear Regression Model (SLRM) which contained only one predictor variable expressing local combustion. Various statistical indices signified the upgraded performance of the MLRM, in comparison with SLRM, thus the participation of long range transport and wind dispersion

variables in the MLRM was successful. According to the MLRM's findings, anthropogenic combustion (traffic, heating) is the strongest source of PM_{10} in the selected background urban area, followed by local non-combustion emissions and long range transport. Extreme $PM_{2.5}$ intrusions from continental Europe also emerged.

Keywords: PM_{10}; MODIS; Aerosol Optical Depth; Wind dispersion; Multiple Linear Regression.

Effect of Butanol Blends on Nano Particle Emissions from a Stationary Conventional Diesel Engine

Mohit Raj Saxena, Rakesh Kumar Maurya

Source: *Volume 16, No. 9, September 2016, Pages 2255-2266 doi:10.4209/aaqr.2016.04.0144*

In this study, combustion characteristics and nano-size soot particle emissions from a stationary conventional diesel engine have been experimentally investigated using butanol/diesel blends. Experiments were conducted on a single cylinder stationary diesel engine at a constant speed of 1500 rpm for neat diesel and butanol/diesel blends (i.e., 10%, 20% and 30% butanol on volume basis) at different engine loads. Piezoelectric pressure transducer installed in the engine combustion chamber was used for measuring cylinder pressure data. In-cylinder pressure data for 2000 consecutive engine cycles was recorded and averaged data was used for the analysis of combustion characteristics. Butanol/diesel blends show higher rate of heat release in comparison to neat diesel and heat release rate increases with butanol percentage in the blend. Opacity meter and exhaust particle sizer were used for analyzing smoke opacity, size and mass distributions of soot particles respectively at different engine operating conditions. Soot particle distribution from 5 nm to 1000 nm was recorded at each test condition. Results show that total particle concentration decreases with an increase in engine operating loads. It was found that butanol/diesel blends have lower total particulate concentration and the surface area.

Keywords: Particulate matter; Smoke opacity; Combustion; Butanol; Diesel.

Phthalate Ester Concentrations, Sources, and Risks in the Ambient Air of Tianjin, China

Zhenyu Zhu, Yaqin Ji, Shijian Zhang, Jingbo Zhao, Jie Zhao

Source: *Volume 16, No. 9, September 2016, Pages 2294-2301 doi:10.4209/aaqr.2015.07.0473*

Phthalate esters (PAEs) are excessively used in industries, which have posed a serious threat to human health. In this study, PAEs concentrations, sources, and risks in the ambient air of Tianjin, China were studied. The average concentrations of dimethyl phthalate (DMP), diethyl phthalate (DEP), di-n-butyl phthalate (DBP), butylbenzyl phthalate (BBP), di (2-ethylhexyl) phthalate (DEHP), and di-n-octyl phthalate (DOP) in PM_{10} were 0.18, 0.08, 2.24, 0.02, 10.79, and 0.10 ng m^{-3}, respectively; in $PM_{2.5}$ the average concentrations were 0.14, 0.07, 1.93, 0.02, 6.26, and 0.05 ng m^{-3}, respectively. DEHP and DBP were the predominant compounds identified, and the concentrations of PAEs in summer were significantly higher than those in winter. A coefficient of divergence analysis indicated that the PAE composition profiles in PM_{10} and $PM_{2.5}$ were significantly different at the HD (Hedong) and HB (Hebei) sites; however, they were similar to each other at the NK (Nankai) and TT (Tieta) sites. A principal component analysis indicated that the emissions from cosmetics, personal care products, industrial processes, plasticizers, and medical devices may be major sources of PAEs in ambient PM in Tianjin. The daily intake (DI) of six PAEs from ambient air inhalation in Tianjin was estimated for five age groups. The results showed that the highest exposure dose was DMP in all age groups. Infants experienced the highest total DI of all six PAEs, whereas adults experienced the lowest total DI. The cancer risks from ambient air inhalation exposure were also estimated on the basis of DEHP concentrations. Although the risks for all population groups were below the U.S. Environmental Protection Agency threshold of 10^{-6}, our result underestimates the actual health risk because only ambient air inhalation exposure to DEHP was considered in this study. Additional studies are necessary to investigate the effects of long-term exposure to air pollution in Tianjin.

Keywords: Phthalate esters; Particulate matter; Principal component analysis; The cancer risks.

Long-Range Correlations in Air Quality Time Series: Effect of Differencing and Shuffling

Asha B. Chelani

Source: *Volume 16, No. 9, September 2016, Pages 2302-2313 doi:10.4209/aaqr.2016.04.0139*

Long-range correlations in the air quality index (AQI) are analysed using rescaled range analysis (*R/S*), detrended fluctuation analysis (*DFA*) and power spectral density analysis. Air quality index in five cities of India is considered for this purpose. Statistical transformations such as differencing and shuffling have been carried out to examine the effect of temporal correlations on long-range correlation property of the time series. All three methods indicated the presence of persistence in original AQI time series. After differencing, long-range correlation property is, however, observed to be distorted. *R/S* analysis did not show the similar results as *DFA* and power spectral density analysis. Shuffled time series is shown to possess persistence as in the original one by using *R/S* analysis, whereas other two methods showed random behaviour at most of the locations. This suggests that the persistence property is largely influenced by short-range correlations in the AQI time series. The incorporation of this information can enhance the performance of the models to forecast the air quality. The similarity in the results of *DFA* and power spectral density analysis suggests that both methods can be relied more than *R/S* analysis in studying the persistence property of the time series.

Keywords: Long-range correlations; Differencing; Shuffling; Air quality index.

Chemical Compositions of PM2.5 at Two Non-Urban Sites from the Polluted Region in Europe

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Source: *Volume 16, No. 10, October 2016, Pages 2333-2348 doi: 10.4209/aaqr.2015.09.0538*

The study presents the analysis of measurement results for the ambient mass concentrations of fine particulate matter ($PM_{2.5}$), $PM_{2.5}$-bound carbonaceous matter (OC, EC) and water-soluble ions (Cl^-, NO_3^-, SO_4^{2-}, Na+, NH_4^+, K^+, Ca^{2+}, Mg^{2+}). The 24-h $PM_{2.5}$ samples were collected in the heating and non-heating seasons at two regional background sites in Southern Poland in 2011–2013. The percentage of the secondary organic and inorganic matter in $PM_{2.5}$ was calculated. Over the whole measurement period, the mean $PM_{2.5}$ concentration was 31.56 μg m^{-3} and 24.92 μg m^{-3} in Racibórz and Złoty Potok, respectively. Regardless of the season, the total carbon percentage in $PM_{2.5}$ was comparable at both sites and amounted ~40%. There were no visible seasonal variations in the secondary organic carbon (SOC) share in $PM_{2.5}$. The mean percentage of the primary organic carbon (POC) in $PM_{2.5}$ was higher than the SOC percentage at both locations. The mean contribution of the water-soluble ions in the $PM_{2.5}$ mass was lower than the TC percentage, with values 20.35% (Złoty Potok) and 33.56% (Racibórz). The total share of the secondary ions (SO_4^{2-}, NO_3^- and NH_4^+) in $PM_{2.5}$ was comparable in both measurement periods. It was shown that $PM_{2.5}$ at regional

background sites in Southern Poland is significantly different than at similar stations across Europe. It is reflected by higher concentrations of $PM_{2.5}$ and its main components and lower percentage of the secondary ions in the $PM_{2.5}$ mass. The carbonaceous matter percentage in $PM_{2.5}$ is higher than in other parts of Europe.

Keywords: $PM_{2.5}$; Organic carbon; Secondary ions; Regional background sites; Southern Poland.

Characterization of Size-Fractionated Particulate Matter and Deposition Fractions in Human Respiratory System in a Typical African City: Nairobi, Kenya

Samuel Mwaniki Gaita, Johan Boman, Michael James Gatari, Annemarie Wagner, Sara Kluge Jonsson

Source: *Volume 16, No. 10, October 2016, Pages 2378-2385 doi: 10.4209/aaqr.2016.01.0019*

Information from elemental and mass composition of size-fractionated airborne particle matter (PM) provides insightful knowledge about their impact on human health, meteorology and climate. To attain insight into the nature of size-fractionated PM from a typical African city, samples were collected from an urban background site in Nairobi, Kenya, during the months of August and September in 2007. PM samples ranging in size from 0.06 to 16 μm aerodynamic diameter were collected on pre-weighed polycarbonate filters with 0.4 μm pore size using a nine-stage cascade impactor. Particles less than 0.06 μm were collected on a backup filter. A total of 170 samples were collected and analysed for trace elements using the Proton Induced X-Ray Emission (PIXE) technique. The analysis showed that Si, Fe and S dominated in all size ranges and displayed unimodal mass-size distribution whereas K, Cu, Zn and Pb, depicted bimodal mass-size distribution highlighting the multiplicity of their sources. To estimate human exposure to PM, deposition fractions of both the coarse and fine PM in the human respiratory system were calculated. The deposited concentration was found to be highest in the head airways region compared to the tracheobronchial and pulmonary regions.

Keywords: Urban air quality; Human health; Particulate pollution.

Characteristics and Relationships between Indoor and Outdoor PM2.5 in Beijing: A Residential Apartment Case Study

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Source: *Volume 16, No. 10, October 2016, Pages 2386-2395 doi: 10.4209/aaqr.2015.12.0682*

In order to understand the characteristics and relationships between indoor and outdoor $PM_{2.5}$ during the heating period of 2014 in Beijing, the investigation of $PM_{2.5}$ and associated species including organic and elemental carbon (OC/EC), water soluble ions, metal elements and trace organic matter (OM) were undertaken at a residential apartment. The average $PM_{2.5}$ concentration was 55.2 ± 47.3 μg m^{-3} for indoor and 100.4 ± 82.1 μg m^{-3} for outdoor, and the indoor $PM_{2.5}$ was found to be mainly from outdoors. OM and $(NH_4)_2SO_4$ were the dominated components of $PM_{2.5}$, accounted for 71.5% in indoor $PM_{2.5}$ and 52.4% in outdoor $PM_{2.5}$, followed by fine soil and NH_4NO_3 (23.7% and 27.9%). The polycyclic aromatic hydrocarbons (PAHs) concentration was 187.3 ng m^{-3} and 387.0 ng m^{-3}, and the phthalic acid esters (PAEs) concentration reached 1054.2 ng m^{-3} and 515.3 ng m^{-3}, for indoor and outdoor, respectively. Hexachlorobenzene (HCB) only existed indoors (5.5 ng m^{-3}). HCB and most PAEs in indoor $PM_{2.5}$ were dominated by indoor sources whereas other species were greatly influenced by outdoor sources especially during the pollution period.

Keywords: Indoor/outdoor $PM_{2.5}$; Chemical composition; Trace organic matter; Heating period.

Vertical Circulation of Atmospheric Pollutants near Mountains during a Southern California Ozone Episode

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Source: *Volume 16, No. 10, October 2016, Pages 2396-2404 doi: 10.4209/aaqr.2015.09.0554*

This study investigates the air pollutant interactions and emission source contributions to ozone (O_3) formation within a complex terrain. Air quality simulations using the Community Multiscale Air Quality (CMAQ) Model focused on vertical distributions of O_3 for the July 14–18, 2005 episode in the South Coast Air Basin (SoCAB). The Zero-Out method was applied in sensitivity tests for seven emission source categories. Elevated O_3 concentrations were found near the top of the planetary

boundary layer (PBL, ~1200 m) and in the free troposphere (~3500 m) over the eastern SoCAB. Low O_3 concentrations were found near the surface at the center of the basin due to nitrogen oxide (NO) titration by fresh vehicle exhaust. Sea and land breezes, enhanced by up-slope flows (the "mountain chimney effect") transported O_3 upward. Formation of O_3 is sensitive to the H_2O_2/HNO_3 ratio, depending on fresh vs. aged pollutant mixtures. Biogenic emissions were important contributors to O_3 formation, both in the SoCAB and at the top of the PBL. In contrast, the highest vehicle contributions to O_3 were found far from urban areas and in the lower free troposphere. Vertical cross-sectional analysis provided some insights into the O_3 formation and mixing processes present in the SoCAB.

Keywords: Ozone formation; Source contribution; Vertical circulation; Model simulation.

Aromatic VOCs at Major Road Junctions of a Metropolis in India: Measurements Using TD-GC-FID and PTR-TOF-MS Instruments

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Source: *Volume 16, No. 10, October 2016, Pages 2405-2420 doi: 10.4209/aaqr.2015.11.0643*

Ambient mass concentrations of benzene and toluene were measured at 12 different road junctions of Ahmedabad city in India during the pre-monsoon season of year 2015. A Thermal Desorption-Gas Chromatography-Flame Ionization Detector (TD-GC-FID) technique was used for the analysis of aromatic volatile organic compounds (VOCs) in air samples. In each of both inner and outer ring roads, air samples were collected at 6 sites to investigate the spatial variation of benzene and toluene. The mass concentrations of benzene and toluene show strong site-to-site and day-to-day variations. The average mass concentration of benzene varied in the ranges of 11–35 μg m^{-3} and 4–12 μg m^{-3}along the inner and outer roads, respectively. The mass concentration of toluene varied in the ranges of 43–142 μg m^{-3} and 11–28 μg m^{-3}along the inner and outer roads, respectively. Overall, the mass concentrations of VOCs along the inner road were 3–5 times higher than those measured along the outer road.The mass concentrations of benzene and toluene show good correlation suggesting their common emission sources (mostly vehicular). However, the enhancement ratios of ΔToluene/ΔBenzene (~4.0 μg μg^{-1}) along both the roads were higher than the typical ratios (1.5–3.5 μg μg^{-1}) reported for vehicular emissions. The higher values of ΔToluene/ΔBenzene are due to the emissions of VOCs also from industrial and other non-traffic sources. During the daytime, the lower mass concentrations of VOCs and lower ΔToluene/ΔBenzene (~2 μg μg^{-1}) indicate the role of photochemical aging. The combined diurnal trend of ΔToluene/ΔBenzene agrees well with that measured at central Ahmedabad using the proton-transfer-reaction time of flight mass spectrometer (PTR-TOF-MS). However, compared to weekdays, the mass concentrations of VOCs

show reduction and increase during the Sunday and Saturday, respectively. The mass concentration of VOCs and their ratio were towards the higher side of data reported for different urban sites of the world.

Keywords: Aromatic VOCs; India; TD-GC-FID; Urban; Traffic.

Loading Rates of Dust and Metals in Residential Houses of Arid and Dry Climatic Regions

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Source: *Volume 16, No. 10, October 2016, Pages 2462-2473 doi: 10.4209/aaqr.2015.10.0611*

Dust samples were collected from 38 naturally ventilated houses for 12 weeks. Effects of three variables in two groups each were evaluated: proximity to traffic density (main- and side-roads), cigarettes smoking (smoking and no-smoking), and houses' age (old and new). No significant differences were identified between the two groups for all variables (p = 0.227–0.247). The average dust loading rate for the entire group was 66.7 ± 30.9 mg m^{-2} $week^{-1}$. The average metal concentrations (μg g^{-1}) for the entire group were 58.7 ± 17.4 for V, 53.8 ± 12.7 (Cr), 473 ± 137 (Mn), 9.68 ± 2.83 (Co), 130 ± 52.1 (Cu), 241 ± 65.3 (Sr), 0.827 ± 0.552 (Cd), 324 ± 143 (Ba), and 58.9 ± 28.9 for Pb. Likewise, the average metal loading rates (μg m^{-2} $week^{-1}$) for the entire group were: 4.01 ± 2.41 for V, 3.62 ± 1.97 (Cr), 31.9 ± 18.3 (Mn), 0.662 ± 0.387 (Co), 8.57 ± 5.30 (Cu), 16.3 ± 9.23 (Sr), 0.051 ± 0.034 (Cd), 21.1 ± 12.4 (Ba), and 3.97 ± 2.74 for Pb. We noticed enrichment factors (EF) of less than 2 and strong correlations between V, Cr, Mn, Co, and Sr indicating their crustal origin. Conversely, Pb, Cu, and Cd showed low to moderate correlations together with moderate to significant EF suggesting anthropogenic pollution of non-crustal origins. Despite the scarcity of rain fall and arid environment in the studied area, our dust and metal loading rates can be considered as intermediate when compared to some international cities. Such a finding could be attributed to the absence of major industries and the relatively low traffic density in our study area.

Keywords: Indoor dust; Dust loading rates; Metals in indoor dust; Almadinah Almunawarah.

Emission Scenarios and Health Impacts of Air Pollutants in Goa

Sumit Sharma, Kavita Vithal Patil

Source: *Volume 16, No. 10, October 2016, Pages 2474-2487 doi: 10.4209/aaqr.2015.12.0664*

This paper presents the first emission inventory for the highly urbanized state of Goa in India, which is an important international tourist destination and also accommodates significant industrial activities, including mining. The observed concentrations of the pollutants like $PM_{10}$and $PM_{2.5}$ show violations at many locations. Sectoral inventories prepared in this study depicts mining (38%), industries (24%), and transport (10% tail-pipe and 15% road dust) as the major contributors to the PM_{10} emissions in the state. Higher emissions intensity is observed in heavily populated and industrialized coastal taluks and mining dominated taluks. Emissions are projected for the future (2030) under two different scenarios (business as usual and alternative) to assess future air quality and impacts. The grid-wise emissions under these future scenarios are fed into an air quality model, to estimate spatial distribution of PM_{10} concentrations in Goa. The model results are validated with actual observations. Thereafter, the grid-wise PM_{10} concentrations are overlaid on the population to compute its health impacts using established dose response functions. The study shows that PM_{10} accounts for 2.6% of mortalities in Goa, which are expected to go up further in a business as usual scenario. Alternative strategies which show reduction in pollution and associated health impacts in the region are evaluated. Based on the alternative scenario, key recommendations are made for air quality improvement in the state.

Keywords: Emission inventory; Air quality modelling; Health impacts; Goa.

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Aerosols Size Distribution Characteristics and Role of Precipitation during Dust Storm Formation over Saudi Arabia

Ashraf Farahat, Hesham El-Askary, A. Umran Dogan

Source: *Volume 16, No. 10, October 2016, Pages 2523-2534 doi: 10.4209/aaqr.2015.11.0656*

Kingdom of Saudi Arabia and the Gulf region are frequently exposed to major dust storms and anthropogenic emissions from rapidly growing industrial activities that affect aerosols optical and physical characteristics. This paper integrates observations from space-borne sensors namely MODIS and CALIPSO, together with AERONET ground observations to examine eight years aerosols characteristics during the (March–May) season of 2003 to 2010 over Saudi Arabia. Aerosol analysis

from the interdependent data assessment show comparable aerosols characteristics over the eight year period with higher aerosols mean optical depths over enhanced dust load region, (46–50°E, 25–29°N), during March–May of 2009 and 2010. The mean angstrom exponent during March–May 2003 to 2008 was found ~17% higher than the same period during 2009. The major dust storm on March 9 and 10, 2009 could have an effect on the coarse mode particles increment during 2009. Over the eight years the highest angstrom exponent was observed on 2004 suggesting dominance of fine-mode particles, whereas a declination in the angstrom exponent values is observed during 2005, 2006, 2007, and 2008. The aerosols size distribution measured by sunphotometer indicates a maximum value of ~ 47% higher in 2009 compared to 2010 suggesting the domination of coarse mode particles in 2009. Using the CALIPSO volume depolarization ratio, a possible mixing of anthropogenic aerosols with dust was observed during March–May of 2009 and 2010 featured by coarse particles domination and high percentage of fine particles during 2009. The effect of precipitation prior to dust storms on dust loading was investigated. Our observation suggests a possible impact of the varying precipitation rate prior to dust storms outbreak and the actual dust loading during dust events.

Keywords: Atmospheric aerosols; Remote Sensing; AERONET; Dust; Pollution.

Classification of Aerosols in an Urban Environment on the Basis of Optical Measurements

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Source: *Volume 16, No. 10, October 2016, Pages 2535-2549 doi: 10.4209/aaqr.2016.06.0219*

The present study investigates various types of aerosol in the Lahore city of Pakistan using Aerosol Robotic Network (AERONET) data over a six year period from 2007 to 2012. Aerosol optical depths (AODs) observed was in the range 0.2–1.12. An analysis of seasonal variations in AOD has indicated that the highest AOD values occurred in summer and the lowest in winter. The urban aerosols of the study area were classified on the basis of optical parameters such as AOD, Extinction Angstrom Exponent (EAE), Absorption Angstrom Exponent (AAE), Single Scattering Albedo (SSA), Asymmetry Parameter (ASY) and Refractive Index (RI). The AAE values were in the range from 0.25 to 3.2. Real Refractive Index (RRI) and Imaginary part of the Refractive Index (IRI) values were in the range from 1.5 to 1.6 and 0 to 0.005 respectively. The major contributions to the atmospheric aerosols over Lahore were from urban industrial emissions, fossil fuel burning and road/soil dust. Higher RRI values reflected larger re-suspended road dust particles and long-range transported particles, while lower values reflected increased anthropogenic absorbing carbonaceous aerosols over the area. The AERONET retrieved SSA (0.80–0.89) and ASY (0.70–0.83) values suggested a

predominance of urban industrial, vehicular and dust aerosols over Lahore. The Derivative of Angstrom Exponent (DAE) was derived at a wavelength of 500 nm and was found to indicate a predominance of fine aerosols across all seasons, particularly during summer and autumn seasons. Back-trajectory analyses using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model revealed that the major air masses over Lahore originated from India, Iran and Afghanistan.

Keywords: Aerosol; AERONET; Classification; AOD; SSA; DAE.

Indoor Air Quality Management by Combined Ventilation and Air Cleaning: An Experimental Study

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Source: *Volume 16, No. 10, October 2016, Pages 2550-2559 doi: 10.4209/aaqr.2015.10.0577*

A combination of ventilation and air cleaning regimes were investigated for the removal of aerosol particles and volatile organic compounds (VOCs) in a test chamber, representing a typical room. A series of portable multi-staged air cleaner efficiency tests were performed examining tobacco smoke as a source of pollution. Portable indoor air cleaners were effective in removing of particles, reaching up to 97% removal efficiency based on particle number concentrations after 30 minutes, while Clean Air Delivery Rate ($CADR_{PNC}$) varied from 37 ± 4 to 237 ± 11 $m^3\ h^{-1}$. The removal of VOCs varied from 21.4 to 45.7% and $CADR_{VOC}$ ranged from 2.2 ± 0.3 to 29.9 ± 2.8 $m^3\ h^{-1}$, indicating substantially lower efficiency. The combination of ventilation and air cleaning provided different responses with respect to pollutant removal and energy efficiency. The air cleaning was the most efficient for removing particulate matter from indoor air, minimizing the requirement for ventilation. On the other hand, the ventilation seemed to be more efficient in the removal of VOCs, while the combination of ventilation and air cleaning increased pollutant removal efficiency by 20% and maximized the energy efficiency.

Keywords: Indoor air quality; Air cleaning; Ventilation; Particulate matter; Volatile organic compounds.

Satellite-Surface Perspectives of Air Quality and Aerosol-Cloud Effects on the Environment: An Overview of 7-SEAS/BASELInE

Si-Chee Tsay, Hal B. Maring, Neng-Huei Lin,Sumaman Buntoung, Somporn Chantara, Hsiao-Chi Chuang, Philip M. Gabriel, Colby S. Goodloe, Brent N. Holben, Ta-Chih Hsiao, N. Christina Hsu, Serm Janjai, William K.M. Lau, Chung-Te Lee, Jaehwa Lee, Adrian M. Loftus, Anh X. Nguyen, Cuong M. Nguyen, Shantanu K. Pani, Peter Pantina, Andrew M. Sayer, Wei-Kuo Tao, Sheng-Hsiang Wang, Ellsworth J. Welton, Wan Wiriya, Ming-Cheng Yen

Source: *Volume 16, No. 11, November 2016, Pages 2581-2602doi:10.4209/aaqr.2016.08.0350*

The objectives of 7-SEAS/BASELInE (Seven SouthEast Asian Studies/Biomass-burning Aerosols & Stratocumulus Environment: Lifecycles & Interactions Experiment) campaigns in spring 2013–2015 were to synergize measurements from uniquely distributed ground-based networks (e.g., AERONET, MPLNET) and sophisticated platforms (e.g., SMARTLabs, regional contributing instruments), along with satellite observations/retrievals and regional atmospheric transport/chemical models to establish a critically needed database, and to advance our understanding of biomass-burning aerosols and trace gases in Southeast Asia (SEA). We present a satellite-surface perspective of 7-SEAS/BASELInE and highlight scientific findings concerning: (1) regional meteorology of moisture fields conducive to the production and maintenance of low-level stratiform clouds over land, (2) atmospheric composition in a biomass-burning environment, particularly tracers/markers to serve as important indicators for assessing the state and evolution of atmospheric constituents, (3) applications of remote sensing to air quality and impact on radiative energetics, examining the effect of diurnal variability of boundary-layer height on aerosol loading, (4) aerosol hygroscopicity and ground-based cloud radar measurements in aerosol-cloud processes by advanced cloud ensemble models, and (5) implications of air quality, in terms of toxicity of nanoparticles and trace gases, to human health. This volume is the third 7-SEAS special issue (after *Atmospheric Research*, vol. 122, 2013; and *Atmospheric Environment*, vol. 78, 2013) and includes 27 papers published, with emphasis on air quality and aerosol-cloud effects on the environment. BASELInE observations of stratiform clouds over SEA are unique, such clouds are embedded in a heavy aerosol-laden environment and feature characteristically greater stability over land than over ocean, with minimal radar surface clutter at a high vertical spatial resolution. To facilitate an improved understanding of regional aerosol-cloud effects, we envision that future *BASELInE-like* measurement/modeling needs fall into two categories: (1) efficient yet critical *in-situ*profiling of the boundary layer for validating remote-sensing/retrievals and for initializing regional transport/chemical and cloud ensemble models, and (2) fully utilizing the high observing frequencies of geostationary satellites for resolving the diurnal cycle of the boundary-layer height as it affects the loading of biomass-burning aerosols, air quality and radiative energetics.

Keywords: 7-SEAS; BASELInE; Biomass-burning; Air Quality; Aerosol; Cloud.

Aerosol Chemical Profile of Near-Source Biomass Burning Smoke in Sonla, Vietnam during 7-SEAS Campaigns in 2012 and 2013

Chung-Te Lee, Shidharth Sankar Ram, Dac Loc Nguyen, Charles C.K. Chou, Shih-Yu Chang, Neng-Huei Lin, Shuenn-Chin Chang, Ta-Chih Hsiao, Guey-Rong Sheu, Chang-Feng Ou-Yang, Kai Hsien Chi, Sheng-Hsiang Wang4, Xue-Chang Wu

Source: *Volume 16, No. 11, November 2016, Pages 2603-2617doi:10.4209/aaqr.2015.07.0465*

This study aimed to investigate aerosol chemical characteristics and to obtain the chemical profile of near-source biomass burning (BB) aerosols at a site (675 m a.s.l.) in Sonla, Northern Vietnam. Particulate matter (PM) with an aerodynamic diameter less than or equal to 2.5 μm ($PM_{2.5}$) was collected over a 24 h sampling period as part of the Seven South East Asian Studies (7-SEAS) campaign. The studies were conducted when BB was highly active — that is, in the spring of 2012 and 2013. The collected particles were analyzed for carbonaceous fractions and water-soluble components, in addition to the mass concentration. Data obtained were further analyzed to determine the stable species profile by classifying the 5-day air-mass backward trajectories. The average $PM_{2.5}$ mass concentrations were 51 ± 19 μg m^{-3} and 57 ± 27 μg m^{-3} in 2012 and 2013, respectively. Carbonaceous contents dominated BB aerosol, with 59% ± 9% and 58% ± 9% in organic carbon (OC) and 9% ± 3% and 10% ± 3% in elemental carbon (EC) of $PM_{2.5}$ in 2012 and 2013, respectively. Of the 8 carbonaceous fractions analyzed thermo-optically for $PM_{2.5}$, OC3 (evolution temperature at 280°C–480°C) was most abundant in OC fractions, and EC1-OP (elemental carbon evolved at 580°C minus the pyrolized OC fractions) was predominant in EC fractions in most occasions. Among the measured water-soluble inorganic ions, NH_4^+ and SO_4^{2-} widely varied, indicating the influence of different trajectory origins. This finding was confirmed by trajectory classification of aerosol data. The trajectories were also distinguished with respect to char-EC to soot-EC ratio, and water-soluble OC. These characteristics were highest in the trajectory from the BB source area.

Keywords: Biomass burning; Near-source aerosol chemical profile; Carbonaceous contents; Water-soluble inorganic ions; Water-soluble organic carbon.

Comprehensive PM2.5 Organic Molecular Composition and Stable Carbon Isotope Ratios at Sonla, Vietnam: Fingerprint of Biomass Burning Components

Dac Loc Nguyen, Kimitaka Kawamura, Kaori Ono, Shidharth Sankar Ram, Guenter Engling, Chung-Te Lee1, Neng-Huei Lin, Shuenn-Chin Chang, Ming-Tung Chuang, Ta-Chih Hsiao, Guey-Rong Sheu, Chang-Feng Ou-Yang, Kai Hsien Chi, Shao-An Sun

Source:*Volume 16, No. 11, November 2016, Pages 2618-2634 doi:10.4209/aaqr.2015.07.0459*

This study presents measurements of aerosol chemical properties at Sonla, northern Vietnam (675 m a.s.l.) during spring time, when biomass burning (BB) was very active in the northern Indochina Peninsula, as part of the 7-SEAS (Seven South East Asian Studies) campaign in 2013. The gas chromatography-mass spectrometry (GC-MS) analysis of BB markers in 14 selected samples indicated that mixed softwood, hardwood, grass, and non-woody vegetation were burned. More than 50 organic compounds including levoglucosan, lignin and resin products, sugar and sugar alcohol compounds, fatty acids, phthalate esters, aromatic acids, poly-acids, and biogenic oxidation products (e.g., 2-methyltetrols, alkene triols, 3-hydroxyglutaric acid) were measured in $PM_{2.5}$. Levoglucosan, a BB tracer, was the predominant species among aerosol sugars, with an average concentration of 1.62 ± 0.89 μg m^{-3}, comprising 2.23 ± 0.5% of $PM_{2.5}$ mass. For the collection period of the selected samples, backward air mass trajectories were classified into the source regions of Indochina (BBIC), southern China (BBSC), and the South China Sea (BBSS). All resolved molecular compounds show their dominance on the trajectory from BBIC, verifying the BB smoke origin of that region. Trajectory classification provides additional information, such as higher level of diethyl phthalate associated with BBSC trajectory, revealing urban or industrial influence, and more low-molecular-weight than high-molecular-weight fatty acids, indicating distributions with more microbial and lesser plant wax/vegetation burning contributions along the BBSC trajectory. In addition, we report, for the first time, stable carbon isotopic data ($\delta^{13}C$) for $PM_{2.5}$ aerosols in northern Vietnam, which ranged from –26.6 to –25.4‰ in $PM_{2.5}$, indicating contributions from burning of C_3 plants and fossil fuel combustion.

Keywords: Indochina; Fine aerosol particles; Biomass burning; Organic molecular markers; $\delta^{13}C$ isotope.

Investigation of Biomass Burning Chemical Components over Northern Southeast Asia during 7-SEAS/BASELInE 2014 Campaign

Chanakarn Khamkaew, Somporn Chantara, Rungruang Janta, Shantanu Kumar Pani, Tippawan Prapamontol, Sawaeng Kawichai, Wan Wiriya, Neng-Huei Lin

Source: *Volume 16, No. 11, November 2016, Pages 2655-2670doi:10.4209/aaqr.2016.03.0105*

This study investigates the chemical components of biomass burning (BB) aerosols obtained from Doi Ang Khang (DAK; near BB source) and Chiang Mai University (CMU; an urban location) over northern Southeast Asia in dry season (March to mid-April) 2014. $PM_{2.5}$ (particulate matter with an aerodynamic diameter less than or equal to 2.5 μm) samples were collected over a 24-h sampling period as a part of the Seven South East Asian Studies (7-SEAS)/BASELInE (BB Aerosols & Stratocumulus Environment: Lifecycles & Interactions Experiment) campaign. The collected aerosols were analyzed for mass concentrations of ions, metals and levoglucosan. The influence of air mass movements on aerosol species was also analyzed. The average $PM_{2.5}$ mass concentrations at DAK (80.8–83.3 μg m^{-3}) and CMU (90.7–93.1 μg m^{-3}) were not significantly different ($p > 0.05$) and well correlated ($r = 0.8$), and likely originated from similar source origins. The number of fire hotspots was particularly high during 20–21 March (greater than 200) and, consequently, peaks of $PM_{2.5}$ were recorded at both sites. The most abundant elements at both sampling sites were K (49–50% of total elements), Al (26–31%), Mg (16%) and Zn (4–7%), whereas SO_4^{2-} (30–38% of total ions), NO_3^- (13–20%), Na^+ (16–20%) and NH_4^+ (14–15%) were the most abundant ions. Concentrations of levoglucosan and K^+ (BB tracers) were well correlated ($r = 0.5$ for CMU and 0.7 for DAK) confirming that the $PM_{2.5}$ detected in these areas were mainly influenced by BB activity. Principal component analysis (PCA) revealed that BB, road traffic, agricultural activity and soil re-suspension were plausible sources of $PM_{2.5}$ over the study locations. Apart from local sources, the influence of long-range transport was also investigated by way of three-day backward trajectory analysis.

Keywords: $PM_{2.5}$; Biomass burning; Chemical composition; Air pollution; Aerosol.

A Case Study of PM2.5 Characterization in Bangi, Selangor, Malaysia during the Southwest Monsoon Season

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Source: *Volume 16, No. 11, November 2016, Pages 2685-2691doi:10.4209/aaqr.2015.04.0277*

A case study was carried out to characterize the ambient $PM_{2.5}$ based on ground-based sampling in Bangi, Selangor, Malaysia in September, 2013 during the southwest monsoon season. We determined the total mass concentration, organic carbon, elemental carbon (EC), and metals in $PM_{2.5}$ samples. The mean $PM_{2.5}$ mass concentration was 44.5 μg m^{-3}, showing that it exceeded the national air quality standard of 35 μg m^{-3} for 24-hour $PM_{2.5}$ by the U.S. Environmental Protection Agency. Relatively high OC and EC concentrations of this study compared to those of other Southeast Asian countries were observed, which indicate that significant sources of OC and EC exist. The results of char-EC/soot-EC ratios strongly suggest that biomass burning is the main contributor to ambient EC concentrations compared to coal combustion and motor vehicle emissions. From calculations using the mass closure model, organic matter was the most abundant component in $PM_{2.5}$ mass at 22.4 ± 6.65 μg m^{-3}, followed by nss-sulfate at 4.84 ± 2.49 μg m^{-3}, and EC at 4.11 ± 0.916 μg m^{-3}. This result indicates that targeting the sources of carbonaceous $PM_{2.5}$ is a crucial step to improve the air quality in this study field.

Keywords: $PM_{2.5}$; OC; EC; Malaysia.

Correlation Analysis, Transportation Mode of Atmospheric Mercury and Criteria Air Pollutants, with Meteorological Parameters at Two Remote Sites of Mountain and Offshore Island in Asia

Wang-Kun Chen, Tsung-Chang Li, Guey-Rong Sheu, Neng-Huei Lin, Liang-Yu Chen, Chung-Shin Yuan

Source: *Volume 16, No. 11, November 2016, Pages 2692-2705doi:10.4209/aaqr.2015.07.0455*

This study presents a comparison of trace (Hg) and criteria (CO, SO_2, NO_x, O_3 and PM_{10}) air pollutants monitored at two remote sites with the same latitude but different altitude: Mt. Lulin and the Penghu Islands, in Taiwan from 2011 to 2012. A filtering technique was comprehensively applied to distinguish the climatic characteristics of the two remote sites, as well as to determine their discriminant factor. The concentrations of air pollutants monitored at Mt. Lulin were generally lower than those at the Penghu Islands, with the exception of O_3concentration. PM_{10} and NO_x were the important factors that can distinguish two clusters of measurement data at the two remote sites, and a criteria discriminant factor of atmospheric parameters derived from these two air pollutants. For both high- and low-frequency patterns, the concentrations of NO_x and PM_{10} exhibit significant differences between the two remote sites. However, O_3 concentrations showed almost no differences between these two remote sites, implying that the pattern for the formation and transportation of O_3 at these two sites resulted from similar mechanisms. Moreover, atmospheric mercury (TGM) had a very good linear correlation with CO. The diurnal variation of Hg concentration was dramatic at the Penghu Islands, while it appeared as low as the North

Hemisphere background mercury concentration at Mt. Lulin, indicating that they were not formed via the mechanism modes. This study thus proposed "scenario mercury" and "background mercury" for interpreting this interesting phenomenon.

Keywords: Remote sites; Atmospheric mercury; Criteria air pollutants; Meteorological parameters; Correlation and transportation.

Evaluation of Atmospheric PCDD/Fs at Two High-Altitude Stations in Vietnam and Taiwan during Southeast Asia Biomass Burning

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Source: *Volume 16, No. 11, November 2016, Pages 2706-2715doi:10.4209/aaqr.2015.11.0653*

Dioxin and dioxin-like compounds such as polychlorinated dibenzo-*p*-dioxins and polychlorinated dibenzofurans (PCDD/Fs) are persistent organic pollutants (POPs) generated through human activities. In recent times, extreme weather events such as wild fires have significantly affected the remobilization and successive bioavailability of PCDD/Fs. In Seven South East Asian Studies (7-SEAS), a Southeast Asia biomass burning event that influenced the environmental outcome and transport of PCDD/Fs in Taiwan was investigated on the basis of a climate change situation. During the 7-SEAS campaign on 20–28 March, 2011, significantly high levels of atmospheric PCDD/Fs were observed at Lulin mountain in central Taiwan and in the source region of Northern Vietnam (Son La). Measurements indicated that the patterns of variation of atmospheric PCDD/Fs at both locations were similar, but with a time lag of approximately 2 to 3 days. At Mt. Lulin, there was a significant increase of PCDD/F concentrations from 3.69 to 11.1 fg I-TEQ m^{-3} and 3.32 to 19.1 fg I-TEQ m^{-3} to reach their peaks on 23 March and 26 March. In this study, a tracer simulation using the Weather Research and Forecasting model coupled with chemistry was conducted to investigate the effects of the Southeast Asia biomass burning. The combined results of air mass paths simulation and satellite data can be used as evidence supporting the hypothesis that the source of the increasing PCDD/F level is originated from biomass-burning regions in Indochina, particularly Northern Vietnam and Northern Thailand.

Keywords: Dioxin; Biomass burning; Vietnam; Long-range transport.

Emission Profiles of PM10-Bound Polycyclic Aromatic Hydrocarbons from Biomass Burning Determined in Chamber for Assessment of Air Pollutants from Open Burning

Wan Wiriya, Somporn Chantara, Sopittaporn Sillapapiromsuk, Neng-Huei Lin

Source: *Volume 16, No. 11, November 2016, Pages 2716-2727doi:10.4209/aaqr.2015.04.0278*

In order to estimate emission factors (EFs) of air pollutants, three types of biomass (rice straw, maize residue and leaf litter) were collected and burnt in a self-designed stainless steel chamber. The EFs of PM_{10} from biomass burning were leaf litter (1.22 ± 0.29 g $kgdry^{-1}$) > rice straw (0.89 ± 0.25 g $kgdry^{-1}$) > maize residue (0.59 ± 0.13 g $kgdry^{-1}$), while those of PM_{10}-bound polycyclic aromatic hydrocarbons (PAHs) were leaf litter (0.91 ± 0.28 mg $kgdry^{-1}$) > maize residue (0.47 ± 0.11 mg kgdry–1) ~ rice straw (0.46 ± 0.21 mg $kgdry^{-1}$). The results revealed that burning of forest leaf litter emitted higher amounts of particulate pollutants than the agricultural residue burning. New values of diagnostic ratios of some PAHs, including FLA/(FLA + PYR, BaA/(BaA + CHR) and IND/(IND + BPER), were proposed to identify biomass burning sources. Emission rates (ERs) of PM10 and PAHs from biomass burning in Chiang Mai, Thailand were estimated based on the EFs and burning areas recorded in the dry season of 2010 and 2011. The ERs of pollutants from forest burning were found to be much higher than those from agricultural field burning, mainly due to larger burnt areas in the forest. In 2010, PM_{10} was mainly emitted from the forest fire (2,250 tons), followed by crop burning (133 tons) and paddy field burning (66.9 tons). The same trend was found in 2011 but with much lower emission rates. The ERs of PAHs from biomass burning were 1,815 kg in 2010 and 416 kg in 2011. The ERs of PM_{10} and PAHs in 2011 were 77% decreased from those in 2010 due to unusually high precipitation in the dry season, causing relatively smaller burnt areas and lower pollutant emissions from open burning. It is expected that the results from this study will be significant information for regulatory actions of air quality management in the northern part of Thailand.

Keywords: Aerosol chemistry; Air pollution; Biomass burning; PM_{10}.

Investigation of the CCN Activity, BC and UVBC Mass Concentrations of Biomass Burning Aerosols during the 2013 BASELInE Campaign

Ta-Chih Hsiao, Wei-Cheng Ye, Sheng-Hsiang Wang, Si-Chee Tsay, Wei-Nai Chen, Neng-Huei Lin, Chung-Te Lee, Hui-Ming Hung, Ming-Tung Chuang, Somporn Chantara

Source: *Volume 16, No. 11, November 2016, Pages 2742-2756doi:10.4209/aaqr.2015.07.0447*

Biomass-burning (BB) aerosols, acting as cloud condensation nuclei (CCN), can influence cloud microphysical and radiative properties. In this study, we present CCN measured near the BB source regions over northern Southeast Asia (Doi Ang Khang, Thailand) and at downwind receptor areas (Lulin Atmospheric Background Station, Taiwan), focusing exclusively on 13–20 March 2013 as part of 2013 spring campaign of the Seven SouthEast Asian Studies (7-SEAS) intensive observation. One of the campaign's objectives is to characterize BB aerosols serving as CCN in SouthEast Asia (SEA). CCN concentrations were measured by a CCN counter at 5 supersaturation (SS) levels: 0.15%, 0.30%, 0.45%, 0.60%, and 0.75%. In addition, $PM_{2.5}$ and black carbon mass concentrations were analyzed by using a tapered element oscillating microbalance and an aethalometer. It was found the number-size distributions and the characteristics of hygroscopicity (e.g., activation ratio and κ) of BB aerosols in SEA have a strong diurnal pattern, and different behaviors of patterns were characterized under two distinct weather systems. The overall average κ value was low (0.05–0.1) but comparable with previous CCN studies in other BB source regions. Furthermore, a large fraction of UV-absorbing organic material (UVBC) and high Delta-C among BB aerosols were also observed, which suggest the existence of substantial particulate organic matter in fresh BB aerosols. These data provide the most extensive characterization of BB aerosols in SEA until now.

Keywords: Cloud condensation nuclei; Biomass burning aerosol; Long-range transport; Diurnal cycle; Hygroscopicity.

In-Situ and Remotely-Sensed Observations of Biomass Burning Aerosols at Doi Ang Khang, Thailand during 7-SEAS/BASELInE 2015

Andrew M. Sayer, N. Christina Hsu, Ta-Chih Hsiao, Peter Pantina, Ferret Kuo, Chang-Feng Ou-Yang, Brent N. Holben, Serm Janjai, Somporn Chantara, Shen-Hsiang Wang, Adrian M. Loftus, Neng-Huei Lin, Si-Chee Tsay

Source: *Volume 16, No. 11, November 2016, Pages 2786-2801doi:10.4209/aaqr.2015.08.0500*

The spring 2015 deployment of a suite of instrumentation at Doi Ang Khang (DAK) in northwestern Thailand enabled the characterization of air masses containing smoke aerosols from burning predominantly in Myanmar. Aerosol Robotic Network (AERONET) Sun photometer data were used to validate Moderate Resolution Imaging Spectroradiometer (MODIS) Collection 6 'Deep Blue' aerosol optical depth (AOD) retrievals; MODIS Terra and Aqua provided results of similar quality, with correlation coefficients of 0.93–0.94 and similar agreement within expected uncertainties to

global-average performance. Scattering and absorption measurements were used to compare surface and total column aerosol single scatter albedo (SSA); while the two were well-correlated, and showed consistent positive relationships with moisture (increasing SSA through the season as surface relative humidity and total columnar water vapor increased), in-situ surface-level SSA was nevertheless significantly lower by 0.12–0.17. This could be related to vertical heterogeneity and/or instrumental issues. DAK is at ~1,500 m above sea level in heterogeneous terrain, and the resulting strong diurnal variability in planetary boundary layer depth above the site leads to high temporal variability in both surface and column measurements, and acts as a controlling factor to the ratio between surface particulate matter (PM) levels and column AOD. In contrast, while some hygroscopic effects were observed relating to aerosol particle size and Ångström exponent, relative humidity variations appear to be less important for the PM:AOD ratio here.

Keywords: Biomass burning; Aerosol; Remote sensing; In situ; 7-SEAS BASELInE.

Radiative Effect of Springtime Biomass-Burning Aerosols over Northern Indochina during 7-SEAS/BASELInE 2013 Campaign

Shantanu Kumar Pani, Sheng-Hsiang Wang, Neng-Huei Lin, Chung-Te Lee, Si-Chee Tsay, Brent N. Holben, Serm Janjai, Ta-Chih Hsiao, Ming-Tung Chuang, Somporn Chantara

Source: *Volume 16, No. 11, November 2016, Pages 2802-2817doi:10.4209/aaqr.2016.03.0130*

The direct aerosol radiative effects of biomass-burning (BB) aerosols over northern Indochina were estimated by using aerosol properties (physical, chemical, and optical) along with the vertical profile measurements from ground-based measurements with integration of an optical and a radiative transfer model during the Seven South East Asian Studies/Biomass-Burning Aerosols & Stratocumulus Environment: Lifecycles & Interactions Experiment (7-SEAS/BASELInE) conducted in spring 2013. Cluster analysis of backward trajectories showed the air masses arriving at mountainous background site (Doi Ang Khang; 19.93°N, 99.05°E, 1536 m above mean sea level) in northern Indochina, mainly from near-source inland BB activities and being confined in the planetary boundary layer. The PM10 and black carbon (BC) mass were 87 ± 28 and 7 ± 2 μg m^{-3}, respectively. The aerosol optical depth (AOD500) was found to be 0.26–1.13 (0.71 ± 0.24). Finer (fine mode fraction ≈0.95, angstrom-exponent at 440–870 nm ≈1.77) and significantly absorbing aerosols (single-scattering albedo ≈0.89, asymmetry-parameter ≈0.67, and absorption AOD ≈0.1 at 440 nm) dominated over this region. BB aerosols (water soluble and BC) were the main contributor to the aerosol radiative forcing (ARF), while others (water insoluble, sea salt and mineral dust) were negligible mainly due to their low extinction efficiency. BC contributed only 6% to the surface aerosol mass but its contribution to AOD was 12% (2 times higher). The overall mean ARF was –8.0 and –31.4 W m^{-2} at top-of-atmosphere (TOA) and at the surface (SFC), respectively. Likely, ARF due to BC was +10.7 and –18.1 W m^{-2} at TOA and SFC, respectively. BC imposed the heating rate of +1.4 K d^{-1} within the atmosphere and highlighting its pivotal role in modifying the radiation budget. We

propose that to upgrade our knowledge on BB aerosol radiative effects in BB source region, the long-term and extensive field measurements are needed.

Keywords: Biomass-burning; Near-source; Aerosol optical properties; Radiative effects; 7-SEAS.

Relationship between Aerosol Optical Depth and Particulate Matter over Singapore: Effects of Aerosol Vertical Distributions

Boon Ning Chew, James R. Campbell, Edward J. Hyer, Santo V. Salinas, Jeffrey S. Reid, Ellsworth J. Welton, Brent N. Holben, Soo Chin Liew

Source: *Volume 16, No. 11, November 2016, Pages 2818-2830doi:10.4209/aaqr.2015.07.0457*

As part of the Seven Southeast Asian Studies (7SEAS) program, an Aerosol Robotic Network (AERONET) sun photometer and a Micro-Pulse Lidar Network (MPLNET) instrument have been deployed at Singapore to study the regional aerosol environment of the Maritime Continent (MC). In addition, the Navy Aerosol Analysis and Prediction System (NAAPS) is used to model aerosol transport over the region. From 24 September 2009 to 31 March 2011, the relationships between ground-, satellite- and model-based aerosol optical depth (AOD) and particulate matter with aerodynamic equivalent diameters less than 2.5 μm ($PM_{2.5}$) for air quality applications are investigated. When MPLNET-derived aerosol scale heights are applied to normalize AOD for comparison with surface $PM_{2.5}$ data, the empirical relationships are shown to improve with an increased 11%, 10% and 5% in explained variances, for AERONET, MODIS and NAAPS respectively. The ratios of root mean square errors to standard deviations for the relationships also show corresponding improvements of 8%, 6% and 2%. Aerosol scale heights are observed to be bimodal with a mode below and another above the strongly-capped/deep near-surface layer (SCD; 0–1.35 km). Aerosol extinctions within the SCD layer are well-correlated with surface $PM_{2.5}$ concentrations, possibly due to strong vertical mixing in the region.

Keywords: Air pollution; Air quality; Aerosol optical depth.

Evaluating the Height of Biomass Burning Smoke Aerosols Retrieved from Synergistic Use of Multiple Satellite Sensors over Southeast Asia

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Source: *Volume 16, No. 11, November 2016, Pages28312842) doi:10.4209/aaqr.2015.08.0506*

This study evaluates the height of biomass burning smoke aerosols retrieved from a combined use of Visible Infrared Imaging Radiometer Suite (VIIRS), Ozone Mapping and Profiler Suite (OMPS), and Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) observations. The retrieved heights are compared against spaceborne and ground-based lidar measurements during the peak biomass burning season (March and April) over Southeast Asia from 2013 to 2015. Based on the comparison against CALIOP, a quality assurance (QA) procedure is developed. It is found that 74% (81–84%) of the retrieved heights fall within 1 km of CALIOP observations for unfiltered (QA-filtered) data, with root-mean-square error (RMSE) of 1.1 km (0.8–1.0 km). Eliminating the requirement for CALIOP observations from the retrieval process significantly increases the temporal coverage with only a slight decrease in the retrieval accuracy; for best QA data, 64% of data fall within 1 km of CALIOP observations with RMSE of 1.1 km. When compared with Micro-Pulse Lidar Network (MPLNET) measurements deployed at Doi Ang Khang, Thailand, the retrieved heights show RMSE of 1.7 km (1.1 km) for unfiltered (QA-filtered) data for the complete algorithm, and 0.9 km (0.8 km) for the simplified algorithm.

Keywords: Aerosol height; Satellite; Biomass burning; Southeast Asia; 7-SEAS.

Aerosol Climatology over the Bay of Bengal and Arabian Sea Inferred from Space-Borne Radiometers and Lidar Observations

Shani Tiwari, Amit K. Mishra, Abhay K. Singh

Source: *Volume 16, No. 11, November 2016, Pages 2855-2868doi:10.4209/aaqr.2015.06.0406*

Atmospheric aerosols over the oceanic region are very important air pollutant and play a vital role in Earth's radiation budget and climate change. This study presents the aerosol climatology over the Bay of Bengal (BoB) and Arabian sea (AS) using long term (2006–2012) data from space-borne radiometers [Moderate-Resolution Imaging Spectroradiometer (MODIS), Ozone Monitoring Instrument (OMI)] and space-based active lidar onboard Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observation (CALIPSO). AS experiences higher AOD as compared to that over BoB during the study period. A good periodicity along with strong intra-seasonal/annual variability in aerosol loading is also observed over both the study regions. Approximately one month lag is found for maximum aerosol loading period over AS and BoB for almost every year i.e., June–July for AS and May–June for BoB. This lag could be explained by pathway and timing of summer monsoon over the Indian subcontinent. Elevated layers of absorbing dust up to 2–4 km altitudes are observed during the pre-monsoon and monsoon seasons over both the regions. The CALIPSO measurements show strong seasonal heterogeneity in aerosol properties over both the regions, which is well corroborated with MODIS and OMI observations. This significant seasonal heterogeneity in aerosol

loading has been explained by the role of transportation of aerosols from various emission sources using NOAA HYSPLIT back trajectory model at three different altitude levels viz. 500, 1500 and 2500 m height. The possible role of Indian summer monsoon in modulating the aerosol behaviour over AS and BoB is another important aspect of this study that need further analyses using higher spatio-temporal resolution data.

Keywords: MODIS; CALIPSO; AOD; Aerosol Index (AI); Fine Mode Fraction (FMF).

Seasonal Variation of Aerosol Optical Properties in an Urban Site of the Yangtze Delta Region of China

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Source:*Volume 16, No. 11, November 2016, Pages 2884-2896 doi:10.4209/aaqr.2015.05.0350*

The characteristics of aerosol optical properties during 2012 at Hangzhou (30°14′N, 120°10′E), an urban site in the Yangtze Delta Region of China, were analyzed, including aerosol scattering and absorption coefficients, aerosol optical depth (AOD), Ångström exponent (α), single scattering albedo (SSA), and aerosol size distribution. Both aerosol absorption and scattering coefficients were lowest in summer; the highest absorption coefficient was observed in autumn; while the scattering was highest in winter. There were no significant differences in the seasonal average of AOD, with values of approximately 1.0 in spring, autumn and winter, and 0.72 in summer. The averaged Ångström exponent was found to be relatively high throughout the year, with a minimum value of 1.14 in spring and a maximum of 1.35 in autumn. The fine modes of the aerosol volume size distributions showed the highest peak around radius 0.15 μm in spring, autumn and winter, and radius 0.25 μm in summer, while the coarse modes showed maximum peaks at radius 2.9 μm in all seasons. The volume concentrations of coarse mode aerosols over Hangzhou were highest in spring compared with other seasons. The mean total SSA values were 0.89, 0.93, 0.89 and 0.88 at 440 nm for spring, summer, autumn and winter, respectively, indicating that the aerosols were mainly composed of scattering particles in Hangzhou. Furthermore, it was found continuous pollution events appeared in East China as a result of biomass burning during June 10–13 in 2012. The main causes of extremely severe air pollution and poor visibility in Hangzhou were related to the long-distance transport of pollutants, local pollution emission and stagnant weather conditions. Aerosol optical properties exhibited high value in absorption AOD and high aerosol volumes, low SSA in coarse mode after biomass burning.

Keywords: Aerosol optical properties; Pollution processes; Hangzhou.

Characterization of Particulate Matter Profiling and Alveolar Deposition from Biomass Burning in Northern Thailand: The 7-SEAS Study

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Source:*Volume 16, No. 11, November 2016, Pages 2897-2906 doi:10.4209/aaqr.2015.08.0502*

Biomass burning (BB) frequently occurs in SouthEast Asia (SEA), which significantly affects the air quality and could consequently lead to adverse health effects. The aim of this study was to characterize particulate matter (PM) and black carbon (BC) emitted from BB source regions in SEA and their potential of deposition in the alveolar region of human lungs. A 31-day characterization of PM profiling was conducted at the Doi Ang Khang (DAK) meteorology station in northern Thailand in March 2013. Substantial numbers of PM (10147 ± 5800 # cm^{-3}) with a geometric mean diameter (GMD) of 114.4 ± 9.2 nm were found at the study site. The PM of less than 2.5 μm in aerodynamic diameter ($PM_{2.5}$) hourly-average mass concentration was 78.0 ± 34.5 μg m^{-3}, whereas the black carbon (BC) mass concentration was 4.4 ± 2.6 μg m^{-3}. Notably, high concentrations of nanoparticle surface area (100.5 ± 54.6 μm^2 cm^{-3}) emitted from biomass burning can be inhaled into the human alveolar region. Significant correlations with fire counts within different ranges around DAK were found for particle number, the surface area concentration of alveolar deposition, and BC. In conclusion, biomass burning is an important PM source in SEA, particularly nanoparticles, which has high potency to be inhaled into the lung environment and interact with alveolar cells, leading to adverse respiratory effects. The fire counts within 100 to 150 km shows the highest Pearson's *r* for particle number and surface area concentration. It suggests 12 to 24 hr could be a fair time scale for initial aging process of BB aerosols. Importantly, the people lives in this region could have higher risk for PM exposure.

Keywords: Air pollution; Alveoli; Biomass burning; Black carbon; Nanoparticle.

Incremental Lifetime Cancer Risk of PM2.5 Bound Polycyclic Aromatic Hydrocarbons (PAHs) before and after the Wildland Fire Episode

Siwatt Pongpiachan

Source: *Volume 16, No. 11, November 2016, Pages 2907-2919doi:10.4209/aaqr.2015.01.0011*

In Northern Thailand, wildland fire during cold period releases large amounts of smoke and fine particles into the atmosphere. The fine particles include several persistent organic compounds such as PAHs. In this study, $PM_{2.5}$-bound PAH concentrations in the air of nine administrative provinces, namely Chiang-Mai, Chiang-Rai, Nan, Phayao, Mae Hong Son, Phrae, Lampang, Lamphun, Uttaradit (N Thailand) were determined during the wildland fire and non-wildland fire seasons. The

monitoring strategy comprised two campaigns in each season. $PM_{2.5}$ was collected using MiniVolTM portable air samplers (Airmetrics) with quartz fibre filters. Both PAHs and their $B[a]P_{Equivalent}$concentrations of other urban cities around the world were significantly higher than those of northern provinces for both seasons. The average cancer risks observed at nine administrative provinces were $8.525 \times 10–4 \pm 3.493 \times 10^{-3}$ and $2.558 \times 10^{-3} \pm 6.986 \times 10^{-3}$ for ingestion rate of 50 and 100 mg day^{-1}, respectively. The excess cancer risks of world cities for ingestion rate of 50 and 100 mg day^{-1} were much higher than those of Northern Thailand for 851 and 567 times in that order. Dust ingestion was exceedingly critical to non-dietary PAH exposure in comparison with $PM_{2.5}$ inhalation. These results are in good agreement with those of previous studies, underlining the significance of indoor air quality on long-term adverse respiratory diseases in Asian cities.

Keywords: Wildland fire; PAHs; Northern Thailand; $PM_{2.5}$; Incremental lifetime cancer risk.

The Simulation of Long-Range Transport of Biomass Burning Plume and Short-Range Transport of Anthropogenic Pollutants to a Mountain Observatory in East Asia during the 7-SEAS/2010 Dongsha Experiment

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The Community Multi-scale Air Quality Model (CMAQ) is used to simulate the long-range transport of biomass burning (BB) pollutants from Southeast Asia (SEA) towards the Taiwan Central Mountain Range (CMR) in March and April 2010. The results show that a proportion of the BB plume was blocked and compressed at the windward side of CMR. High-altitude BB plume is shown to influence air quality on the ground via three mechanisms: (1) the subsidence in the anticyclone, (2) the downward motion in the cold surge, and (3) the vertical mixing of the boundary layer over land. Two case studies are further investigated to probe the chemical evolution of the air parcel approaching Mt. Lulin. The first case shows that the third mechanism also explained the increase in the concentrations of peroxyacyl nitrate (PAN), higher peroxyacyl nitrate (PANX), NH_3, SO_2, and volatile organic compounds in the BB plume when entering the land over western Taiwan. Meanwhile, the percentage of NO_3^- in the plume is also significantly increased. The second case reveals that valley wind transported air pollutants from the ground to the mountains. The air parcel, accompanied with considerable concentrations of PAN, PANX, SULF, and anthropogenic secondary organic aerosol, moved up Mt. Lulin. The pollutant concentrations, except for elemental carbon, in the air parcel decreased on approach to Mt. Lulin because the air parcel was mixed with a clean air.

Keywords: Biomass burning; Lulin high-mountain site; Simulation; 2010 Dongsha experiment.

An Overview of PCDD/F Inventories and Emission Factors from Stationary and Mobile Sources: What We Know and What is Missing

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This overview attempts to outline what we currently know about the PCDD/F emission inventories and the source categories therein. Besides the best available control techniques, suggestions are offered on how to reduce the PCDD/F emission factors and emission quantity of some important PCDD/F emission sources. The PCDD/F combustion sources can be classified as either stationary or mobile or minimally/uncontrolled combustion sources. The major stationary sources of PCDD/Fs are metal production processes, waste incineration, heat and power plants, and fly ash treatment plant. Crematories, vehicles, residential boilers and stoves are of key concern due to their proximity to residential areas and their relatively lower lying stacks and exhaust gases, which may result in great impact to their surrounding environment.Moreover, we offered our perspectives on how to improve the quality and representative of the PCDD/F emission factors to attain PCDD/F inventories which correspond more to reality. These points of view include: (1) PCDD/F contributions during start-up procedures of MSWIs should be considered, (2) the sampling times of stack flue gases for EAFs and secondary metal smelters should correspond to whole smelting process stages, (3) longer flue gas sampling time should be executed for power plants, (4) direct exhaust samplings from tailpipes for mobile sources, (5) development of an open burn testing facility that can reflect the real open burning conditions, and (6) long-term sampling techniques like AMESA are suggested to used exclusively for the most contributed PCDD/F stationary sources.

Keywords: PCDD/F inventory; Stationary sources; Mobile sources; Biomass burning; AMESA; Electric arc furnaces.

Design and Characterization of an Inhalation System to Expose Rodents to Nanoaerosols

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The number of workers potentially exposed to nanoparticles (NPs) in industrial processes is constantly increasing, even though the toxicological effects of these compounds have not yet been fully characterized. The hazards associated with this exposure can be assessed most relevantly by toxicology studies involving inhalation of nanoaerosols by animals.In this paper, we describe and characterize an aerosol generated in a nose-only exposure system used to study the respiratory effects of NPs in rat; this system was designed to meet the most stringent requirements for animal testing in terms of protection of operators against risks associated with NPs and biohazards and to comply with the OECD guidelines for chemical testing. The inhalation facility was fully validated by exposing Fisher 344 rats to TiO_2 P25 aerosols at 10 mg m^{-3}. Aerosol monitoring and in-depth characterization were ensured by real-time devices (condensation particle counter, optical particle sizer, scanning mobility particle sizer, aerodynamic particle sizer and electrical low pressure impactor) and samples taken for off-line analyses (gravimetric analysis, mass size distribution from cascade impactor, TEM observations). The test atmosphere was stable in terms of concentrations and distributions (mass or number) between different inhalation towers on a given day and between days (intra-experiment), as well as between inhalation campaigns (between experiments). In terms of the respiratory deposition profile, preliminary results after exposure for one month indicate that this system is relevant, and should therefore be appropriate for in vivo inhalation toxicity studies.

Keywords: Inhalation; Rodent; Nanoaerosols; Titanium dioxide.

Improving Urban Air Quality Measurements by a Diffusion Charger Based Electrical Particle Sensors – A Field Study in Beijing, China

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High aerosol loadings contribute significantly to the air quality problems of Asian megacities. To address this, monitoring data for aerosol mass and number that is spatially and temoprally of high resolution is needed, while the cost of obtaining such data remains high. Here, we present a field study in a polluted megacity, Beijing, using a diffusion-charge-based electrical aerosol sensor, the Pegasor PPS-M, which is a robust and comparatively low-cost instrument for the monitoring of both aerosol mass and number simultaneously. We present data over several months in the year 2014, and for varying aerosol size distributions, and analyze the performance against particle number and mass (volume) measured using a wide range particle sizer (WPS) and beta-attenuation-based $PM_{2.5}$ observations. We show that using a single trap voltage, the PPS-M correlates well with particle mass, but not so well with particle number due to the variability in particle size distributions. However, the instrument response to number was improved by running the

instrument with a variable trap voltage, and using the ratio of the different signals to gain information on the particle average volume. With this method, we were able to improve the correlation of the PPS-M; with the observed particle number from R = 0.14 to R = 0.72 for the measurement time period. Altogether, the PPS-M instrument displayed robustness and low maintenance requirements, and it showed good correlation with the other instruments in this study.

Keywords: Diffusion charging; Aerosol instrumentation; China air quality; Urban aerosol.

Particulate Matter, Ozone, and Nitrogen Species in Aged Wildfire Plumes Observed at the Mount Bachelor Observatory

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During the summer of 2012 and 2013, we measured carbon monoxide (CO), carbon dioxide (CO_2), ozone (O_3), nitrogen oxides (NO_x), reactive nitrogen (NO_y), peroxyacetyl nitrate (PAN), aerosol scattering (σ_{sp}) and absorption, elemental and organic carbon (EC and OC), and aerosol chemistry at the Mount Bachelor Observatory (2.8 km above sea level, Oregon, US). Here we analyze 23 of the individual plumes from regional wildfires to better understand production and loss of aerosols and gaseous species. We also developed a new method to calculate enhancement ratios and Modified Combustion Efficiency (MCE), which takes into account possible changes in background concentrations during transport. We compared this new method to existing methods for calculating enhancement ratios. The MCE values ranged from 0.79–0.98, $\Delta O_3/\Delta CO$ ranged from 0.01–0.07 ppbv ppbv^{-1}, $\Delta\sigma_{sp}/\Delta CO$ ranged from 0.23–1.32 Mm^{-1} (at STP) ppbv^{-1}, $\Delta NO_y/\Delta CO$ ranged from 2.89–12.82 pptv ppbv^{-1}, and $\Delta PAN/\Delta CO$ ranged from 1.46–6.25 pptv ppbv^{-1}. A comparison of three different methods to calculate enhancement ratios (ER) showed that the methods generally resulted in similar $\Delta\sigma_{sp}/\Delta CO$, $\Delta NO_y/\Delta CO$, and $\Delta PAN/\Delta CO$; however, there was a significant bias between the methods when calculating $\Delta O_3/\Delta CO$ due to the small absolute enhancement of O_3 in the plumes. The $\Delta O_3/\Delta CO$ ERs calculated using two common methods were biased low (~20–30%) when compared to the new proposed method. Two pieces of evidence suggest moderate secondary particulate formation in many of the plumes studied: 1) mean observed $\Delta OC/\Delta CO_2$ was 0.028 g particulate-C gC^{-1} (as CO_2)—27% higher than the midpoint of the biomass burning emission ratio range reported by a recent review—and 2) single scattering albedo (ω) was relatively constant at all MCE values, in contrast with results for fresh plumes. The observed NO_x, PAN, and aerosol nitrate represented 6–48%, 25–57%, and 20–69% of the observed NO_y in the aged plumes, respectively, and other species represented on average 11% of the observed NO_y.

Keywords: Particulate matter; Ozone; NOy; Enhancement ratio; Modified Combustion Efficiency.

Variability of Anthropogenic Gases: Nitrogen Oxides, Sulfur Dioxide, Ozone and Ammonia in Kathmandu Valley, Nepal

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Kathmandu Valley is one of the largest and most polluted metropolitan regions in the Himalayan foothills. Rapidly expanding urban sprawl and a growing fleet of vehicles, and industrial facilities such as brick factories across the valley have led to conditions where ambient concentrations of key gaseous air pollutants are expected to exceed Nepal's National Ambient Air Quality Standards (NAAQS) and World Health Organization (WHO) guidelines. In order to understand the spatial variation of the trace gases in the Kathmandu Valley, passive samples of SO_2, NO_x, NO_2, NH_3, and O_3 were collected simultaneously from fifteen locations between March and May 2013. A follow-up study during two separate campaigns in 2014 sampled these gases, except ammonia, one site at a time from thirteen urban, suburban and rural stationary sites. In 2013, urban sites were observed to have higher weekly averaged NO_2 and SO_2 (22.4 ± 8.1 μg m^{-3} and 14.5 ± 11.1 μg m^{-3}, respectively) than sub-urban sites (9.2 ± 3.9 μg m^{-3} and 7.6 ± 2.8 μg m^{-3}, respectively). Regions located within 3 km of brick factories had higher SO_2 concentrations (22.3 ± 14.7 μg m^{-3}) than distant sites (5.8 ± 1.1 μg m^{-3}). Higher O_3 (108.5 ± 31.4 μg m^{-3}) was observed in rural locations compared to urban sites (87.1 ± 9.2 μg m^{-3}), emphasizing the importance of meteorological factors and precursor species for ozone production and titration. Parallel to previous studies, these results suggest that ground-level O_3, as its levels frequently exceeded guidelines throughout the sampling periods, is an important concern throughout the valley. NH_3 near polluted rivers and SO_2around brick factories are also important pollutants that need more intensive monitoring, primarily due to their importance in particulate matter formation chemistry.

Keywords: Nitrogen oxides; Sulfur dioxide; Ozone, Tropospheric; Passive samplers; Air quality.

Characterization of a Smog Chamber for Studying Formation and Physicochemical Properties of Secondary Organic Aerosol

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Source: *Volume 16, No. 12, December 2016, Pages 3102-3113doi:10.4209/aaqr.2015.10.0580*

An indoor smog chamber facility has been built for carrying out secondary organic aerosol (SOA) formation and for studying physicochemical properties of SOA. This facility comprises of ~7 m^3 FEP Teflon reactor placed in temperature controlled room coupled with instruments for gas and particle phase data. Detailed characterization experiments have been presented describing control of reactor temperature, relative humidity (RH), effective mixing time, wall loss rates of gases and particles, light source, and air purification. This chamber showed a wide range of temperature control with acceptable precision (i.e., 18–33 ± 0.5°C). The gas wall loss rates for NO, NO_2, and O_3 were found to be 3.78×10^{-4} min^{-1}, 4.48×10^{-5} min^{-1}, and 6.47×10^{-5} min^{-1}, respectively. NO_2 photolysis rate constant was 0.17 min^{-1}. Particle wall loss constant was found to be 3.96×10^{-3} min^{-1} at D_p = 100 nm. SOA yields of dark α-pinene ozonolysis ranged from 0.025 to 0.378 for α-pinene concentrations from 10 ppb to 100 ppb.

Keywords: Smog chamber; Secondary organic aerosol (SOA); α-pinene; Ozonolysis.

Variability in Transport Pathways and Source Areas of PM10 in Beijing during 2009–2012

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The transport pathways and source areas of PM_{10} in Beijing were examined on the basis of a model-assisted analysis. Computed back trajectories were used to trace the air history. The aim of this work was to study the main source areas of PM_{10}, the variability of transport pathways, and potential source areas in Beijing. The results reveal that the major potential source areas of PM_{10} in Beijing were Hebei, Shandong, Tianjin, northwest of Inner Mongolia, and Outer Mongolia. The main source areas of PM_{10} have changed from the northwest to the south and southeast of Beijing during 2009–2012. During the study period, the regional contributions of PM_{10} from Shandong, Tianjin and Henan increased, whereas those from Inner Mongolia and Mongolia decreased compared with 2003–2009. The northwest airflow is a key factor in extreme pollution episodes. Sand storm partly

contributed to the PM_{10} concentration in fast northwesterly transport paths. PM_{10}concentrations in winter and spring were higher than autumn and summer. In spring and summer, Beijing was strongly affected by long-range transport. Long-range transport had a weaker effect on PM_{10} concentrations during autumn and winter. The clustering of back-trajectories and PSCF results indicate the need to reduce PM_{10} transport from areas surrounding Beijing, particularly from the south of Beijing.

Keywords: PM_{10}; Back trajectories; Regional sources; Extreme pollution episodes.

Characterization of PAHs and PCBs in Fly Ashes of Eighteen Coal-Fired Power Plants

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For the management of coal fly ashes (CFAs) from coal-fired power plants (CFPPs), characterization of PAHs and PCBs in CFAs is imperative. The 18 PAH and 86 PCB congeners in CFAs collected from 18 large-scale CFPPs in China were detected using GC/MS system. The PAH concentrations were in the range of 5.51–70.9 ng g^{-1} for 16 CFPPs with individual block power capacity as 600 MW (IBPC-600), significantly lower than 886–916 ng g^{-1} for 2 CFPPs with IBPC as 200 and 300 MW (IBPC-200/300). Both PAH and PCB congeners for 18 CFPPs were dominated by low molecular weight ones. The 3- and 2-ring PAHs, di-, tri- and tetra-PCBs were the predominant homologs. PAH profiles for 16 CFPPs with IBPC-600 were significantly different from other industrial stacks based on higher coefficients of divergence. The BaP-based toxic equivalency (BaPeq) concentration and BaP-based equivalent carcinogenic power (BaPE) for 16 CFPPs with IBPC-600 were 0.834 ng g^{-1} and 0.570, much lower than corresponding 20.5 ng g^{-1} and 15.4 for 2 CFPPs with IBPC-200/300. No difference existed for Σ_{86}PCBs between CPFFs with IBPC-600 and -200/300, which ranged from 9.60 to 32.1 ng g^{-1}. Higher mean carcinogenic PAH concentrations for 2 CFPPs with IBPC-200/300 and PCBs-TEQ concentration for 18 CFPPs indicated the application of CFAs as soil amendment should be prohibited. The PAH concentrations for 18 CFPPs were well correlated with the total organic carbon (TOC) values, while PCB concentrations showed not this trend, indicated the different formation mechanism between PCBs and PAHs.

Keywords: PAHs; PCBs; Coal fly ash; Coal-fired power plant.

Characterization of Bioaerosols and their Relation with OC, EC and Carbonyl VOCs at a Busy Roadside Restaurants-Cluster in New Delhi

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Source: *Volume 16, No. 12, December 2016, Pages 3198-3211doi:10.4209/aaqr.2016.01.0043*

Measurement of biological particles (bioaerosols) in ambient air is of great importance as it is directly linked with the health issues. However, data on the bioaerosols characterization are scarce. Here we report on the concentration and characterization of bioaerosols (including bacterial and fungal aerosols) as well as determination of organic and elemental carbon (OC and EC) in total suspended particulate matter (TSPM) at a busy roadside restaurants-cluster site in New Delhi. In addition, 14 carbonyl volatile organic compounds (carbonyl VOCs) were also measured and their relationship with bioaerosols and OC/EC is assessed. The culturable airborne bacterial and fungal concentrations (CAB and CAF) at restaurant area varied significantly in different seasons ranging from 1.7×10^4–9.8×10^4 (averaged $6.3 \times 10^4 \pm 2.6 \times 10^4$ cfu m^{-3}) and 3.5×10^2–9.5×10^3 ($3.9 \times 10^3 \pm 3.1 \times 10^3$ cfu m^{-3}) cfu m^{-3}, respectively. Major concentration peaks of TSPM, OC, EC as well as bacterial and fungal aerosols were found in winter and spring seasons. These peaks can be attributed to the low atmospheric boundary layer (ABL) height and favourable meteorological conditions for microbial growth in winter and spring seasons in New Delhi. Good correlations ($R^2 > 0.5$) were observed between CAB, CAF, TSPM and OC. On the other hand, CAB and CAF were not found to be correlated with carbonyl compounds ($R^2 < 0.2$) indicative of their diverse sources. The bacterial identification was done by 16s rDNA sequencing and the identified strains were *Bacillus sp*., *Bacillus firmus*, *Bacillus licheniformis*, *Bacillus cereus*, *Bacillus pumilus*, *Acinetobacter sp*. and *Acinetobactor radioresistens* gene. Predominant fungal genera identified were *Aspergillus*, *Cladosporium*, *Alternaria*and *Fusarium*, which are known for adverse health effects causing numerous allergic and pathogenic inflammations.

Keywords: Bioaerosol; Bacteria; Fungi; Carbonyls; Organic and elemental carbon.

Analysis of the Effect of Meteorological Factors on PM2.5-Associated PAHs during Autumn-Winter in Urban Nanchang

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Source: *Volume 16, No. 12, December 2016, Pages 3222-3229doi:10.4209/aaqr.2016.08.0351*

The objectives of this study were to examine the association between $PM_{2.5}$-associated polycyclic aromatic hydrocarbons (PAHs) and their meteorological factors, and the relationship between $PM_{2.5}$-associated PAH concentrations and temperature, air pressure and air humidity. Data collected by Center for Disease Control (CDC) in Nanchang urban areas during the fall and winter seasons of 2014–2015 were analyzed by using high performance liquid chromatography (HPLC). Our study showed that the $PM_{2.5}$ mass concentration had a mean of 0.088 μg m^{-3}. Our results showed that the total concentration of PAHs in Nanchang was 22.54 ng m^{-3} (RSD: 8.50) and 15 different types of PAHs examined in this study all exceeded the China national standard. Multiple regression analysis revealed that the daily average concentration of total PAH was significantly associated with the temperature and daily minimum humidity ($p < 0.05$), but not daily wind speed and rainfall. Principal component analysis and characteristic ratio study indicated that the source of PAHs in $PM_{2.5}$ were mainly from vehicle exhaust and coal and gas combustion in Nanchang.

Keywords: $PM_{2.5}$; Meteorological factors; Polycyclic Aromatic Hydrocarbons (PAHs); HPLC.

Multi-Year Analysis of Aerosol Properties Retrieved from the Ångström Parameters for Different Spectral Ranges over Pune

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Source:*Volume 16, No. 12, December 2016, Pages 3266-3280 doi:10.4209/aaqr.2016.06.0268*

The present study evaluates the temporal variation of aerosol optical depth (AOD_{500} nm) and the Ångström parameters [viz., Ångström exponent (AE, α), Ångström turbidity coefficient (β) and second order Ångström exponent (α′)] at a tropical observing site, Pune (18°32′N; 73°49′E, 559 m AMSL) during 2008–15. Six-year means for winter and pre-monsoon seasons together are found to be 0.534 ± 0.13, 1.054 ± 0.27, 0.254 ± 0.08 and 0.167 ± 1.33 for AOD_{500} nm, AE, β and α′ respectively. Average month-to-month variability of AOD_{500} nm, AE, β and α′ during 2008–15 depicts seasonal cycle with strong departures with respect to long-term means. Frequency distributions for AOD, AE and β are positively skewed (skewness = 0.77, 0.32 and 1.14 respectively) while it is negatively skewed for α′ (skewness = –0.18). Analysis of AE difference, curvature parameter difference ($\alpha_2-\alpha_1$) and AOD_{500} nm–$AE_{440\text{-}870}$ nm contour density map reveals that the aerosol ensemble at Pune consists of four aerosol types viz., UI (urban/industrial), CM (clear maritime), DD (desert dust) and MT (mixed type). Their relative magnitudes, however, differ during winter and pre-monsoon seasons. Thus, the contour density map shows dominance of UI and relatively less occurrence of MT type aerosols during winter. In pre-monsoon, however, the aerosol scenario is driven by MT type aerosol although UI and DD type aerosols show their remarkable existence.

Keywords: Aerosol optical depth; Ångström exponent; Ångström turbidity coefficient; Aerosol loading; Aerosol sources.

Emission Characteristics and Control Prospects of Primary PM2.5 from Fossil Fuel Power Plants in China

Yan Wang, Ke Cheng, He-Zhong Tian, Peng Yi, Zhi-Gang Xue

Source: *Volume 16, No. 12, December 2016, Pages 3290-3301doi:10.4209/aaqr.2016.07.0324*

In this study, a unit-based approach was used to establish an integrated emission inventory of primary $PM_{2.5}$ from fossil fuel (coal, oil, and natural gas) power plants in China. The inventory was of high spatial and temporal resolution, and composed of detailed chemical speciation. In 2014, the total emissions were estimated to be approximately 669.53 kt. The emissions of primary $PM_{2.5}$ from coal-fired power plants (CFPs) were 668.56 kt, making CFPs the largest contributor. The emissions of primary $PM_{2.5}$ from oil-fired power plants (OFPs) and natural gas-fired power plants (GFPs) were approximately 17.41 t and 945.60 t, respectively. Spatial distribution features demonstrated that the emissions in the eastern and central provinces of China were much higher than those in the west, except for provinces involved in the "west-to-east power transmission" project. For CFPs, crustal elements and water-soluble inorganic ions were the primary species of $PM_{2.5}$. By contrast, for OFPs and GFPs, carbonaceous components were the predominant species of $PM_{2.5}$. Moreover, this study conducted a scenario analysis of changes in $PM_{2.5}$ emissions resulting from technical advancement, penetration, and substitution of CFPs by GFPs for the target years.

Keywords: $PM_{2.5}$ emission; Unit-based approach; Chemical composition; Scenario projection.

Reducing Cold Start Emissions from Automotive Diesel Engine at Cold Ambient Temperatures

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Source: *Volume 16, No. 12, December 2016, Pages 3330-3337doi:10.4209/aaqr.2015.11.0616*

Cold start performance of diesel engines is determined by engine design, fuel type, fuel injection strategies, lubricant and ambient temperature conditions. Prevailing emissions legislation regarding low temperature emission tests applicable for gasoline vehicles is likely to be implemented to diesel vehicles. The present research work investigates the effects of intake air heating on a Euro 5 diesel engine's performance and exhaust emission (gaseous and particulate

emissions) characteristics during the cold start followed by idling at cold ambient conditions. Heating of intake air entering the engine at cold ambient temperature conditions improved fuel combustion as well as reduced the cranking period and improved the fuel economy. More than 50% reduction in HC and 17% reduction in NOx emissions were achieved by intake air heating. Number count of accumulation mode particulates were higher during cold start compared to idle operation for all the temperature conditions. Intake air heating decreased the particulate number and size that led to reduction in total particulate mass by higher than 50% and 75% during cold start and idle respectively. The intake air heating strategy improved the cold start performance of the diesel engine at cold ambient temperature conditions and thereby would reduce the overall driving cycle emissions.

Keywords: Cold start; Particulates; Emissions; Diesel engines.

Atmospheric deposition of organochlorine pesticides by precipitation in a coastal area

S. Sıddık Cindoru, Erman Ozturk

Source: *Environmental Science and Pollution Research, December 2016, Volume 23, Issue 24, pp 24504–24513*

Wet deposition fluxes of organochlorine pesticides (OCPs) were determined for rain samples collected in a coastal area of Turkey. Seventeen precipitation samples were collected over a 1-year period from 2008 to 2009. Rainwater was accumulated at the beginning of rain events using real time monitoring. Atmospheric concentrations were also measured in parallel with deposition samples. Both atmospheric concentrations and deposition fluxes were determined as particle and gas phases. The particle phase and dissolved phase deposition fluxes were 794.26 ± 756.70 ngm−2 day−1 and 800.77 ± 672.63 ngm−2 day−1, respectively. The washout ratios for OCP compounds were calculated separately for the particle and dissolved phases using the atmospheric concentrations and rain concentrations. The minimum washout ratio for the particle phase was 2339.47 for Endrin aldehyde, whereas the maximum washout ratio was 497593.34 for Methoxychlor. The maximum washout ratio for the dissolved phase was 247523.89 for Endosulfan beta, whereas the minimum washout ratio was 10169.69 for p,p′-DDT. The dry deposition velocities ranged from 0.01 to 1.67 cms−1. The partitioning of wet deposition between the particle and dissolved phases was 50 % in terms of total OCP deposition.

Keywords: OCPs, Wet Dry deposition, Rainwater concentration, Flux Persistent organic pollutants

Chemical characterization of rainwater at a high-altitude site “Nainital” in the central Himalayas, India

Deewan Singh Bisht, A.K. Srivastava, H. Joshi, K. Ram, N. Singh, M. Naja, M.K. Srivastava,S. Tiwari

Source: *Environmental Science and Pollution Research, pp 1–11, DOI: 10.1007/s11356-016-8093-z*

The present study investigates the chemical composition of rainwater (RW) from a high-altitude site “Nainital” (1958 m above msl) in the central Himalaya region, to understand the influence of local, regional, and long-range transport of pollutants. A total of 55 (2 in pre-monsoon and 53 in monsoon) RW samples were collected during the study period (June–September 2012) and were analyzed for major anions and cations using an ion chromatograph. The pH of precipitation events ranged from 4.95 to 6.50 (average 5.6 ± 0.3) was observed during the monsoon period (near to the acidic), whereas during the pre-monsoon, the pH was 6.25 ± 0.49 (alkaline) over the study region; it is due the mixture of anthropogenic as well as the natural chemical constituents. The average ionic concentration (sum of measured chemical constituents) was ∼3 times higher during the pre-monsoon (986 ± 101 μeq/1) compared to that in the monsoon season (373 ± 37 μeq/1). This is mainly due to the presence of more natural aerosols in the pre-monsoon season which is also reflected in the pH of rainwater (average 6.25 ± 0.50) as well as ionic concentration. The chemical composition suggests that Ca2+ was the major contributor (34%) among cations, followed by Na+ (10%), K+ (8%), and Mg2+ (9%), whereas Cl−, NO3−, and SO42− contributed ∼13, 11, and 9%, respectively, among anions. The average ratio of acidic species (SO42−/NO3−) is 1.56, suggesting 61 and 39% contribution of SO42− and NO3−, respectively, which is very close to the estimated contribution of H2SO4 (60–70%) and HNO3 (30–40%) in the precipitation samples. Neutralization factors for Ca2+, Mg2+, and NH4+ in RW at Nainital are 4.94, 1.21, and 0.37, respectively, indicating their crucial role in neutralization of acidic species. The non-sea-salt (NSS) contribution to total Ca2+, K+, and Mg2+is estimated to be ∼98, 97, and 74%, respectively, suggesting the dominance of crustal sources for cations. In contrast, the NSS contribution to the total Cl− and SO42− is 16 and 69% indicating their anthropogenic origin, respectively. Principle component analysis also suggests that the first factor (i.e., natural sources, mainly dust, and sea-salts) accounts for ∼33% variance, whereas the second factor (i.e., fossil fuel and biomass burning) accounts for ∼18% variance of the measured ionic composition. The remaining contributions are attributed to the mixed emission sources and transport of pollutants from Indo-Gangetic Plain (IGP) and western parts of India. The results of the present study reveal a significant contribution of crustal and anthropogenic sources in the RW and neutralization processes in the central Himalaya.

Keywords: Rainwater chemistry, High altitude, Ionic concentration, Neutralizing factor, PCA

Source identification of combustion-related air pollution during an episode and afterwards in winter-time in Istanbul

S. Levent Kuzu

Source: *Environmental Science and Pollution Research, pp 1–10, DOI: 10.1007/s11356-016-7831-6*

Conventional air pollutants (PM10, CO, NOx) gradually increased from fall to winter during 2015 in Istanbul. Several air pollution episodes were observed during this period. This study was made in order to determine polycyclic aromatic hydrocarbon (PAH) levels, identify the sources of air pollution, and make toxicity assessment based on Benzo(a)pyrene equivalent concentrations. The sampling took 14 sequential days during winter. High-pressure weather conditions prevailed at the start of the sampling. The conditions were then changed to low-pressure condition towards the end of the sampling. Strong inversion was effective on the onset of the sampling. Strong inversion was effective at the onset of the sampling. A high-volume sampler was used to collect gas and particle phase samples. Total suspended particle concentrations were between 27 and 252 μg m−3. Sixteen PAH species were investigated. Total (gas + particle) PAH concentrations were between 76.4 and 1280.3 ng m−3, with an average of 301.4 ng m−3. Individual PAH concentrations were between not detected (n.d.) and 99.2 ng m−3in the gaseous phase, and between n.d. and 11.5 ng m−3 in the particle phase. Phenanthrene had the highest share among 16 PAH compounds. Benzo(a)pyrene was not detected in 8 days. On the remaining days, its concentration ranged between 5.5 and 14.8 ng m−3 with an average of 3.7 ng m−3. Low-molecular-weight PAHs dominated gaseous phase; inversely, high-molecular-weight PAHs dominated particle phase. Possible sources were identified by diagnostic ratios. These ratios suggested that coal combustion and diesel vehicle exhaust emissions had a substantial impact on ambient air quality. Benzo(a)pyrene equivalencies were calculated for each PAH compound in order to make toxicity assessment. Total benzo(a)pyrene equivalencies ranged between 0.4 and 30.0 ng m−3 with an average of 7.2 ng m−3.

Keywords: Air pollution episode, PAH Diagnostic ratio, Source identification, Gas-particle partitioning, Toxicity assessment

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Chemical characterization of rainwater at a high-altitude site “Nainital” in the central Himalayas, India

Deewan Singh Bisht, A.K. Srivastava, H. Joshi, K. Ram, N. Singh, M. Naja, M.K. Srivastava,S. Tiwari

Source: *Environmental Science and Pollution Research, pp 1–11, DOI: 10.1007/s11356-016-8093-z*

The present study investigates the chemical composition of rainwater (RW) from a high-altitude site “Nainital” (1958 m above msl) in the central Himalaya region, to understand the influence of local, regional, and long-range transport of pollutants. A total of 55 (2 in pre-monsoon and 53 in monsoon) RW samples were collected during the study period (June–September 2012) and were analyzed for major anions and cations using an ion chromatograph. The pH of precipitation events ranged from 4.95 to 6.50 (average 5.6 ± 0.3) was observed during the monsoon period (near to the acidic), whereas during the pre-monsoon, the pH was 6.25 ± 0.49 (alkaline) over the study region; it is due the mixture of anthropogenic as well as the natural chemical constituents. The average ionic concentration (sum of measured chemical constituents) was ~3 times higher during the pre-monsoon (986 ± 101 μeq/1) compared to that in the monsoon season (373 ± 37 μeq/1). This is mainly due to the presence of more natural aerosols in the pre-monsoon season which is also reflected in the pH of rainwater (average 6.25 ± 0.50) as well as ionic concentration. The chemical composition suggests that Ca2+ was the major contributor (34%) among cations, followed by Na+ (10%), K+ (8%), and Mg2+ (9%), whereas Cl−, NO3−, and SO42− contributed ~13, 11, and 9%, respectively, among anions. The average ratio of acidic species (SO42−/NO3−) is 1.56, suggesting 61 and 39% contribution of SO42− and NO3−, respectively, which is very close to the estimated contribution of H2SO4 (60–70%) and HNO3 (30–40%) in the precipitation samples. Neutralization factors for Ca2+, Mg2+, and NH4+ in RW at Nainital are 4.94, 1.21, and 0.37, respectively, indicating their crucial role in neutralization of acidic species. The non-sea-salt (NSS) contribution to total Ca2+, K+, and Mg2+is estimated to be ~98, 97, and 74%, respectively, suggesting the dominance of crustal sources for cations. In contrast, the NSS contribution to the total Cl− and SO42− is 16 and 69% indicating their anthropogenic origin, respectively. Principle component analysis also suggests that the first factor (i.e., natural sources, mainly dust, and sea-salts) accounts for ~33% variance, whereas the second factor (i.e., fossil fuel and biomass burning) accounts for ~18% variance of the measured ionic composition. The remaining contributions are attributed to the mixed emission sources and transport of pollutants from Indo-Gangetic Plain (IGP) and western parts of India. The results of the present study reveal a significant contribution of crustal and anthropogenic sources in the RW and neutralization processes in the central Himalaya.

Keywords: Rainwater chemistry, High altitude, Ionic concentration, Neutralizing factor, PCA

Advanced mortar coatings for cultural heritage protection. Durability towards prolonged UV and outdoor exposure

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Source: *Environmental Science and Pollution Research, pp 1–10, DOI: 10.1007/s11356-016-7611-3*

In the present work, two kinds of hybrid polymeric–inorganic coatings containing TiO2 or SiO2particles and prepared starting from two commercial resins (Alpha®SI30 and

Bluesil®BP9710) were developed and applied to two kinds of mortars (an air-hardening calcic lime mortar [ALM] and a natural hydraulic lime mortar [HLM]) to achieve better performances in terms of water repellence and consequently damage resistance. The two pure commercial resins were also applied for comparison purposes. Properties of the coated materials and their performance were studied using different techniques such as contact angle measurements, capillary absorption test, mercury intrusion porosimetry, surface free energy, colorimetric measurements and water vapour permeability tests. Tests were also performed to determine the weathering effects on both the commercial and the hybrid coatings in order to study their durability. Thus, exposures to UV radiation, to UV radiation/condensed water cycles and to a real polluted atmospheric environment have been performed. The effectiveness of the hybrid SiO2 based coating was demonstrated, especially in the case of the HLM mortar.

Keywords: Mortars, Hybrid coatings, Cultural heritage, Surface modification, Aging, Exposure

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Size-resolved source apportionment of particulate matter in urban Beijing during haze and non-haze episodes

S. L. Tian, Y. P. Pan, and Y. S. WangState

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Additional size-resolved chemical information is needed before the physicochemical characteristics and sources of airborne particles can be understood; however, this information remains unavailable in most regions of China due to lacking measurement data. In this study, we report observations of various chemical species in size-segregated particle samples that were collected over 1 year in the urban area of Beijing, a megacity that experiences severe haze episodes. In addition to fine particles, high concentrations of coarse particles were measured during the periods of haze. The abundance and chemical compositions of the particles in this study were temporally and spatially variable, with major contributions from organic matter and secondary inorganic aerosols. The contributions of organic matter to the particle mass decreased from 37.9 to 31.2 %, and the total contribution of sulfate, nitrate and ammonium increased from 19.1 to 33.9 % between non-haze and haze days, respectively. Due to heterogeneous reactions and hygroscopic growth, the peak concentrations of the organic carbon, cadmium and sulfate, nitrate, ammonium, chloride and potassium shifted from 0.43 to 0.65 μm on non-haze days to 0.65–1.1 μm on haze days. Although the size distributions of lead and thallium were similar during the observation period, their concentrations increased by a factor of more than 1.5 on haze days compared with non-haze days. We observed that sulfate and ammonium, which have a size range of 0.43–0.65 μm, sulfate and nitrate, which have a size range of 0.65–1.1 μm, calcium, which has a size range of 5.8–9 μm, and the meteorological factors of relative humidity and wind speed were responsible for haze pollution when the visibility was less than 10 km. Source apportionment using Positive Matrix Factorization

showed six $PM_{2.1}$ sources and seven $PM_{2.1–9}$ common sources: secondary inorganic aerosol (25.1 % for fine particles vs. 9.8 % for coarse particles), coal combustion (17.7 % vs. 7.8 %), biomass burning (11.1 % vs. 11.8 %), industrial pollution (12.1 % vs. 5.1 %), road dust (8.4 % vs. 10.9 %), vehicle emissions (19.6 % for fine particles), mineral dust (22.6 % for coarse particles) and organic aerosol (23.6 % for coarse particles). The contributions of the first four factors and vehicle emissions were higher on haze days than non-haze days, while the reverse is true for road dust and mineral dust. The sources' contribution generally increased as the size decreased, with the exception of mineral dust. However, two peaks were consistently found in the fine and coarse particles. In addition, the sources' contribution varied with the wind direction, with coal and oil combustion products increasing during southern flows. This result suggests that future air pollution control strategies should consider wind patterns, especially during episodes of haze. Furthermore, the findings of this study indicated that the $PM_{2.5}$-based data set is insufficient for determining source control policies for haze in China and that detailed size-resolved information is needed to characterize the important sources of particulate matter in urban regions and better understand severe haze pollution.

Improving aerosol interaction with clouds and precipitation in a regional chemical weather modeling system

C. Zhou, X. Zhang, S. Gong, Y. Wang, and M. Xue

Source: *Atmos. Chem. Phys., 16, 145-160, 2016*
http://www.atmos-chem-phys.net/16/145/2016/doi:10.5194/acp-16-145-2016

A comprehensive aerosol–cloud–precipitation interaction (ACI) scheme has been developed under a China Meteorological Administration (CMA) chemical weather modeling system, GRAPES/CUACE (Global/Regional Assimilation and PrEdiction System, CMA Unified Atmospheric Chemistry Environment). Calculated by a sectional aerosol activation scheme based on the information of size and mass from CUACE and the thermal-dynamic and humid states from the weather model GRAPES at each time step, the cloud condensation nuclei (CCN) are interactively fed online into a two-moment cloud scheme (WRF Double-Moment 6-class scheme – WDM6) and a convective parameterization to drive cloud physics and precipitation formation processes. The modeling system has been applied to study the ACI for January 2013 when several persistent haze-fog events and eight precipitation events occurred.The results show that aerosols that interact with the WDM6 in GRAPES/CUACE obviously increase the total cloud water, liquid water content, and cloud droplet number concentrations, while decreasing the mean diameters of cloud droplets with varying magnitudes of the changes in each case and region. These interactive microphysical properties of clouds improve the calculation of their collection growth rates in some regions and hence the precipitation rate and distributions in the model, showing 24 to 48 % enhancements of threat score for 6 h precipitation in almost all regions. The aerosols that interact with the WDM6 also reduce the regional mean bias of temperature by 3 °C during certain precipitation events, but the monthly means bias is only reduced by about 0.3 °C.

Can we explain the observed methane variability after the Mount Pinatubo eruption?

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Source: *Atmos. Chem. Phys., 16, 195-214, 2016*
http://www.atmos-chem-phys.net/16/195/2016/doi:10.5194/acp-16-195-2016

The CH4 growth rate in the atmosphere showed large variations after the Pinatubo eruption in June 1991. A decrease of more than 10 ppb yr−1 in the growth rate over the course of 1992 was reported, and a partial recovery in the following year. Although several reasons have been proposed to explain the evolution of CH4 after the eruption, their contributions to the observed variations are not yet resolved. CH4 is removed from the atmosphere by the reaction with tropospheric OH, which in turn is produced by O3 photolysis under UV radiation. The CH4removal after the Pinatubo eruption might have been affected by changes in tropospheric UV levels due to the presence of stratospheric SO2 and sulfate aerosols, and due to enhanced ozone depletion on Pinatubo aerosols. The perturbed climate after the eruption also altered both sources and sinks of atmospheric CH4. Furthermore, CH4 concentrations were influenced by other factors of natural variability in that period, such as El Niño–Southern Oscillation (ENSO) and biomass burning events. Emissions of CO, NOX and non-methane volatile organic compounds (NMVOCs) also affected CH4 concentrations indirectly by influencing tropospheric OH levels. Potential drivers of CH4 variability are investigated using the TM5 global chemistry model. The contribution that each driver had to the global CH4variability during the period 1990 to 1995 is quantified. We find that a decrease of 8–10 ppb yr−1 CH4 is explained by a combination of the above processes. However, the timing of the minimum growth rate is found 6&nash;9 months later than observed. The long-term decrease in CH4growth rate over the period 1990 to 1995 is well captured and can be attributed to an increase in OH concentrations over this time period. Potential uncertainties in our modelled CH4 growth rate include emissions of CH4 from wetlands, biomass burning emissions of CH4 and other compounds, biogenic NMVOC and the sensitivity of OH to NMVOC emission changes. Two inventories are used for CH4 emissions from wetlands, ORCHIDEE and LPJ, to investigate the role of uncertainties in these emissions. Although the higher climate sensitivity of ORCHIDEE improves the simulated CH4 growth rate change after Pinatubo, none of the two inventories properly captures the observed CH4 variability in this period.

Effects of emission reductions on organic aerosol in the southeastern United States

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http://www.atmos-chem-phys.net/16/215/2016/doi:10.5194/acp-16-215-2016

Long-term (1999 to 2013) data from the Southeastern Aerosol Research and Characterization (SEARCH) network are used to show that anthropogenic emission reductions led to important decreases in fine-particle organic aerosol (OA) concentrations in the southeastern US On average, 45 % (range 25 to 63 %) of the 1999 to 2013 mean organic carbon (OC) concentrations are attributed to combustion processes, including fossil fuel use and biomass burning, through associations of measured OC with combustion products such as elemental carbon (EC), carbon monoxide (CO), and nitrogen oxides (NOx). The 2013 mean combustion-derived OC concentrations were 0.5 to 1.4 μg m−3 at the five sites operating in that year. Mean annual combustion-derived OC concentrations declined from 3.8 ± 0.2 μg m−3 (68 % of total OC) to 1.4 ± 0.1 μg m−3 (60 % of total OC) between 1999 and 2013 at the urban Atlanta, Georgia, site (JST) and from 2.9 ± 0.4 μg m−3 (39 % of total OC) to 0.7 ± 0.1 μg m−3 (30 % of total OC) between 2001 and 2013 at the urban Birmingham, Alabama (BHM), site. The urban OC declines coincide with reductions of motor vehicle emissions between 2006 and 2010, which may have decreased mean OC concentrations at the urban SEARCH sites by > 2 μg m−3. BHM additionally exhibits a decline in OC associated with SO2 from 0.4 ± 0.04 μg m−3 in 2001 to 0.2 ± 0.03 μg m−3in 2013, interpreted as the result of reduced emissions from industrial sources within the city. Analyses using non-soil potassium as a biomass burning tracer indicate that biomass burning OC occurs throughout the year at all sites. All eight SEARCH sites show an association of OC with sulfate (SO4) ranging from 0.3 to 1.0 μg m−3 on average, representing ∼ 25 % of the 1999 to 2013 mean OC concentrations. Because the mass of OC identified with SO4 averages 20 to 30 % of the SO4 concentrations, the mean SO4-associated OC declined by ∼ 0.5 to 1 μg m−3 as SO4concentrations decreased throughout the SEARCH region. The 2013 mean SO4 concentrations of 1.7 to 2.0 μg m−3 imply that future decreases in mean SO4-associated OC concentrations would not exceed ∼ 0.3 to 0.5 μg m−3. Seasonal OC concentrations, largely identified with ozone (O3), vary from 0.3 to 1.4 μg m−3 (∼ 20 % of the total OC concentrations).

Radiative and thermodynamic responses to aerosol extinction profiles during the pre-monsoon month over South Asia

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http://www.atmos-chem-phys.net/16/247/2016/doi:10.5194/acp-16-247-2016

Aerosol radiative effects and thermodynamic responses over South Asia are examined with the Weather Research and Forecasting model coupled with Chemistry (WRF-Chem) for March 2012. Model results of aerosol optical depths (AODs) and extinction profiles are analyzed and compared to satellite retrievals and two ground-based lidars located in northern India. The WRF-Chem model is found to heavily underestimate the AOD during the simulated pre-monsoon month and about 83 % of the model's low bias is due to aerosol extinctions below ∼ 2 km. Doubling the calculated aerosol extinctions below 850 hPa generates much better agreement with the observed AOD and

extinction profiles averaged over South Asia. To separate the effect of absorption and scattering properties, two runs were conducted: in one run (Case I), the calculated scattering and absorption coefficients were increased proportionally, while in the second run (Case II) only the calculated aerosol scattering coefficient was increased. With the same AOD and extinction profiles, the two runs produce significantly different radiative effects over land and oceans. On the regional mean basis, Case I generates 48 % more heating in the atmosphere and 21 % more dimming at the surface than Case II. Case I also produces stronger cooling responses over the land from the longwave radiation adjustment and boundary layer mixing. These rapid adjustments offset the stronger radiative heating in Case I and lead to an overall lower-troposphere cooling up to −0.7 K day−1, which is smaller than that in Case II. Over the ocean, direct radiative effects dominate the heating rate changes in the lower atmosphere lacking such surface and lower atmosphere adjustments due to fixed sea surface temperature, and the strongest atmospheric warming is obtained in Case I. Consequently, atmospheric dynamics (boundary layer heights and meridional circulation) and thermodynamic processes (water vapor and cloudiness) are shown to respond differently between Case I and Case II, underlining the importance of determining the exact portion of scattering or absorbing aerosols that lead to the underestimation of aerosol optical depth in the model. In addition, the model results suggest that both the direct radiative effect and rapid thermodynamic responses need to be quantified for understanding aerosol radiative impacts.

The sensitivities of emissions reductions for the mitigation of UK PM2.5

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Source: *Atmos. Chem. Phys., 16, 265-276, 2016*
http://www.atmos-chem-phys.net/16/265/2016/doi:10.5194/acp-16-265-2016

The reduction of ambient concentrations of fine particulate matter (PM2.5) is a key objective for air pollution control policies in the UK and elsewhere. Long-term exposure to PM2.5 has been identified as a major contributor to adverse human health effects in epidemiological studies and underpins ambient PM2.5 legislation. As a range of emission sources and atmospheric chemistry transport processes contribute to PM2.5 concentrations, atmospheric chemistry transport models are an essential tool to assess emissions control effectiveness. The EMEP4UK atmospheric chemistry transport model was used to investigate the impact of reductions in UK anthropogenic emissions of primary PM2.5, NH3, NOx, SOx or non-methane VOC on surface concentrations of PM2.5 in the UK for a recent year (2010) and for a future current legislation emission (CLE) scenario (2030). In general, the sensitivity to UK mitigation is rather small. A 30 % reduction in UK emissions of any one of the above components yields (for the 2010 simulation) a maximum reduction in PM2.5 in any given location of ∼ 0.6 µg m−3 (equivalent to ∼ 6 % of the modelled PM2.5). On average across the UK, the sensitivity of PM2.5 concentrations to a 30 % reduction in UK emissions of individual contributing components, for both the 2010 and 2030 CLE baselines, increases in the order NMVOC, NOx, SOx, NH3 and primary PM2.5; however there are strong spatial

differences in the PM2.5 sensitivities across the UK. Consequently, the sensitivity of PM2.5 to individual component emissions reductions varies between area and population weighting. Reductions in NH3 have the greatest effect on area-weighted PM2.5. A full UK population weighting places greater emphasis on reductions of primary PM2.5 emissions, which is simulated to be the most effective single-component control on PM2.5 for the 2030 scenario. An important conclusion is that weighting corresponding to the average exposure indicator metric (using data from the 45 model grids containing a monitor whose measurements are used to calculate the UK AEI) further increases the emphasis on the effectiveness of primary PM2.5 emissions reductions (and of NOx emissions reductions) relative to the effectiveness of NH3emissions reductions. Reductions in primary PM2.5 have the largest impact on the AEI in both 2010 and the 2030 CLE scenario. The summation of the modelled reductions to the UK PM2.5 AEI from 30 % reductions in UK emissions of primary PM2.5, NH3, SOx, NOx and VOC totals 1.17 and 0.82 μg m−3 for the 2010 and 2030 CLE simulations, respectively (not accounting for non-linearity)

Radiative and climate impacts of a large volcanic eruption during stratospheric sulfur geoengineering

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Source: *Atmos. Chem. Phys., 16, 305-323, 2016*
http://www.atmos-chem-phys.net/16/305/2016/doi:10.5194/acp-16-305-2016

Both explosive volcanic eruptions, which emit sulfur dioxide into the stratosphere, and stratospheric geoengineering via sulfur injections can potentially cool the climate by increasing the amount of scattering particles in the atmosphere. Here we employ a global aerosol-climate model and an Earth system model to study the radiative and climate changes occurring after an erupting volcano during solar radiation management (SRM). According to our simulations the radiative impacts of the eruption and SRM are not additive and the radiative effects and climate changes occurring after the eruption depend strongly on whether SRM is continued or suspended after the eruption. In the former case, the peak burden of the additional stratospheric sulfate as well as changes in global mean precipitation are fairly similar regardless of whether the eruption takes place in a SRM or non-SRM world. However, the maximum increase in the global mean radiative forcing caused by the eruption is approximately 21 % lower compared to a case when the eruption occurs in an unperturbed atmosphere. In addition, the recovery of the stratospheric sulfur burden and radiative forcing is significantly faster after the eruption, because the eruption during the SRM leads to a smaller number and larger sulfate particles compared to the eruption in a non-SRM world. On the other hand, if SRM is suspended immediately after the eruption, the peak increase in global forcing caused by the eruption is about 32 % lower compared to a corresponding eruption into a clean background atmosphere. In this simulation, only about one-third of the global ensemble-mean cooling occurs after the eruption, compared to that occurring after an eruption under unperturbed atmospheric conditions. Furthermore, the global cooling signal is seen only for

the 12 months after the eruption in the former scenario compared to over 40 months in the latter. In terms of global precipitation rate, we obtain a 36 % smaller decrease in the first year after the eruption and again a clearly faster recovery in the concurrent eruption and SRM scenario, which is suspended after the eruption. We also found that an explosive eruption could lead to significantly different regional climate responses depending on whether it takes place during geoengineering or into an unperturbed background atmosphere. Our results imply that observations from previous large eruptions, such as Mount Pinatubo in 1991, are not directly applicable when estimating the potential consequences of a volcanic eruption during stratospheric geoengineering.

Nitrogen speciation in various types of aerosols in spring over the northwestern Pacific Ocean

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Source: *Atmos. Chem. Phys., 16, 325-341, 2016*
http://www.atmos-chem-phys.net/16/325/2016/ doi:10.5194/acp-16-325-2016

The cumulative atmospheric nitrogen deposition has been found to profoundly impact the nutrient stoichiometry of the eastern China seas (ECSs: the Yellow Sea and East China Sea) and the northwestern Pacific Ocean (NWPO). In spite of the potential significance of dry deposition in those regions, shipboard observations of atmospheric aerosols remain insufficient, particularly regarding the compositions of water-soluble nitrogen species (nitrate, ammonium and water-soluble organic nitrogen – WSON). We conducted a cruise covering the ECSs and the NWPO during the spring of 2014 and observed three types of atmospheric aerosols. Aluminum content, air mass backward trajectories, weather conditions, and ion stoichiometry allowed us to discern dust aerosol patches and sea-fog-modified aerosols (widespread over the ECSs) from background aerosols (open ocean). Among the three types, sea-fog-modified aerosols contained the highest concentrations of nitrate (536 ± 300 nmol N m−3), ammonium (442 ± 194 nmol N m−3) and WSON (147 ± 171 nmol N m−3); furthermore, ammonium and nitrate together occupied ∼ 65 % of the molar fraction of total ions. The dust aerosols also contained significant amounts of nitrate (100 ± 23 nmol N m−3) and ammonium (138 ± 24 nmol N m−3) which were obviously larger than those in the background aerosols (26 ± 32 for nitrate and 54 ± 45 nmol N m−3 for ammonium), yet this was not the case for WSON. It appeared that dust aerosols had less of a chance to come in contact with WSON during their transport. In the open ocean, we found that sea salt (e.g., Na+, Cl−, Mg2+), as well as WSON, correlated positively with wind speed. Apparently, marine dissolved organic nitrogen (DON) was emitted from breaking waves. Regardless of the variable wind speeds from 0.8 to as high as 18 m s−1, nitrate and ammonium, by contrast, remained in narrow ranges, implying that some supply and consumption processes of nitrate and ammonium were required to maintain such a quasi-static condition. Mean dry deposition of total dissolved nitrogen (TDN) for sea-fog-modified aerosols (1090 ± 671 μmol N m−2 d−1) was 5 times higher than that for dust aerosols

(190 ± 41.6 μmol N m−2 d−1) and around 20 times higher than that for background aerosols (56.8 ± 59.1 μmol N m−2 d−1). Apparently, spring sea fog on the ECSs played an important role in removing atmospheric reactive nitrogen from the Chinese mainland and depositing it into the ECSs, thus effectively preventing its seaward export to the NWPO.

Stratospheric ozone change and related climate impacts over 1850–2100 as modelled by the ACCMIP ensemble

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http://www.atmos-chem-phys.net/16/343/2016/ doi:10.5194/acp-16-343-2016

Stratospheric ozone and associated climate impacts in the Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP) simulations are evaluated in the recent past (1980–2000), and examined in the long-term (1850–2100) using the Representative Concentration Pathways (RCPs) low- and high-emission scenarios (RCP2.6 and RCP8.5, respectively) for the period 2000–2100. ACCMIP multi-model mean total column ozone (TCO) trends compare favourably, within uncertainty estimates, against observations. Particularly good agreement is seen in the Antarctic austral spring (−11.9 % dec−1 compared to observed ∼ −13.9 ± 10.4 % dec−1), although larger deviations are found in the Arctic's boreal spring (−2.1 % dec−1 compared to observed ∼ −5.3 ± 3.3 % dec−1). The simulated ozone hole has cooled the lower stratosphere during austral spring in the last few decades (−2.2 K dec−1). This cooling results in Southern Hemisphere summertime tropospheric circulation changes captured by an increase in the Southern Annular Mode (SAM) index (1.3 hPa dec−1). In the future, the interplay between the ozone hole recovery and greenhouse gases (GHGs) concentrations may result in the SAM index returning to pre-ozone hole levels or even with a more positive phase from around the second half of the century (−0.4 and 0.3 hPa dec−1 for the RCP2.6 and RCP8.5, respectively). By 2100, stratospheric ozone sensitivity to GHG concentrations is greatest in the Arctic and Northern Hemisphere midlatitudes (37.7 and 16.1 DU difference between the RCP2.6 and RCP8.5, respectively), and smallest over the tropics and Antarctica continent (2.5 and 8.1 DU respectively). Future TCO changes in the tropics are mainly determined by the upper stratospheric ozone sensitivity to GHG concentrations, due to a large compensation between tropospheric and lower stratospheric column ozone changes in the two RCP scenarios. These results demonstrate how changes in stratospheric ozone are tightly linked to climate and show the benefit of including the processes interactively in climate models.

Global tropospheric ozone variations from 2003 to 2011 as seen by SCIAMACHY

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http://www.atmos-chem-phys.net/16/417/2016/doi:10.5194/acp-16-417-2016

An analysis of the tropospheric ozone (O3) columns (TOCs) derived from SCIAMACHY limb-nadir-matching (LNM) observations during the period 2003–2011, focusing on global variations in TOC, is described. The changes are derived using a multivariate linear regression model. TOC shows changes of −0.2 ± 0.4, 0.3 ± 0.4, 0.1 ± 0.5 and 0.1 ± 0.2 % yr−1, which are not statistically significant at the 2σ level in the latitude bands 30–50° N, 20° S–0, 0–20° N and 50–30° S, respectively. Tropospheric O3 shows statistically significant increases over some regions of South Asia (1–3 % yr−1), the South American continent (up to 2 % yr−1), Alaska (up to 2 % yr−1) and around Congo in Africa (up to 2 % yr−1). Significant increase in TOC is determined off the continents including Australia (up to 2 % yr−1), Eurasia (1–3 % yr−1) and South America (up to 3 % yr−1). Significant decrease in TOC (up to −3 % yr−1) is observed over some regions of the continents of North America, Europe and South America. Over the oceanic regions including the Pacific, North Atlantic and Indian oceans, significant decreases in TOC (−1 to −3 % yr−1) were observed. In addition, the response of the El Niño–Southern Oscillation (ENSO) and quasi-biennial oscillation (QBO) to changes in TOC for the period 2003–2011 was investigated. The result shows extensive regions, mostly in the tropics and Northern Hemisphere extratropics, of significant ENSO responses to changes in TOC and a significant QBO response to TOC changes over some regions.

Fog composition at Baengnyeong Island in the eastern Yellow Sea: detecting markers of aqueous atmospheric oxidations

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http://www.atmos-chem-phys.net/16/437/2016/doi:10.5194/acp-16-437-2016

Samples of fog water were collected at Baengnyeong Island (BYI) in the Yellow Sea during the summer of 2014. The most abundant chemical species in the fog water were NH4+ (mean of 2220 μM), NO3− (1260 μM), SO4−2 (730 μM), and Na+ (551 μM), with substantial contributions from other species consistent with marine and biomass burning influence on some dates. The pH of the samples ranged between 3.48 and 5.00, with a mean of 3.94, intermediate within pH values of fog/cloud water reported previously in Southeast Asia. Back trajectories (72 h) showed that high relative humidity (> 80 %) was encountered upwind of the sampling site by all but one of the sampled air masses, and that the fog composition at BYI can be impacted by several different source regions, including the Sea of Japan, southeastern China, northeastern China, and the East China Sea.

Sulfur in the collected fog was highly oxidized: low S(IV) concentrations were measured (mean of 2.36 μM) in contrast to SO4−2 and in contrast to fog/cloud S(IV) concentrations from pollutant source regions; organosulfate species were also observed and were most likely formed through aging of mainly biogenic volatile organic compounds. Low-molecular-mass organic acids were major contributors to total organic carbon (TOC; 36–69 %), comprising a fraction of TOC at the upper end of that seen in fogs and clouds in other polluted environments. Large contributions were observed from not only acetic and formic acids but also oxalic, succinic, maleic, and other organic acids that can be produced in aqueous atmospheric organic processing (AAOP) reactions. These samples of East Asian fog water containing highly oxidized components represent fog downwind of pollutant sources and can provide new insight into the fate of regional emissions. In particular, these samples demonstrate the result of extensive photochemical aging during multiday transport, including oxidation within wet aerosols and fogs.

Overview of the Chemistry-Aerosol Mediterranean Experiment/Aerosol Direct Radiative Forcing on the Mediterranean Climate (ChArMEx/ADRIMED) summer 2013 campaign

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http://www.atmos-chem-phys.net/16/455/2016/doi:10.5194/acp-16-455-2016

The Chemistry-Aerosol Mediterranean Experiment (ChArMEx; http://charmex.lsce.ipsl.fr) is a collaborative research program federating international activities to investigate Mediterranean regional chemistry-climate interactions. A special observing period (SOP-1a) including intensive airborne measurements was performed in the framework of the Aerosol Direct Radiative Impact on the regional climate in the MEDiterranean region (ADRIMED) project during the Mediterranean dry season over the western and central Mediterranean basins, with a focus on aerosol-radiation measurements and their modeling. The SOP-1a took place from 11 June to 5 July 2013. Airborne measurements were made by both the ATR-42 and F-20 French research aircraft operated from Sardinia (Italy) and instrumented for in situ and remote-sensing measurements, respectively, and by sounding and drifting balloons, launched in Minorca. The experimental setup also involved several ground-based measurement sites on islands including two ground-based reference stations

in Corsica and Lampedusa and secondary monitoring sites in Minorca and Sicily. Additional measurements including lidar profiling were also performed on alert during aircraft operations at EARLINET/ACTRIS stations at Granada and Barcelona in Spain, and in southern Italy. Remote-sensing aerosol products from satellites (MSG/SEVIRI, MODIS) and from the AERONET/PHOTONS network were also used. Dedicated meso-scale and regional modeling experiments were performed in relation to this observational effort. We provide here an overview of the different surface and aircraft observations deployed during the ChArMEx/ADRIMED period and of associated modeling studies together with an analysis of the synoptic conditions that determined the aerosol emission and transport. Meteorological conditions observed during this campaign (moderate temperatures and southern flows) were not favorable to producing high levels of atmospheric pollutants or intense biomass burning events in the region. However, numerous mineral dust plumes were observed during the campaign, with the main sources located in Morocco, Algeria and Tunisia, leading to aerosol optical depth (AOD) values ranging between 0.2 and 0.6 (at 440 nm) over the western and central Mediterranean basins. One important point of this experiment concerns the direct observations of aerosol extinction onboard the ATR-42, using the CAPS system, showing local maxima reaching up to 150 M m−1 within the dust plume. Non-negligible aerosol extinction (about 50 M m−1) has also been observed within the marine boundary layer (MBL). By combining the ATR-42 extinction coefficient observations with absorption and scattering measurements, we performed a complete optical closure revealing excellent agreement with estimated optical properties. This additional information on extinction properties has allowed calculation of the dust single scattering albedo (SSA) with a high level of confidence over the western Mediterranean. Our results show a moderate variability from 0.90 to 1.00 (at 530 nm) for all flights studied compared to that reported in the literature on this optical parameter. Our results underline also a relatively low difference in SSA with values derived near dust sources. In parallel, active remote-sensing observations from the surface and onboard the F-20 aircraft suggest a complex vertical structure of particles and distinct aerosol layers with sea spray and pollution located within the MBL, and mineral dust and/or aged North American smoke particles located above (up to 6–7 km in altitude). Aircraft and balloon-borne observations allow one to investigate the vertical structure of the aerosol size distribution showing particles characterized by a large size (> 10 μm in diameter) within dust plumes. In most of cases, a coarse mode characterized by an effective diameter ranging between 5 and 10 μm, has been detected above the MBL. In terms of shortwave (SW) direct forcing, in situ surface and aircraft observations have been merged and used as inputs in 1-D radiative transfer codes for calculating the aerosol direct radiative forcing (DRF). Results show significant surface SW instantaneous forcing (up to −90 W m−2 at noon). Aircraft observations provide also original estimates of the vertical structure of SW and LW radiative heating revealing significant instantaneous values of about 5° K per day in the solar spectrum (for a solar angle of 30°) within the dust layer. Associated 3-D modeling studies from regional climate (RCM) and chemistry transport (CTM) models indicate a relatively good agreement for simulated AOD compared with observations from the AERONET/PHOTONS network and satellite data, especially for long-range dust transport. Calculations of the 3-D SW (clear-sky) surface DRF indicate an average of about −10 to −20 W m−2 (for the whole period) over the Mediterranean Sea together with maxima (−50 W m−2) over northern Africa. The top of the atmosphere (TOA) DRF is shown to be highly variable within the domain, due to moderate absorbing properties of dust and changes in the surface albedo.

Indeed, 3-D simulations indicate negative forcing over the Mediterranean Sea and Europe and positive forcing over northern Africa. Finally, a multi-year simulation, performed for the 2003 to 2009 period and including an ocean–atmosphere (O–A) coupling, underlines the impact of the aerosol direct radiative forcing on the sea surface temperature, O–A fluxes and the hydrological cycle over the Mediterranean.

Aerosol properties, source identification, and cloud processing in orographic clouds measured by single particle mass spectrometry on a central European mountain site during HCCT-2010

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http://www.atmos-chem-phys.net/16/505/2016/doi:10.5194/acp-16-505-2016

Cloud residues and out-of-cloud aerosol particles with diameters between 150 and 900 nm were analysed by online single particle aerosol mass spectrometry during the 6-week study Hill Cap Cloud Thuringia (HCCT)-2010 in September–October 2010. The measurement location was the mountain Schmücke (937 m a.s.l.) in central Germany. More than 160 000 bipolar mass spectra from out-of-cloud aerosol particles and more than 13 000 bipolar mass spectra from cloud residual particles were obtained and were classified using a fuzzy c-means clustering algorithm. Analysis of the uncertainty of the sorting algorithm was conducted on a subset of the data by comparing the clustering output with particle-by-particle inspection and classification by the operator. This analysis yielded a false classification probability between 13 and 48 %. Additionally, particle types were identified by specific marker ions. The results from the ambient aerosol analysis show that 63 % of the analysed particles belong to clusters having a diurnal variation, suggesting that local or regional sources dominate the aerosol, especially for particles containing soot and biomass burning particles. In the cloud residues, the relative percentage of large soot-containing particles and particles containing amines was found to be increased compared to the out-of-cloud aerosol, while, in general, organic particles were less abundant in the cloud residues. In the case of amines, this can be explained by the high solubility of the amines, while the large soot-containing particles were found to be internally mixed with inorganics, which explains their activation as cloud condensation nuclei. Furthermore, the results show that during cloud processing, both sulfate and nitrate are added to the residual particles, thereby changing the mixing state and increasing the fraction of particles with nitrate and/or sulfate. This is expected to lead to higher hygroscopicity after cloud evaporation, and therefore to an increase of the particles' ability to act as cloud condensation nuclei after their cloud passage.

Sources of nitrogen deposition in Federal Class I areas in the US

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It is desired to control excessive reactive nitrogen (Nr) deposition due to its detrimental impact on ecosystems. Using a three-dimensional atmospheric chemical transport model, GEOS-Chem, Nr deposition in the contiguous US and eight selected Class I areas (Voyageurs (VY), Smoky Mountain (SM), Shenandoah (SD), Big Bend (BB), Rocky Mountain (RM), Grand Teton (GT), Joshua Tree (JT), and Sequoia (SQ)) is investigated. First, modeled Nr deposition is compared with National Trends Network (NTN) and Clean Air Status and Trends Network (CASTNET) deposition values. The seasonality of measured species is generally well represented by the model ($R2 > 0.6$), except in JT. While modeled Nr is generally within the range of seasonal observations, large overestimates are present in sites such as SM and SD in the spring and summer (up to 0.6 kg N ha month−1), likely owing to model high-biases in surface HNO3. The contribution of non-measured species (mostly dry deposition of NH3) to total modeled Nr deposition ranges from 1 to 55 %. The spatial distribution of the origin of Nr deposited in each Class I area and the contributions of individual emission sectors are estimated using the GEOS-Chem adjoint model. We find the largest role of long-range transport for VY, where 50 % (90 %) of annual Nr deposition originates within 670 (1670) km of the park. In contrast, the Nr emission footprint is most localized for SQ, where 50 % (90 %) of the deposition originates from within 130 (370) km. Emissions from California contribute to the Nr deposition in remote areas in the western US (RM, GT). Mobile NOx and livestock NH3 are found to be the major sources of Nr deposition in all sites except BB, where contributions of NOx from lightning and soils to natural levels of Nr deposition are significant (~ 40 %). The efficiency in terms of Nr deposition per kg emissions of NH3-N, NOx-N, and SO2-S are also estimated. Unique seasonal features are found in JT (opposing efficiency distributions for winter and summer), RM (large fluctuations in the range of effective regions), and SD (upwind NH3 emissions hindering Nr deposition). We also evaluate the contributions of emissions to the total area of Class I regions in critical load exceedance, and to the total magnitude of exceedance. We find that while it is effective to control emissions in the western US to reduce the area of regions in CL exceedance, it can be more effective to control emissions in the eastern US to reduce the magnitude of Nr deposition above the CL. Finally, uncertainty in the nitrogen deposition caused by uncertainty in the NH3 emission inventory is explored by comparing results based on two different NH3 inventories; noticeable differences in the emission inventories and thus sensitivities of up to a factor of four found in individual locations.

Possible influence of atmospheric circulations on winter haze pollution in the Beijing–Tianjin–Hebei region, northern China

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Using the daily records derived from the synoptic weather stations and the NCEP/NCAR and ERA-Interim reanalysis data, the variability of the winter haze pollution (indicated by the mean visibility and number of hazy days) in the Beijing–Tianjin–Hebei (BTH) region during the period 1981 to 2015 and its relationship with the atmospheric circulations at middle–high latitude were analyzed in this study. The winter haze pollution in BTH had distinct inter-annual and inter-decadal variabilities without a significant long-term trend. According to the spatial distribution of correlation coefficients, six atmospheric circulation indices (I1 to I6) were defined from the key areas in sea level pressure (SLP), zonal and meridional winds at 850 hPa (U850, V850), geopotential height field at 500 hPa (H500), zonal wind at 200 hPa (U200), and air temperature at 200 hPa (T200), respectively. All of the six indices have significant and stable correlations with the winter visibility and number of hazy days in BTH. In the raw (unfiltered) correlations, the correlation coefficients between the six indices and the winter visibility (number of hazy days) varied from 0.57 (0.47) to 0.76 (0.6) with an average of 0.65 (0.54); in the high-frequency (< 10 years) correlations, the coefficients varied from 0.62 (0.58) to 0.8 (0.69) with an average of 0.69 (0.64). The six circulation indices together can explain 77.7 % (78.7 %) and 61.7 % (69.1 %) variances of the winter visibility and the number of hazy days in the year-to-year (inter-annual) variability, respectively. The increase in Ic (a comprehensive index derived from the six individual circulation indices) can cause a shallowing of the East Asian trough at the middle troposphere and a weakening of the Siberian high-pressure field at sea level, and is then accompanied by a reduction (increase) of horizontal advection and vertical convection (relative humidity) in the lowest troposphere and a reduced boundary layer height in BTH and its neighboring areas, which are favorable for the formation of haze pollution in BTH winter, and vice versa. The high level of the prediction statistics and the reasonable mechanism suggested that the winter haze pollution in BTH can be forecasted or estimated credibly based on the optimized atmospheric circulation indices. Thus it is helpful for government decision-making departments to take action in advance in dealing with probably severe haze pollution in BTH indicated by the atmospheric circulation conditions.

Reactive nitrogen partitioning and its relationship to winter ozone events in Utah

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http://www.atmos-chem-phys.net/16/573/2016/doi:10.5194/acp-16-573-2016

High wintertime ozone levels have been observed in the Uintah Basin, Utah, a sparsely populated rural region with intensive oil and gas operations. The reactive nitrogen budget plays an important role in tropospheric ozone formation. Measurements were taken during three field campaigns in the winters of 2012, 2013 and 2014, which experienced varying climatic conditions. Average concentrations of ozone and total reactive nitrogen were observed to be 2.5 times higher in 2013 than 2012, with 2014 an intermediate year in most respects. However, photochemically active NOx (NO + NO2) remained remarkably similar all three years. Nitric acid comprised roughly half of NOz (≡ NOy – NOx) in 2013, with nighttime nitric acid formation through heterogeneous uptake of N2O5 contributing approximately 6 times more than daytime formation. In 2012, N2O5 and ClNO2 were larger components of NOz relative to HNO3. The nighttime N2O5 lifetime between the high-ozone year 2013 and the low-ozone year 2012 is lower by a factor of 2.6, and much of this is due to higher aerosol surface area in the high-ozone year of 2013. A box-model simulation supports the importance of nighttime chemistry on the reactive nitrogen budget, showing a large sensitivity of NOx and ozone concentrations to nighttime processes.

A sub-decadal trend in diacids in atmospheric aerosols in eastern Asia

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http://www.atmos-chem-phys.net/16/585/2016/doi:10.5194/acp-16-585-2016

Change in secondary organic aerosols (SOAs) has been predicted to be highly uncertain in the future atmosphere in Asia. To better quantify the SOA change, we examine the sub-decadal (2001–2008) trend in major surrogate compounds (C2–C10 diacids) of SOA in atmospheric aerosols from Gosan site on Cheju Island, South Korea. The Gosan site is influenced by pollution outflows from eastern Asia. The molecular distributions of diacids were characterized by the predominance of oxalic (C2) acid followed by malonic (C3) and succinic (C4) acids in each year. The seasonal variations in diacids in each year were characterized by the highest concentrations of saturated diacids in spring and unsaturated diacids in winter. The consistent molecular distributions and seasonal variations along with significantly similar air mass transport patterns are indicative of similar pollution sources for diacids in eastern Asia on a sub-decadal scale. However, the intensity of the pollution sources has increased as evidenced by the increases in major diacids at the rate of 3.9–47.4 % per year, particularly in April. The temporal variations in atmospheric tracer compounds (carbon monoxide, levoglucosan, 2-methyltetrols, pinic acid, glyoxylic acid, glyoxal and

methylglyoxal) suggest that the increases in diacids are due to enhanced precursor emissions associated with more anthropogenic than biogenic activities followed by the compounds' chemical processing in the atmosphere. The trends in diacids contrast with the reported decreases in sulfate, nitrate and ammonium in recent years in eastern Asia. This study demonstrates that recent pollution control strategies in eastern Asia were not able to decrease organic acidic species in the atmosphere. The increases in water-soluble organic acid fraction could modify the aerosol organic composition and its sensitivity to climate relevant physical properties.

Fine particulate matter in the tropical environment: monsoonal effects, source apportionment, and health risk assessment

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Source: *Atmos. Chem. Phys., 16, 597-617, 2016*
http://www.atmos-chem-phys.net/16/597/2016/doi:10.5194/acp-16-597-2016

The health implications of PM2.5 in the tropical region of Southeast Asia (SEA) are significant as PM2.5 can pose serious health concerns. PM2.5 concentration and sources here are strongly influenced by changes in the monsoon regime from the south-west quadrant to the north-east quadrant in the region. In this work, PM2.5 samples were collected at a semi-urban area using a high-volume air sampler at different seasons on 24 h basis. Analysis of trace elements and water-soluble ions was performed using inductively coupled plasma mass spectroscopy (ICP-MS) and ion chromatography (IC), respectively. Apportionment analysis of PM2.5 was carried out using the United States Environmental Protection Agency (US EPA) positive matrix factorization (PMF) 5.0 and a mass closure model. We quantitatively characterized the health risks posed to human populations through the inhalation of selected heavy metals in PM2.5. 48 % of the samples collected exceeded the World Health Organization (WHO) 24 h PM2.5 guideline but only 19 % of the samples exceeded 24 h US EPA National Ambient Air Quality Standard (NAAQS). The PM2.5 concentration was slightly higher during the north-east monsoon compared to south-west monsoon. The main trace metals identified were As, Pb, Cd, Ni, Mn, V, and Cr while the main ions were SO42−, NO3−, NH4+, and Na. The mass closure model identified four major sources of PM2.5 that account for 55 % of total mass balance. The four sources are mineral matter (MIN) (35 %), secondary inorganic aerosol (SIA) (11 %), sea salt (SS) (7 %), and trace elements (TE) (2 %). PMF 5.0 elucidated five potential sources: motor vehicle emissions coupled with biomass burning (31 %) were the most dominant, followed by marine/sulfate aerosol (20 %), coal burning (19 %), nitrate aerosol (17 %), and mineral/road dust (13 %). The hazard quotient (HQ) for four selected metals (Pb, As, Cd, and Ni) in PM2.5 mass was highest in PM2.5 mass from the coal burning source and least in PM2.5 mass originating from the mineral/road dust source. The main carcinogenic heavy metal of concern to health at the current location was As; the other heavy metals (Ni, Pb, and Cd) did not pose a significant cancer risk in PM2.5 mass concentration. Overall, the associated lifetime cancer risk posed by the exposure of hazardous metals in PM2.5 is 3–4 per 1 000 000 people at this location.

Formation of secondary aerosols from gasoline vehicle exhaust when mixing with SO2

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Source: *Atmos. Chem. Phys., 16, 675-689, 2016*
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Sulfur dioxide (SO2) can enhance the formation of secondary aerosols from biogenic volatile organic compounds (VOCs), but its influence on secondary aerosol formation from anthropogenic VOCs, particularly complex mixtures like vehicle exhaust, remains uncertain. Gasoline vehicle exhaust (GVE) and SO2, a typical pollutant from coal burning, are directly co-introduced into a smog chamber, in this study, to investigate the formation of secondary organic aerosols (SOA) and sulfate aerosols through photooxidation. New particle formation was enhanced, while substantial sulfate was formed through the oxidation of SO2 in the presence of high concentration of SO2. Homogenous oxidation by OH radicals contributed a negligible fraction to the conversion of SO2 to sulfate, and instead the oxidation by stabilized Criegee intermediates (sCIs), formed from alkenes in the exhaust reacting with ozone, dominated the conversion of SO2. After 5 h of photochemical aging, GVE's SOA production factor revealed an increase by 60–200 % in the presence of high concentration of SO2. The increase could principally be attributed to acid-catalyzed SOA formation as evidenced by the strong positive linear correlation ($R2 = 0.97$) between the SOA production factor and in situ particle acidity calculated by the AIM-II model. A high-resolution time-of-flight aerosol mass spectrometer (HR-TOF-AMS) resolved OA's relatively lower oxygen-to-carbon (O : C) (0.44 ± 0.02) and higher hydrogen-to-carbon (H : C) (1.40 ± 0.03) molar ratios for the GVE / SO2 mixture, with a significantly lower estimated average carbon oxidation state (OSc) of -0.51 ± 0.06 than -0.19 ± 0.08 for GVE alone. The relative higher mass loading of OA in the experiments with SO2 might be a significant explanation for the lower SOA oxidation degree.

Aircraft-measured indirect cloud effects from biomass burning smoke in the Arctic and subarctic

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http://www.atmos-chem-phys.net/16/715/2016/doi:10.5194/acp-16-715-2016

The incidence of wildfires in the Arctic and subarctic is increasing; in boreal North America, for example, the burned area is expected to increase by 200–300 % over the next 50–100 years, which previous studies suggest could have a large effect on cloud microphysics, lifetime, albedo, and

precipitation. However, the interactions between smoke particles and clouds remain poorly quantified due to confounding meteorological influences and remote sensing limitations. Here, we use data from several aircraft campaigns in the Arctic and subarctic to explore cloud microphysics in liquid-phase clouds influenced by biomass burning. Median cloud droplet radii in smoky clouds were ∼ 40–60 % smaller than in background clouds. Based on the relationship between cloud droplet number (Nliq) and various biomass burning tracers (BBt) across the multi-campaign data set, we calculated the magnitude of subarctic and Arctic smoke aerosol–cloud interactions (ACIs, where ACI = (1⁄3) × dln(Nliq)⁄dln(BBt)) to be ∼ 0.16 out of a maximum possible value of 0.33 that would be obtained if all aerosols were to nucleate cloud droplets. Interestingly, in a separate subarctic case study with low liquid water content (∼ 0.02 g m−3) and very high aerosol concentrations (2000–3000 cm−3) in the most polluted clouds, the estimated ACI value was only 0.05. In this case, competition for water vapor by the high concentration of cloud condensation nuclei (CCN) strongly limited the formation of droplets and reduced the cloud albedo effect, which highlights the importance of cloud feedbacks across scales. Using our calculated ACI values, we estimate that the smoke-driven cloud albedo effect may decrease local summertime short-wave radiative flux by between 2 and 4 W m−2 or more under some low and homogeneous cloud cover conditions in the subarctic, although the changes should be smaller in high surface albedo regions of the Arctic. We lastly explore evidence suggesting that numerous northern-latitude background Aitken particles can interact with combustion particles, perhaps impacting their properties as cloud condensation and ice nuclei.

The impact of shipping emissions on air pollution in the greater North Sea region – Part 1: Current emissions and concentrations

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http://www.atmos-chem-phys.net/16/739/2016/doi:10.5194/acp-16-739-2016

The North Sea is one of the areas with the highest ship traffic densities worldwide. At any time, about 3000 ships are sailing its waterways. Previous scientific publications have shown that ships contribute significantly to atmospheric concentrations of NOx, particulate matter and ozone. Especially in the case of particulate matter and ozone, this influence can even be seen in regions far away from the main shipping routes. In order to quantify the effects of North Sea shipping on air quality in its bordering states, it is essential to determine the emissions from shipping as accurately as possible. Within Interreg IVb project Clean North Sea Shipping (CNSS), a bottom-up approach was developed and used to thoroughly compile such an emission inventory for 2011 that served as the base year for the current emission situation. The innovative aspect of this approach was to use load-dependent functions to calculate emissions from the ships' current activities instead of averaged emission factors for the entire range of the engine loads. These functions were applied to ship activities that were derived from hourly records of Automatic Identification System signals together with a database containing the engine characteristics of the vessels that traveled the North

Sea in 2011. The emission model yielded ship emissions among others of NOx and SO2 at high temporal and spatial resolution that were subsequently used in a chemistry transport model in order to simulate the impact of the emissions on pollutant concentration levels. The total emissions of nitrogen reached 540 Gg and those of sulfur oxides 123 Gg within the North Sea – including the adjacent western part of the Baltic Sea until 5° W. This was about twice as much of those of a medium-sized industrialized European state like the Netherlands. The relative contribution of ships to, for example, NO2 concentration levels ashore close to the sea can reach up to 25 % in summer and 15 % in winter. Some hundred kilometers away from the sea, the contribution was about 6 % in summer and 4 % in winter. The relative contribution of the secondary pollutant NO3− was found to reach 20 % in summer and 6 % in winter even far from the shore.

Impact of vehicular emissions on the formation of fine particles in the Sao Paulo Metropolitan Area: a numerical study with the WRF-Chem model

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***Source**: Atmos. Chem. Phys., 16, 777-797, 2016*
http://www.atmos-chem-phys.net/16/777/2016/doi:10.5194/acp-16-777-2016

The objective of this work is to evaluate the impact of vehicular emissions on the formation of fine particles (PM2.5; ≤ 2.5 μm in diameter) in the Sao Paulo Metropolitan Area (SPMA) in Brazil, where ethanol is used intensively as a fuel in road vehicles. The Weather Research and Forecasting with Chemistry (WRF-Chem) model, which simulates feedbacks between meteorological variables and chemical species, is used as a photochemical modelling tool to describe the physico-chemical processes leading to the evolution of number and mass size distribution of particles through gas-to-particle conversion. A vehicular emission model based on statistical information of vehicular activity is applied to simulate vehicular emissions over the studied area. The simulation has been performed for a 1-month period (7 August–6 September 2012) to cover the availability of experimental data from the NUANCE-SPS (Narrowing the Uncertainties on Aerosol and Climate Changes in Sao Paulo State) project that aims to characterize emissions of atmospheric aerosols in the SPMA. The availability of experimental measurements of atmospheric aerosols and the application of the WRF-Chem model made it possible to represent some of the most important properties of fine particles in the SPMA such as the mass size distribution and chemical composition, besides allowing us to evaluate its formation potential through the gas-to-particle conversion processes. Results show that the emission of primary gases, mostly from vehicles, led to a production of secondary particles between 20 and 30 % in relation to the total mass concentration of PM2.5 in the downtown SPMA. Each of PM2.5 and primary natural aerosol (dust and sea salt) contributed with 40–50 % of the total PM10 (i.e. those ≤ 10 μm in diameter) concentration. Over 40 % of the formation of fine particles, by mass, was due to the emission of hydrocarbons, mainly aromatics. Furthermore, an increase in the number of small particles impaired the ultraviolet radiation and induced a decrease in ozone formation. The ground-level

O3 concentration decreased by about 2 % when the aerosol-radiation feedback is taken into account.

The impact of residential combustion emissions on atmospheric aerosol, human health, and climate

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http://www.atmos-chem-phys.net/16/873/2016/doi:10.5194/acp-16-873-2016

Combustion of fuels in the residential sector for cooking and heating results in the emission of aerosol and aerosol precursors impacting air quality, human health, and climate. Residential emissions are dominated by the combustion of solid fuels. We use a global aerosol microphysics model to simulate the impact of residential fuel combustion on atmospheric aerosol for the year 2000. The model underestimates black carbon (BC) and organic carbon (OC) mass concentrations observed over Asia, Eastern Europe, and Africa, with better prediction when carbonaceous emissions from the residential sector are doubled. Observed seasonal variability of BC and OC concentrations are better simulated when residential emissions include a seasonal cycle. The largest contributions of residential emissions to annual surface mean particulate matter (PM2.5) concentrations are simulated for East Asia, South Asia, and Eastern Europe. We use a concentration response function to estimate the human health impact due to long-term exposure to ambient PM2.5 from residential emissions. We estimate global annual excess adult (> 30 years of age) premature mortality (due to both cardiopulmonary disease and lung cancer) to be 308 000 (113 300–497 000, 5th to 95th percentile uncertainty range) for monthly varying residential emissions and 517 000 (192 000–827 000) when residential carbonaceous emissions are doubled. Mortality due to residential emissions is greatest in Asia, with China and India accounting for 50 % of simulated global excess mortality. Using an offline radiative transfer model we estimate that residential emissions exert a global annual mean direct radiative effect between −66 and +21 mW m−2, with sensitivity to the residential emission flux and the assumed ratio of BC, OC, and SO2 emissions. Residential emissions exert a global annual mean first aerosol indirect effect of between −52 and −16 mW m−2, which is sensitive to the assumed size distribution of carbonaceous emissions. Overall, our results demonstrate that reducing residential combustion emissions would have substantial benefits for human health through reductions in ambient PM2.5 concentrations.

Historic records of organic compounds from a high Alpine glacier: influences of biomass burning, anthropogenic emissions, and dust transport

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Historic records of α-dicarbonyls (glyoxal, methylglyoxal), carboxylic acids (C6–C12 dicarboxylic acids, pinic acid, p-hydroxybenzoic acid, phthalic acid, 4-methylphthalic acid), and ions (oxalate, formate, calcium) were determined with annual resolution in an ice core from Grenzgletscher in the southern Swiss Alps, covering the time period from 1942 to 1993. Chemical analysis of the organic compounds was conducted using ultra-high-performance liquid chromatography (UHPLC) coupled to electrospray ionization high-resolution mass spectrometry (ESI-HRMS) for dicarbonyls and long-chain carboxylic acids and ion chromatography for short-chain carboxylates. Long-term records of the carboxylic acids and dicarbonyls, as well as their source apportionment, are reported for western Europe. This is the first study comprising long-term trends of dicarbonyls and long-chain dicarboxylic acids (C6–C12) in Alpine precipitation. Source assignment of the organic species present in the ice core was performed using principal component analysis. Our results suggest biomass burning, anthropogenic emissions, and transport of mineral dust to be the main parameters influencing the concentration of organic compounds. Ice core records of several highly correlated compounds (e.g., p-hydroxybenzoic acid, pinic acid, pimelic, and suberic acids) can be related to the forest fire history in southern Switzerland. P-hydroxybenzoic acid was found to be the best organic fire tracer in the study area, revealing the highest correlation with the burned area from fires. Historical records of methylglyoxal, phthalic acid, and dicarboxylic acids adipic acid, sebacic acid, and dodecanedioic acid are comparable with that of anthropogenic emissions of volatile organic compounds (VOCs). The small organic acids, oxalic acid and formic acid, are both highly correlated with calcium, suggesting their records to be affected by changing mineral dust transport to the drilling site.

High-resolution inventory of ammonia emissions from agricultural fertilizer in China from 1978 to 2008

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Source: *Atmos. Chem. Phys., 16, 1207-1218, 2016*
http://www.atmos-chem-phys.net/16/1207/2016/doi:10.5194/acp-16-1207-2016

The quantification of ammonia (NH3) emissions is essential to the more accurate quantification of atmospheric nitrogen deposition, improved air quality and the assessment of ammonia-related agricultural policy and climate mitigation strategies. The quantity, geographic distribution and historical trends of these emissions remain largely uncertain. In this paper, a new Chinese agricultural fertilizer NH3(CAF_NH3) emissions inventory has been compiled that exhibits the following improvements: (1) a 1 × 1 km gridded map on the county level was developed for 2008; (2) a combined bottom-up and top-down method was used for the local correction of emission factors (EFs) and parameters; (3) the temporal patterns of historical time trends for 1978–2008 were estimated and the uncertainties were quantified for the inventories; and (4) a sensitivity test was performed in which a province-level disaggregated map was compared with CAF_NH3 emissions for 2008. The total CAF_NH3 emissions for 2008 were 8.4 TgNH3 yr−1 (a 6.6–9.8 Tg interquartile range). From 1978 to 2008, annual NH3 emissions fluctuated with three peaks (1987, 1996 and 2005), and total emissions increased from 3.2 to 8.4 Tg at an annual rate of 3.0 %. During the study period, the contribution of livestock manure spreading increased from 37.0 to 45.5 % because of changing fertilization practices and the rapid increase in egg, milk, and meat consumption. The average contribution of synthetic fertilizer, which has a positive effect on crop yields, was approximately 38.3 % (minimum: 33.4 %; maximum: 42.7 %). With rapid urbanization causing a decline in the rural population, the contribution of the rural excrement sector varied widely between 20.3 % and 8.5 %. The average contributions of cake fertilizer and straw returning were approximately 3.8 and 4.5 %, respectively, thus small and stable. Collectively, the CAF_NH3 emissions reflect the nation's agricultural policy to a certain extent. An effective approach to decreasing PM2.5 concentrations in China would be to simultaneously decrease NOx, SO2, and NH3 emissions.

On the long-term impact of emissions from central European cities on regional air quality

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http://www.atmos-chem-phys.net/16/1331/2016/doi:10.5194/acp-16-1331-2016

For the purpose of qualifying and quantifying the impact of urban emission from Central European cities on the present-day regional air quality, the regional climate model RegCM4.2 was coupled with the chemistry transport model CAMx, including two-way interactions. A series of simulations was carried out for the 2001–2010 period either with all urban emissions included (base case) or without considering urban emissions. Further, the sensitivity of ozone production to urban emissions was examined by performing reduction experiments with −20 % emission perturbation of NOx and/or non-methane volatile organic compounds (NMVOC).

The modeling system's air quality related outputs were evaluated using AirBase, and EMEP surface measurements showed reasonable reproduction of the monthly variation for ozone (O3), but the

annual cycle of nitrogen dioxide (NO2) and sulfur dioxide (SO2) is more biased. In terms of hourly correlations, values achieved for ozone and NO2 are 0.5–0.8 and 0.4–0.6, but SO2 is poorly or not correlated at all with measurements (r around 0.2–0.5). The modeled fine particulates (PM2.5) are usually underestimated, especially in winter, mainly due to underestimation of nitrates and carbonaceous aerosols.

European air quality measures were chosen as metrics describing the cities emission impact on regional air pollution. Due to urban emissions, significant ozone titration occurs over cities while over rural areas remote from cities, ozone production is modeled, mainly in terms of number of exceedances and accumulated exceedances over the threshold of 40 ppbv. Urban NOx, SO2 and PM2.5 emissions also significantly contribute to concentrations in the cities themselves (up to 50–70 % for NOx and SO2, and up to 60 % for PM2.5), but the contribution is large over rural areas as well (10–20 %). Although air pollution over cities is largely determined by the local urban emissions, considerable (often a few tens of %) fraction of the concentration is attributable to other sources from rural areas and minor cities. For the case of Prague (Czech Republic capital), it is further shown that the inter-urban interference between large cities does not play an important role which means that the impact on a chosen city of emissions from all other large cities is very small. At last, it is shown that to achieve significant ozone reduction over cities in central Europe, the emission control strategies have to focus on the reduction of NMVOC, as reducing NOx (due to suppressed titration) often leads to increased O3. The influence over rural areas is however always in favor of improved air quality, i.e. both NOx and/or NMVOC reduction ends up in decreased ozone pollution, mainly in terms of exceedances.

PLAM – a meteorological pollution index for air quality and its applications in fog-haze forecasts in North China

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Source: Using surface meteorological observation and high-resolution emission data, this paper discusses the application of the PLAM/h index (Parameter Linking Air-quality to Meteorological conditions/haze) in the prediction of large-scale low visibility and fog-haze events. Based on the two-dimensional probability density function diagnosis model for emissions, the study extends the diagnosis and prediction of the meteorological pollution index PLAM to the regional visibility fog-haze intensity. The results show that combining the influence of regular meteorological conditions and emission factors together in the PLAM/h parameterization scheme is very effective in improving the diagnostic identification ability of the fog-haze weather in North China. The determination coefficients for four seasons (spring, summer, autumn, and winter) between PLAM/h and visibility observation are 0.76, 0.80, 0.96, and 0.86, respectively, and all of their significance levels exceed 0.001, showing the ability of PLAM/h to predict the seasonal changes and differences of fog-haze weather in the North China region. The high-value correlation zones are located in Jing-Jin-Ji (Beijing, Tianjin, Hebei), Bohai Bay rim, and southern Hebei–northern Henan, indicating that

the PLAM/h index is related to the distribution of frequent heavy fog-haze weather in North China and the distribution of emission high-value zone. Through comparative analysis of the heavy fog-haze events and large-scale clear-weather processes in winter and summer, it is found that PLAM/h index 24 h forecast is highly correlated with the visibility observation. Therefore, the PLAM/h index has good capability in identification, analysis, and forecasting.

Climate modulation of the Tibetan Plateau on haze in China

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Source: *Atmos. Chem. Phys., 16, 1365-1375, 2016*
http://www.atmos-chem-phys.net/16/1365/2016/doi:10.5194/acp-16-1365-2016

Rapid increases in pollutant emissions in conjunction with stagnant meteorological conditions result in haze pollution in China. Recent frequent haze in China has attracted worldwide attention. Here we show a relationship between the haze events and Tibetan Plateau (TP)'s environment and climate changes. Based on observational data taken over recent decades, we identify central-eastern China (CEC) as a climatological large-scale “susceptible region” of frequent haze, which is harbored by the TP with its impact on midlatitude westerly winds. The observational and modeling studies demonstrate that the interannual variations in the thermal forcing of TP are positively correlated with the incidences of wintertime haze over CEC. Further analysis indicates that the climate warming of the TP induced changes in atmospheric circulation, driving frequent haze events in CEC. The frequent haze occurrences in CEC are consistent with decreasing winter monsoon winds, intensifying downward air flows and increasing atmospheric stability in the lower troposphere over the CEC in association with upstream plateau's thermal anomalies. Therefore, variations of haze in China are related to mechanical and thermal forcing by the TP. Our results also suggest that implications of the large TP topography for environment and climate changes should be taken into account for air pollution mitigation policies in China.

Oil and gas impacts on air quality in federal lands in the Bakken region: an overview of the Bakken Air Quality Study and first results

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Source: The Bakken formation contains billions of barrels of oil and gas trapped in rock and shale. Horizontal drilling and hydraulic fracturing methods have allowed for extraction of these resources, leading to exponential growth of oil production in the region over the past decade. Along with this

development has come an increase in associated emissions to the atmosphere. Concern about potential impacts of these emissions on federal lands in the region prompted the National Park Service to sponsor the Bakken Air Quality Study over two winters in 2013–2014. Here we provide an overview of the study and present some initial results aimed at better understanding the impact of local oil and gas emissions on regional air quality. Data from the study, along with long-term monitoring data, suggest that while power plants are still an important emissions source in the region, emissions from oil and gas activities are impacting ambient concentrations of nitrogen oxides and black carbon and may dominate recent observed trends in pollutant concentrations at some of the study sites. Measurements of volatile organic compounds also definitively show that oil and gas emissions were present in almost every air mass sampled over a period of more than 4 months.

Light absorption of brown carbon aerosol in the PRD region of China

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http://www.atmos-chem-phys.net/16/1433/2016/doi:10.5194/acp-16-1433-2016

The strong spectral dependence of light absorption of brown carbon (BrC) aerosol is regarded to influence aerosol's radiative forcing significantly. The Absorption Angstrom Exponent (AAE) method has been widely used in previous studies to attribute light absorption of BrC at shorter wavelengths for ambient aerosols, with a theoretical assumption that the AAE of "pure" black carbon (BC) aerosol equals to 1.0. In this study, the AAE method was applied to both urban and rural environments in the Pearl River Delta (PRD) region of China, with an improvement of constraining the realistic AAE of "pure" BC through statistical analysis of on-line measurement data. A three-wavelength photo-acoustic soot spectrometer (PASS-3) and aerosol mass spectrometers (AMS) were used to explore the relationship between the measured AAE and the relative abundance of organic aerosol to BC. The regression and extrapolation analysis revealed that more realistic AAE values for "pure" BC aerosol (AAEBC) were 0.86, 0.82, and 1.02 between 405 and 781 nm, and 0.70, 0.71, and 0.86 between 532 and 781 nm, in the campaigns of urbanwinter, urbanfall, and ruralfall, respectively. Roadway tunnel experiments were conducted and the results further confirmed the representativeness of the obtained AAEBC values for the urban environment. Finally, the average light absorption contributions of BrC (± relative uncertainties) at 405 nm were quantified to be 11.7 % (±5 %), 6.3 % (±4 %), and 12.1 % (±7 %) in the campaigns of urbanwinter, urbanfall, and ruralfall, respectively, and those at 532 nm were 10.0 % (±2 %), 4.1 % (±3 %), and 5.5 % (±5 %), respectively. The relatively higher BrC absorption contribution at 405 nm in the ruralfall campaign could be reasonably attributed to the biomass burning events nearby, which was then directly supported by the biomass burning simulation experiments performed in this study. This paper indicates that the BrC contribution to total aerosol light absorption at shorter wavelengths is not negligible in the highly urbanized and industrialized PRD region.

Pesticides in the atmosphere: a comparison of gas-particle partitioning and particle size distribution of legacy and current-use pesticides

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Source: *Atmos. Chem. Phys., 16, 1531-1544, 2016*
http://www.atmos-chem-phys.net/16/1531/2016/doi:10.5194/acp-16-1531-2016

This study presents a comparison of seasonal variation, gas-particle partitioning, and particle-phase size distribution of organochlorine pesticides (OCPs) and current-use pesticides (CUPs) in air. Two years (2012/2013) of weekly air samples were collected at a background site in the Czech Republic using a high-volume air sampler. To study the particle-phase size distribution, air samples were also collected at an urban and rural site in the area of Brno, Czech Republic, using a cascade impactor separating atmospheric particulates according to six size fractions. Major differences were found in the atmospheric distribution of OCPs and CUPs. The atmospheric concentrations of CUPs were driven by agricultural activities while secondary sources such as volatilization from surfaces governed the atmospheric concentrations of OCPs. Moreover, clear differences were observed in gas-particle partitioning; CUP partitioning was influenced by adsorption onto mineral surfaces while OCPs were mainly partitioning to aerosols through absorption. A predictive method for estimating the gas-particle partitioning has been derived and is proposed for polar and non-polar pesticides. Finally, while OCPs and the majority of CUPs were largely found on fine particles, four CUPs (carbendazim, isoproturon, prochloraz, and terbuthylazine) had higher concentrations on coarse particles (> 3.0 μm), which may be related to the pesticide application technique. This finding is particularly important and should be further investigated given that large particles result in lower risks from inhalation (regardless the toxicity of the pesticide) and lower potential for long-range atmospheric transport.

Mid-21st century air quality at the urban scale under the influence of changed climate and emissions – case studies for Paris and Stockholm

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Source: *Atmos. Chem. Phys., 16, 1877-1894, 2016*
http://www.atmos-chem-phys.net/16/1877/2016/doi:10.5194/acp-16-1877-2016

Ozone, PM10 and PM2.5 concentrations over Paris, France and Stockholm, Sweden were modelled at 4 and 1 km horizontal resolutions respectively for the present and 2050 periods employing decade-long simulations. We account for large-scale global climate change (RCP-4.5) and fine-

resolution bottom-up emission projections developed by local experts and quantify their impact on future pollutant concentrations. Moreover, we identify biases related to the implementation of regional-scale emission projections by comparing modelled pollutant concentrations between the fine- and coarse-scale simulations over the study areas. We show that over urban areas with major regional contribution (e.g. the city of Stockholm) the bias related to coarse-scale projections may be significant and lead to policy misclassification. Our results stress the need to better understand the mechanism of bias propagation across the modelling scales in order to design more successful local-scale strategies. We find that the impact of climate change is spatially homogeneous in both regions, implying strong regional influence. The climate benefit for ozone (daily mean and maximum) is up to −5 % for Paris and −2 % for Stockholm city. The climate benefit on PM2.5 and PM10 in Paris is between −5 and −10 %, while for Stockholm we estimate mixed trends of up to 3 % depending on season and size class. In Stockholm, emission mitigation leads to concentration reductions up to 15 % for daily mean and maximum ozone and 20 % for PM. Through a sensitivity analysis we show that this response is entirely due to changes in emissions at the regional scale. On the contrary, over the city of Paris (VOC-limited photochemical regime), local mitigation of NOx emissions increases future ozone concentrations due to ozone titration inhibition. This competing trend between the respective roles of emission and climate change, results in an increase in 2050 daily mean ozone by 2.5 % in Paris. Climate and not emission change appears to be the most influential factor for maximum ozone concentration over the city of Paris, which may be particularly interesting from a health impact perspective.

Contribution of ship emissions to the concentration and deposition of air pollutants in Europe

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Source: *Atmos. Chem. Phys., 16, 1895-1906, 2016*
http://www.atmos-chem-phys.net/16/1895/2016/doi:10.5194/acp-16-1895-2016

Emissions from the marine transport sector are one of the least-regulated anthropogenic emission sources and contribute significantly to air pollution. Although strict limits were introduced recently for the maximum sulfur content in marine fuels in the SECAs (sulfur emission control areas) and in EU ports, sulfur emissions outside the SECAs and emissions of other components in all European maritime areas have continued to increase in the last two decades. We have used the air quality model CAMx (Comprehensive Air Quality Model with Extensions) with and without ship emissions for the year 2006 to determine the effects of international shipping on the annual as well as seasonal concentrations of ozone, primary and secondary components of PM2.5, and the dry and wet deposition of nitrogen and sulfur compounds in Europe. The largest changes in pollutant concentrations due to ship emissions were predicted for summer. Concentrations of particulate sulfate increased due to ship emissions in the Mediterranean (up to 60 %), the English Channel and the North Sea (30–35 %), while increases in particulate nitrate levels were found especially in the

north, around the Benelux area (20 %), where there were high NH3 land-based emissions. Our model results showed that not only are the atmospheric concentrations of pollutants affected by ship emissions, but also depositions of nitrogen and sulfur compounds increase significantly along the shipping routes. NOx emissions from the ships, especially in the English Channel and the North Sea, cause a decrease in the dry deposition of reduced nitrogen at source regions by moving it from the gas phase to the particle phase which then contributes to an increase in the wet deposition at coastal areas with higher precipitation. In the western Mediterranean region, on the other hand, model results show an increase in the deposition of oxidized nitrogen (mostly HNO3) due to the ship traffic. Dry deposition of SO2 seems to be significant along the shipping routes, whereas sulfate wet deposition occurs mainly along the Scandinavian and Adriatic coasts. The results presented in this paper suggest that evolution of NOx emissions from ships and land-based NH3 emissions will play a significant role in future European air quality.

Sensitivity of simulated CO2 concentration to sub-annual variations in fossil fuel CO2 emissions

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Recent advances in fossil fuel CO2 (FFCO2) emission inventories enable sensitivity tests of simulated atmospheric CO2 concentrations to sub-annual variations in FFCO2 emissions and what this implies for the interpretation of observed CO2. Six experiments are conducted to investigate the potential impact of three cycles of FFCO2 emission variability (diurnal, weekly and monthly) using a global tracer transport model. Results show an annual FFCO2 rectification varying from −1.35 to +0.13 ppm from the combination of all three cycles. This rectification is driven by a large negative diurnal FFCO2 rectification due to the covariation of diurnal FFCO2 emissions and diurnal vertical mixing, as well as a smaller positive seasonal FFCO2 rectification driven by the covariation of monthly FFCO2 emissions and monthly atmospheric transport. The diurnal FFCO2 emissions are responsible for a diurnal FFCO2 concentration amplitude of up to 9.12 ppm at the grid cell scale. Similarly, the monthly FFCO2 emissions are responsible for a simulated seasonal CO2 amplitude of up to 6.11 ppm at the grid cell scale. The impact of the diurnal FFCO2 emissions, when only sampled in the local afternoon, is also important, causing an increase of +1.13 ppmv at the grid cell scale. The simulated CO2 concentration impacts from the diurnally and seasonally varying FFCO2 emissions are centered over large source regions in the Northern Hemisphere, extending to downwind regions. This study demonstrates the influence of sub-annual variations in FFCO2 emissions on simulated CO2 concentration and suggests that inversion studies must take account of these variations in the affected regions.

Ammonia in the summertime Arctic marine boundary layer: sources, sinks, and implications

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Continuous hourly measurements of gas-phase ammonia (NH3(g)) were taken from 13 July to 7 August 2014 on a research cruise throughout Baffin Bay and the eastern Canadian Arctic Archipelago. Concentrations ranged from 30 to 650 ng m−3 (40–870 pptv) with the highest values recorded in Lancaster Sound (74°13′ N, 84°00′ W). Simultaneous measurements of total ammonium ([NHx]), pH and temperature in the ocean and in melt ponds were used to compute the compensation point (χ), which is the ambient NH3(g) concentration at which surface–air fluxes change direction. Ambient NH3(g) was usually several orders of magnitude larger than both χocean and χMP (< 0.4–10 ng m3) indicating these surface pools are net sinks of NH3. Flux calculations estimate average net downward fluxes of 1.4 and 1.1 ng m−2 s−1for the open ocean and melt ponds, respectively. Sufficient NH3(g) was present to neutralize non-sea-salt sulfate (nss-SO42−) in the boundary layer during most of the study. This finding was corroborated with a historical data set of PM2.5 composition from Alert, Nunavut (82°30′ N, 62°20′ W) wherein the median ratio of NH4+/nss-SO42− equivalents was greater than 0.75 in June, July and August. The GEOS-Chem chemical transport model was employed to examine the impact of NH3(g) emissions from seabird guano on boundary-layer composition and nss-SO42−neutralization. A GEOS-Chem simulation without seabird emissions underestimated boundary layer NH3(g) by several orders of magnitude and yielded highly acidic aerosol. A simulation that included seabird NH3 emissions was in better agreement with observations for both NH3(g)concentrations and nss-SO42− neutralization. This is strong evidence that seabird colonies are significant sources of NH3 in the summertime Arctic, and are ubiquitous enough to impact atmospheric composition across the entire Baffin Bay region. Large wildfires in the Northwest Territories were likely an important source of NH3, but their influence was probably limited to the Central Canadian Arctic. Implications of seabird-derived N-deposition to terrestrial and aquatic ecosystems are also discussed.

On the progress of the 2015–2016 El Niño event

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http://www.atmos-chem-phys.net/16/2007/2016/doi:10.5194/acp-16-2007-2016

It has been recently reported that the current 2015–2016 El Niño could become "one of the strongest on record". To further explore this claim, we performed the new analysis described in detail in Varotsos et al. (2015) that allows the detection of precursory signals of the strong El Niño events by using a recently developed non-linear dynamics tool. In this context, the analysis of the Southern Oscillation Index time series for the period 1876–2015 shows that the running 2015–2016 El Niño would be rather a "moderate to strong" or even a "strong" event and not “one of the strongest on record", as that of 1997–1998.

Volatility of organic aerosol and its components in the megacity of Paris

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http://www.atmos-chem-phys.net/16/2013/2016/doi:10.5194/acp-16-2013-2016

Using a mass transfer model and the volatility basis set, we estimate the volatility distribution for the organic aerosol (OA) components during summer and winter in Paris, France as part of the collaborative project MEGAPOLI. The concentrations of the OA components as a function of temperature were measured combining data from a thermodenuder and an aerosol mass spectrometer (AMS) with Positive Matrix Factorization (PMF) analysis. The hydrocarbon-like organic aerosol (HOA) had similar volatility distributions for the summer and winter campaigns with half of the material in the saturation concentration bin of 10 μg m−3 and another 35–40 % consisting of low and extremely low volatility organic compounds (LVOCs with effective saturation concentrations C* of 10−3–0.1 μg m−3 and ELVOCs C* less or equal than 10−4 μg m−3, respectively). The winter cooking OA (COA) was more than an order of magnitude less volatile than the summer COA. The low-volatility oxygenated OA (LV-OOA) factor detected in the summer had the lowest volatility of all the derived factors and consisted almost exclusively of ELVOCs. The volatility for the semi-volatile oxygenated OA (SV-OOA) was significantly higher than that of the LV-OOA, containing both semi-volatile organic components (SVOCs with C* in the 1–100 μg m−3 range) and LVOCs. The oxygenated OA (OOA) factor in winter consisted of SVOCs (45 %), LVOCs (25 %) and ELVOCs (30 %). The volatility of marine OA (MOA) was higher than that of the other factors

containing around 60 % SVOCs. The biomass burning OA (BBOA) factor contained components with a wide range of volatilities with significant contributions from both SVOCs (50 %) and LVOCs (30 %). Finally, combining the bulk average O : C ratios and volatility distributions of the various factors, our results are placed into the two-dimensional volatility basis set (2D-VBS) framework. The OA factors cover a broad spectrum of volatilities with no direct link between the average volatility and average O : C of the OA components.

High-resolution ammonia emissions inventories in China from 1980 to 2012

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***Source**: Atmos. Chem. Phys., 16, 2043-2058, 2016*
http://www.atmos-chem-phys.net/16/2043/2016/doi:10.5194/acp-16-2043-2016

Ammonia (NH3) can interact in the atmosphere with other trace chemical species, which can lead to detrimental environmental consequences, such as the formation of fine particulates and ultimately global climate change. China is a major agricultural country, and livestock numbers and nitrogen fertilizer use have increased drastically since 1978, following the rapid economic and industrial development experienced by the country. In this study, comprehensive NH3 emissions inventories were compiled for China for 1980–2012. In a previous study, we parameterized emissions factors (EFs) considering ambient temperature, soil acidity, and the method and rate of fertilizer application. In this study, we refined these EFs by adding the effects of wind speed and new data from field experiments of NH3 flux in cropland in northern China. We found that total NH3 emissions in China increased from 5.9 to 11.1 Tg from 1980 to 1996, and then decreased to 9.7 Tg in 2012. The two major contributors were livestock manure and synthetic fertilizer application, which contributed 80–90 % of the total emissions. Emissions from livestock manure rose from 2.86 Tg (1980) to 6.16 Tg (2005), and then decreased to 5.0 Tg (2012); beef cattle were the largest source followed by laying hens and pigs. The remarkable downward trend in livestock emissions that occurred in 2007 was attributed to a decrease in the numbers of various livestock animals, including beef cattle, goats, and sheep. Meanwhile, emissions from synthetic fertilizer ranged from 2.1 Tg (1980) to 4.7 Tg (1996), and then declined to 2.8 Tg (2012). Urea and ammonium bicarbonate (ABC) dominated this category of emissions, and a decline in ABC application led to the decrease in emissions that took place from the mid-1990s onwards. High emissions were concentrated in eastern and southwestern China. Seasonally, peak NH3 emissions occurred in spring and summer. The inventories had a monthly temporal resolution and a spatial resolution of 1000 m, and thus are suitable for global and regional air-quality modeling.

Atmospheric OH reactivity in central London: observations, model predictions and estimates of in situ ozone production

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Source: *Atmos. Chem. Phys., 16, 2109-2122, 2016*
http://www.atmos-chem-phys.net/16/2109/2016/doi:10.5194/acp-16-2109-2016

Near-continuous measurements of hydroxyl radical (OH) reactivity in the urban background atmosphere of central London during the summer of 2012 are presented. OH reactivity behaviour is seen to be broadly dependent on air mass origin, with the highest reactivity and the most pronounced diurnal profile observed when air had passed over central London to the east, prior to measurement. Averaged over the entire observation period of 26 days, OH reactivity peaked at $\sim$ 27 s−1 in the morning, with a minimum of $\sim$ 15 s−1 during the afternoon. A maximum OH reactivity of 116 s−1 was recorded on one day during morning rush hour. A detailed box model using the Master Chemical Mechanism was used to calculate OH reactivity, and was constrained with an extended measurement data set of volatile organic compounds (VOCs) derived from a gas chromatography flame ionisation detector (GC-FID) and a two-dimensional GC instrument which included heavier molecular weight (up to C12) aliphatic VOCs, oxygenated VOCs and the biogenic VOCs α-pinene and limonene. Comparison was made between observed OH reactivity and modelled OH reactivity using (i) a standard suite of VOC measurements (C2–C8 hydrocarbons and a small selection of oxygenated VOCs) and (ii) a more comprehensive inventory including species up to C12. Modelled reactivities were lower than those measured (by 33 %) when only the reactivity of the standard VOC suite was considered. The difference between measured and modelled reactivity was improved, to within 15 %, if the reactivity of the higher VOCs ($\geqslant$ C9) was also considered, with the reactivity of the biogenic compounds of α-pinene and limonene and their oxidation products almost entirely responsible for this improvement. Further improvements in the model's ability to reproduce OH reactivity (to within 6 %) could be achieved if the reactivity and degradation mechanism of unassigned two-dimensional GC peaks were estimated. Neglecting the contribution of the higher VOCs ($\geqslant$ C9) (particularly α-pinene and limonene) and model-generated intermediates increases the modelled OH concentrations by 41 %, and the magnitude of in situ ozone production calculated from the production of RO2 was significantly lower (60 %). This work highlights that any future ozone abatement strategies should consider the role that biogenic emissions play alongside anthropogenic emissions in influencing London's air quality.

Air quality and radiative impacts of Arctic shipping emissions in the summertime in northern Norway: from the local to the regional scale

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***Source**: Atmos. Chem. Phys., 16, 2359-2379, 2016*
http://www.atmos-chem-phys.net/16/2359/2016/doi:10.5194/acp-16-2359-2016

In this study, we quantify the impacts of shipping pollution on air quality and shortwave radiative effect in northern Norway, using WRF-Chem (Weather Research and Forecasting with chemistry) simulations combined with high-resolution, real-time STEAM2 (Ship Traffic Emissions Assessment Model version 2) shipping emissions. STEAM2 emissions are evaluated using airborne measurements from the ACCESS (Arctic Climate Change, Economy and Society) aircraft campaign, which was conducted in the summer 2012, in two ways. First, emissions of nitrogen oxides (NOx) and sulfur dioxide (SO2) are derived for specific ships by combining in situ measurements in ship plumes and FLEXPART-WRF plume dispersion modeling, and these values are compared to STEAM2 emissions for the same ships. Second, regional WRF-Chem runs with and without STEAM2 ship emissions are performed at two different resolutions, 3 km × 3 km and 15 km × 15 km, and evaluated against measurements along flight tracks and average campaign profiles in the marine boundary layer and lower troposphere. These comparisons show that differences between STEAM2 emissions and calculated emissions can be quite large (−57 to +148 %) for individual ships, but that WRF-Chem simulations using STEAM2 emissions reproduce well the average NOx, SO2 and O3 measured during ACCESS flights. The same WRF-Chem simulations show that the magnitude of NOx and ozone (O3) production from ship emissions at the surface is not very sensitive (< 5 %) to the horizontal grid resolution (15 or 3 km), while surface PM10 particulate matter enhancements due to ships are moderately sensitive (15 %) to resolution. The 15 km resolution WRF-Chem simulations are used to estimate the regional impacts of shipping pollution in northern Norway. Our results indicate that ship emissions are an important source of pollution along the Norwegian coast, enhancing 15-day-averaged surface concentrations of NOx (∼ +80 %), SO2 (∼ +80 %), O3 (∼ +5 %), black carbon (∼ +40 %), and PM2.5 (∼ +10 %). The residence time of black carbon originating from shipping emissions is 1.4 days. Over the same 15-day period, ship emissions in northern Norway have a global shortwave (direct + semi-direct + indirect) radiative effect of −9.3 m Wm−2.

Mixing layer height and its implications for air pollution over Beijing, China

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Source: *Atmos. Chem. Phys., 16, 2459-2475, 2016*
http://www.atmos-chem-phys.net/16/2459/2016/doi:10.5194/acp-16-2459-2016

The mixing layer is an important meteorological factor that affects air pollution. In this study, the atmospheric mixing layer height (MLH) was observed in Beijing from July 2009 to December 2012 using a ceilometer. By comparison with radiosonde data, we found that the ceilometer underestimates the MLH under conditions of neutral stratification caused by strong winds, whereas it overestimates the MLH when sand-dust is crossing. Using meteorological, PM2.5, and PM10 observational data, we screened the observed MLH automatically; the ceilometer observations were fairly consistent with the radiosondes, with a correlation coefficient greater than 0.9. Further analysis indicated that the MLH is low in autumn and winter and high in spring and summer in Beijing. There is a significant correlation between the sensible heat flux and MLH, and the diurnal cycle of the MLH in summer is also affected by the circulation of mountainous plain winds. Using visibility as an index to classify the degree of air pollution, we found that the variation in the sensible heat and buoyancy term in turbulent kinetic energy (TKE) is insignificant when visibility decreases from 10 to 5 km, but the reduction of shear term in TKE is near 70 %. When visibility decreases from 5 to 1 km, the variation of the shear term in TKE is insignificant, but the decrease in the sensible heat and buoyancy term in TKE is approximately 60 %. Although the correlation between the daily variation of the MLH and visibility is very poor, the correlation between them is significantly enhanced when the relative humidity increases beyond 80 %. This indicates that humidity-related physicochemical processes is the primary source of atmospheric particles under heavy pollution and that the dissipation of atmospheric particles mainly depends on the MLH. The presented results of the atmospheric mixing layer provide useful empirical information for improving meteorological and atmospheric chemistry models and the forecasting and warning of air pollution.

Transport pathways from the Asian monsoon anticyclone to the stratosphere

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http://www.atmos-chem-phys.net/16/2703/2016/doi:10.5194/acp-16-2703-2016

Transport pathways of air originating in the upper-tropospheric Asian monsoon anticyclone are investigated based on three-dimensional trajectories. The Asian monsoon anticyclone emerges in response to persistent deep convection over India and southeast Asia in northern summer, and this convection is associated with rapid transport from the surface to the upper troposphere and possibly into the stratosphere. Here, we investigate the fate of air that originates within the upper-tropospheric anticyclone from the outflow of deep convection, using trajectories driven by ERA-interim reanalysis data. Calculations include isentropic estimates, plus fully three-dimensional results based on kinematic and diabatic transport calculations. Isentropic calculations show that air parcels are typically confined within the anticyclone for 10–20 days and spread over the tropical belt within a month of their initialization. However, only few parcels (3 % at 360 K, 8 % at 380 K) reach the extratropical stratosphere by isentropic transport. When considering vertical transport we find that 31 % or 48 % of the trajectories reach the stratosphere within 60 days when using vertical velocities or diabatic heating rates to calculate vertical transport, respectively. In both cases, most parcels that reach the stratosphere are transported upward within the anticyclone and enter the stratosphere in the tropics, typically 10–20 days after their initialization at 360 K. This suggests that trace gases, including pollutants, that are transported into the stratosphere via the Asian monsoon system are in a position to enter the tropical pipe and thus be transported into the deep stratosphere. Sensitivity calculations with respect to the initial altitude of the trajectories showed that air needs to be transported to levels of 360 K or above by deep convection to likely (≧ 50 %) reach the stratosphere through transport by the large-scale circulation.

Drivers of changes in stratospheric and tropospheric ozone between year 2000 and 2100

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A stratosphere-resolving configuration of the Met Office's Unified Model (UM) with the United Kingdom Chemistry and Aerosols (UKCA) scheme is used to investigate the atmospheric response to changes in (a) greenhouse gases and climate, (b) ozone-depleting substances (ODSs) and (c) non-methane ozone precursor emissions. A suite of time-slice experiments show the separate, as well as pairwise, impacts of these perturbations between the years 2000 and 2100. Sensitivity to uncertainties in future greenhouse gases and aerosols is explored through the use of the Representative Concentration Pathway (RCP) 4.5 and 8.5 scenarios.

The results highlight an important role for the stratosphere in determining the annual mean tropospheric ozone response, primarily through stratosphere–troposphere exchange (STE) of

ozone. Under both climate change and reductions in ODSs, increases in STE offset decreases in net chemical production and act to increase the tropospheric ozone burden. This opposes the effects of projected decreases in ozone precursors through measures to improve air quality, which act to reduce the ozone burden.

The global tropospheric lifetime of ozone (τO3) does not change significantly under climate change at RCP4.5, but it decreases at RCP8.5. This opposes the increases in τO3 simulated under reductions in ODSs and ozone precursor emissions.

The additivity of the changes in ozone is examined by comparing the sum of the responses in the single-forcing experiments to those from equivalent combined-forcing experiments. Whilst the ozone responses to most forcing combinations are found to be approximately additive, non-additive changes are found in both the stratosphere and troposphere when a large climate forcing (RCP8.5) is combined with the effects of ODSs.

Urban stress-induced biogenic VOC emissions and SOA-forming potentials in Beijing

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Source: *Atmos. Chem. Phys., 16, 2901-2920, 2016*
http://www.atmos-chem-phys.net/16/2901/2016/doi:10.5194/acp-16-2901-2016

Trees can significantly impact the urban air chemistry by the uptake and emission of reactive biogenic volatile organic compounds (BVOCs), which are involved in ozone and particle formation. Here we present the emission potentials of "constitutive" (cBVOCs) and "stress-induced" BVOCs (sBVOCs) from the dominant broadleaf woody plant species in the megacity of Beijing. Based on the municipal tree census and cuvette BVOC measurements on leaf level, we built an inventory of BVOC emissions, and assessed the potential impact of BVOCs on secondary organic aerosol (SOA) formation in 2005 and 2010, i.e., before and after realizing the large tree-planting program for the 2008 Olympic Games. We found that sBVOCs, such as fatty acid derivatives, benzenoids, and sesquiterpenes, constituted a significant fraction (∼ 40 %) of the total annual BVOC emissions, and we estimated that the overall annual BVOC budget may have doubled from ∼ 4.8 × 109 g C year−1 in 2005 to ∼ 10.3 × 109 g C year−1 in 2010 due to the increase in urban greening, while at the same time the emission of anthropogenic VOCs (AVOCs) decreased by 24 %. Based on the BVOC emission assessment, we estimated the biological impact on SOA mass formation potential in Beijing. Constitutive and stress-induced BVOCs might produce similar amounts of secondary aerosol in Beijing. However, the main contributors of SOA-mass formations

originated from anthropogenic sources (> 90 %). This study demonstrates the general importance to include sBVOCs when studying BVOC emissions. Although the main problems regarding air quality in Beijing still originate from anthropogenic activities, the present survey suggests that in urban plantation programs, the selection of low-emitting plant species has some potential beneficial effects on urban air quality.

Ozone and carbon monoxide over India during the summer monsoon: regional emissions and transport

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Source: *Atmos. Chem. Phys., 16, 3013-3032, 2016*
http://www.atmos-chem-phys.net/16/3013/2016/doi:10.5194/acp-16-3013-2016

We compare in situ measurements of ozone (O3) and carbon monoxide (CO) profiles from the CARIBIC program with the results from the regional chemistry transport model (WRF-Chem) to investigate the role of local and regional emissions and long-range transport over southern India during the summer monsoon of 2008. WRF-Chem successfully reproduces the general features of O3 and CO distributions over the South Asian region. However, absolute CO concentrations in the lower troposphere are typically underestimated. Here we investigate the influence of local relative to remote emissions through sensitivity simulations.

The influence of 50 % increased CO emissions over South Asia leads to a significant enhancement (upto 20 % in July) in upper tropospheric CO in the northern and central Indian regions. Over Chennai in southern India, this causes a 33 % increase in surface CO during June. However, the influence of enhanced local and regional emissions is found to be smaller (5 %) in the free troposphere over Chennai, except during September. Local to regional emissions are therefore suggested to play a minor role in the underestimation of CO by WRF-Chem during June–August. In the lower troposphere, a high pollution (O3: 146.4 ± 12.8, CO: 136.4 ± 12.2 nmol mol−1) event (15 July 2008), not reproduced by the model, is shown to be due to transport of photochemically processed air masses from the boundary layer in southern India. A sensitivity simulation combined with backward trajectories indicates that long-range transport of CO to southern India is significantly underestimated, particularly in air masses from the west, i.e., from Central Africa. This study highlights the need for more aircraft-based measurements over India and adjacent regions and the improvement of global emission inventories.

Brown carbon aerosols from burning of boreal peatlands: microphysical properties, emission factors, and implications for direct radiative forcing

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Source: *Atmos. Chem. Phys., 16, 3033-3040, 2016*
http://www.atmos-chem-phys.net/16/3033/2016/doi:10.5194/acp-16-3033-2016

The surface air warming over the Arctic has been almost twice as much as the global average in recent decades. In this region, unprecedented amounts of smoldering peat fires have been identified as a major emission source of climate-warming agents. While much is known about greenhouse gas emissions from these fires, there is a knowledge gap on the nature of particulate emissions and their potential role in atmospheric warming. Here, we show that aerosols emitted from burning of Alaskan and Siberian peatlands are predominantly brown carbon (BrC) – a class of visible light-absorbing organic carbon (OC) – with a negligible amount of black carbon content. The mean fuel-based emission factors for OC aerosols ranged from 3.8 to 16.6 g kg−1. Their mass absorption efficiencies were in the range of 0.2–0.8 m2 g−1 at 405 nm (violet) and dropped sharply to 0.03–0.07 m2 g−1 at 532 nm (green), characterized by a mean Ångström exponent of ≈ 9. Electron microscopy images of the particles revealed their morphologies to be either single sphere or agglomerated "tar balls". The shortwave top-of-atmosphere aerosol radiative forcing per unit optical depth under clear-sky conditions was estimated as a function of surface albedo. Only over bright surfaces with albedo greater than 0.6, such as snow cover and low-level clouds, the emitted aerosols could result in a net warming (positive forcing) of the atmosphere.

Development of a vehicle emission inventory with high temporal–spatial resolution based on NRT traffic data and its impact on air pollution in Beijing – Part 1: Development and evaluation of vehicle emission inventory

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Source: *Atmos. Chem. Phys., 16, 3161-3170, 2016*
http://www.atmos-chem-phys.net/16/3161/2016/doi:10.5194/acp-16-3161-2016

This paper presents a bottom-up methodology based on the local emission factors, complemented with the widely used emission factors of Computer Programme to Calculate Emissions from Road

Transport (COPERT) model and near-real-time traffic data on road segments to develop a vehicle emission inventory with high temporal–spatial resolution (HTSVE) for the Beijing urban area. To simulate real-world vehicle emissions accurately, the road has been divided into segments according to the driving cycle (traffic speed) on this road segment. The results show that the vehicle emissions of NOx, CO, HC and PM were 10.54 × 104, 42.51 × 104 and 2.13 × 104 and 0.41 × 104 Mg respectively. The vehicle emissions and fuel consumption estimated by the model were compared with the China Vehicle Emission Control Annual Report and fuel sales thereafter. The grid-based emissions were also compared with the vehicular emission inventory developed by the macro-scale approach. This method indicates that the bottom-up approach better estimates the levels and spatial distribution of vehicle emissions than the macro-scale method, which relies on more information. Based on the results of this study, improved air quality simulation and the contribution of vehicle emissions to ambient pollutant concentration in Beijing have been investigated in a companion paper (He et al., 2016).

Development of a vehicle emission inventory with high temporal–spatial resolution based on NRT traffic data and its impact on air pollution in Beijing – Part 2: Impact of vehicle emission on urban air quality

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Source: *Atmos. Chem. Phys., 16, 3171-3184, 2016*
http://www.atmos-chem-phys.net/16/3171/2016/doi:10.5194/acp-16-3171-2016

A companion paper developed a vehicle emission inventory with high temporal–spatial resolution (HTSVE) with a bottom-up methodology based on local emission factors, complemented with the widely used emission factors of COPERT model and near-real-time (NRT) traffic data on a specific road segment for 2013 in urban Beijing (Jing et al., 2016), which is used to investigate the impact of vehicle pollution on air pollution in this study. Based on the sensitivity analysis method of switching on/off pollutant emissions in the Chinese air quality forecasting model CUACE, a modelling study was carried out to evaluate the contributions of vehicle emission to the air pollution in Beijing's main urban areas in the periods of summer (July) and winter (December) 2013. Generally, the CUACE model had good performance of the concentration simulation of pollutants. The model simulation has been improved by using HTSVE. The vehicle emission contribution (VEC) to ambient pollutant concentrations not only changes with seasons but also changes with time. The mean VEC, affected by regional pollutant transports significantly, is 55.4 and 48.5 % for NO2 and 5.4 and 10.5 % for PM2.5 in July and December 2013 respectively. Regardless of regional transports, relative vehicle emission contribution (RVEC) to NO2 is 59.2 and 57.8 % in July and December 2013, while it is 8.7 and 13.9 % for PM2.5. The RVEC to PM2.5 is lower than the PM2.5 contribution rate for vehicle emission in total emission, which may be due to dry deposition

of PM2.5 from vehicle emission in the near-surface layer occuring more easily than from elevated source emission.

New insights into PM2.5 chemical composition and sources in two major cities in China during extreme haze events using aerosol mass spectrometry

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Source: *Atmos. Chem. Phys., 16, 3207-3225, 2016*
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During winter 2013–2014 aerosol mass spectrometer (AMS) measurements were conducted for the first time with a novel PM2.5(particulate matter with aerodynamic diameter ≤ 2.5 μm) lens in two major cities of China: Xi'an and Beijing. We denote the periods with visibility below 2 km as extreme haze and refer to the rest as reference periods. During the measurements in Xi'an an extreme haze covered the city for about a week and the total non-refractory (NR)-PM2.5 mass fraction reached peak concentrations of over 1000 μg m−3. During the measurements in Beijing two extreme haze events occurred, but the temporal extent and the total concentrations reached during these events were lower than in Xi'an. Average PM2.5 concentrations of 537 ± 146 and 243 ± 47 μg m−3 (including NR species and equivalent black carbon, eBC) were recorded during the extreme haze events in Xi'an and Beijing, respectively. During the reference periods the measured average concentrations were 140 ± 99 μg m−3 in Xi'an and 75 ± 61 μg m−3 in Beijing. The relative composition of the NR-PM2.5 evolved substantially during the extreme haze periods, with increased contributions of the inorganic components (mostly sulfate and nitrate). Our results suggest that the high relative humidity present during the extreme haze events had a strong effect on the increase of sulfate mass (via aqueous phase oxidation of sulfur dioxide). Another relevant characteristic of the extreme haze is the size of the measured particles. During the extreme haze events, the AMS showed much larger particles, with a volume weighted mode at about 800 to 1000 nm, in contrast to about 400 nm during reference periods. These large particle sizes made the use of the PM2.5 inlet crucial, especially during the severe haze events, where 39 ± 5 % of the mass would have been lost in the conventional PM1 (particulate matter with aerodynamic diameter ≤ 1 μm) inlet. A novel positive matrix factorization procedure was developed to apportion the sources of organic aerosols (OA) based on their mass spectra using the multilinear engine (ME-2) controlled via the source finder (SoFi). The procedure allows for an effective exploration of the solution space, a more objective selection of the best solution and an estimation of the rotational uncertainties. Our results clearly show an increase of the oxygenated organic aerosol (OOA) mass during extreme haze

events. The contribution of OOA to the total OA increased from the reference to the extreme haze periods from 16.2 ± 1.1 to 31.3 ± 1.5 % in Xi'an and from 15.7 ± 0.7 to 25.0 ± 1.2 % in Beijing. By contrast, during the reference periods the total OA mass was dominated by domestic emissions of primary aerosols from biomass burning in Xi'an (42.2 ± 1.5 % of OA) and coal combustion in Beijing (55.2 ± 1.6 % of OA). These two sources are also mostly responsible for extremely high polycyclic aromatic hydrocarbon (PAH) concentrations measured with the AMS (campaign average of 2.1 ± 2.0 µg m−3 and frequent peak concentrations above 10 µg m−3). To the best of our knowledge, this is the first data set where the simultaneous extraction of these two primary sources could be achieved in China by conducting on-line AMS measurements at two areas with contrasted emission patterns.

Contribution of oil and natural gas production to renewed increase in atmospheric methane (2007–2014): top–down estimate from ethane and methane column observations

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Harmonized time series of column-averaged mole fractions of atmospheric methane and ethane over the period 1999–2014 are derived from solar Fourier transform infrared (FTIR) measurements at the Zugspitze summit (47° N, 11° E; 2964 m a.s.l.) and at Lauder (45° S, 170° E; 370 m a.s.l.). Long-term trend analysis reveals a consistent renewed methane increase since 2007 of 6.2 [5.6, 6.9] ppb yr−1 (parts-per-billion per year) at the Zugspitze and 6.0 [5.3, 6.7] ppb yr−1 at Lauder (95 % confidence intervals). Several recent studies provide pieces of evidence that the renewed methane increase is most likely driven by two main factors: (i) increased methane emissions from tropical wetlands, followed by (ii) increased thermogenic methane emissions due to growing oil and natural gas production. Here, we quantify the magnitude of the second class of sources, using long-term measurements of atmospheric ethane as a tracer for thermogenic methane emissions. In 2007, after years of weak decline, the Zugspitze ethane time series shows the sudden onset of a significant positive trend (2.3 [1.8, 2.8] × 10−2 ppb yr−1for 2007–2014), while a negative trend persists at Lauder after 2007 (−0.4 [−0.6, −0.1] × 10−2 ppb yr−1). Zugspitze methane and ethane time series are significantly correlated for the period 2007–2014 and can be assigned to thermogenic methane emissions with an ethane-to-methane ratio (EMR) of 12–19 %. We present optimized emission scenarios for 2007–2014 derived from an atmospheric two-box model. From our trend observations we infer a total ethane emission increase over the period 2007–2014 from oil and natural gas sources of 1–11 Tg yr−1along with an overall methane emission increase of 24–45 Tg yr−1. Based on these results, the oil and natural gas emission contribution (C) to the renewed methane increase is deduced using three different emission

scenarios with dedicated EMR ranges. Reference scenario 1 assumes an oil and gas emission combination with EMR = 7.0–16.2 %, which results in a minimum contribution C > 39 % (given as lower bound of 95 % confidence interval). Beside this most plausible scenario 1, we consider two less realistic limiting cases of pure oil-related emissions (scenario 2 with EMR = 16.2–31.4 %) and pure natural gas sources (scenario 3 with EMR = 4.4–7.0 %), which result in C > 18 % and C > 73 %, respectively. Our results suggest that long-term observations of column-averaged ethane provide a valuable constraint on the source attribution of methane emission changes and provide basic knowledge for developing effective climate change mitigation strategies.

Top-down estimates of benzene and toluene emissions in the Pearl River Delta and Hong Kong, China

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Source: *Atmos. Chem. Phys., 16, 3369-3382, 2016*
http://www.atmos-chem-phys.net/16/3369/2016/doi:10.5194/acp-16-3369-2016

Benzene (C6H6) and toluene (C7H8) are toxic to humans and the environment. They are also important precursors of ground-level ozone and secondary organic aerosols and contribute substantially to severe air pollution in urban areas in China. Discrepancies exist between different bottom-up inventories for benzene and toluene emissions in the Pearl River Delta (PRD) and Hong Kong (HK), which are emission hot spots in China. This study provides top-down estimates of benzene and toluene emissions in the PRD and HK using atmospheric measurement data from a rural site in the area, Heshan, an atmospheric transport model, and an inverse modeling method. The model simulations captured the measured mixing ratios during most pollution episodes. For the PRD and HK, the benzene emissions estimated in this study for 2010 were 44 (12–75) and 5 (2–7) Gg yr−1 for the PRD and HK, respectively, and the toluene emissions were 131 (44–218) and 6 (2–9) Gg yr−1, respectively. Temporal and spatial differences between the inversion estimate and four different bottom-up emission estimates are discussed, and it is proposed that more observations at different sites are urgently needed to better constrain benzene and toluene (and other air pollutant) emissions in the PRD and HK in the future.

A global simulation of brown carbon: implications for photochemistry and direct radiative effect

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http://www.atmos-chem-phys.net/16/3413/2016/doi:10.5194/acp-16-3413-2016

Recent observations suggest that a certain fraction of organic carbon (OC) aerosol effectively absorbs solar radiation, which is also known as brown carbon (BrC) aerosol. Despite much observational evidence of its presence, very few global modelling studies have been conducted because of poor understanding of global BrC emissions. Here we present an explicit global simulation of BrC in a global 3-D chemical transport model (GEOS-Chem), including global BrC emission estimates from primary (3.9 ± 1.7 and 3.0 ± 1.3 TgC yr−1 from biomass burning and biofuel) and secondary (5.7 TgC yr−1 from aromatic oxidation) sources. We evaluate the model by comparing the results with observed absorption by water-soluble OC in surface air in the United States, and with single scattering albedo observations at Aerosol Robotic Network (AERONET) sites all over the globe. The model successfully reproduces the seasonal variations of observed light absorption by water-soluble OC, but underestimates the magnitudes, especially in regions with high secondary source contributions. Our global simulations show that BrC accounts for 21 % of the global mean surface OC concentration, which is typically assumed to be scattering. We find that the global direct radiative effect of BrC is nearly zero at the top of the atmosphere, and consequently decreases the direct radiative cooling effect of OC by 16 %. In addition, the BrC absorption leads to a general reduction of NO2 photolysis rates, whose maximum decreases occur in Asia up to −8 % (−17 %) on an annual (spring) mean basis. The resulting decreases of annual (spring) mean surface ozone concentrations are up to −6 % (−13 %) in Asia, indicating a non-negligible effect of BrC on photochemistry in this region.

Production of peroxy nitrates in boreal biomass burning plumes over Canada during the BORTAS campaign

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Source: *tmos. Chem. Phys., 16, 3485-3497, 2016*
http://www.atmos-chem-phys.net/16/3485/2016/doi:10.5194/acp-16-3485-2016

The observations collected during the BOReal forest fires on Tropospheric oxidants over the Atlantic using Aircraft and Satellites (BORTAS) campaign in summer 2011 over Canada are analysed to study the impact of forest fire emissions on the formation of ozone (O3) and total

peroxy nitrates ∑PNs, ∑ROONO2). The suite of measurements on board the BAe-146 aircraft, deployed in this campaign, allows us to calculate the production of O3 and of ∑PNs, a long-lived NOx reservoir whose concentration is supposed to be impacted by biomass burning emissions. In fire plumes, profiles of carbon monoxide (CO), which is a well-established tracer of pyrogenic emission, show concentration enhancements that are in strong correspondence with a significant increase of concentrations of ∑PNs, whereas minimal increase of the concentrations of O3 and NO2 is observed. The ∑PN and O3 productions have been calculated using the rate constants of the first- and second-order reactions of volatile organic compound (VOC) oxidation. The ∑PN and O3 productions have also been quantified by 0-D model simulation based on the Master Chemical Mechanism. Both methods show that in fire plumes the average production of ∑PNs and O3 are greater than in the background plumes, but the increase of ∑PN production is more pronounced than the O3 production. The average ∑PN production in fire plumes is from 7 to 12 times greater than in the background, whereas the average O3 production in fire plumes is from 2 to 5 times greater than in the background. These results suggest that, at least for boreal forest fires and for the measurements recorded during the BORTAS campaign, fire emissions impact both the oxidized NOy and O3, but (1 ∑PN production is amplified significantly more than O3 production and (2) in the forest fire plumes the ratio between the O3 production and the ∑PN production is lower than the ratio evaluated in the background air masses, thus confirming that the role played by the ∑PNs produced during biomass burning is significant in the O3 budget. The implication of these observations is that fire emissions in some cases, for example boreal forest fires and in the conditions reported here, may influence more long-lived precursors of O3 than short-lived pollutants, which in turn can be transported and eventually diluted in a wide area.

Exploring the uncertainty associated with satellite-based estimates of premature mortality due to exposure to fine particulate matter

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The negative impacts of fine particulate matter (PM2.5) exposure on human health are a primary motivator for air quality research. However, estimates of the air pollution health burden vary considerably and strongly depend on the data sets and methodology. Satellite observations of aerosol optical depth (AOD) have been widely used to overcome limited coverage from surface monitoring and to assess the global population exposure to PM2.5 and the associated premature mortality. Here we quantify the uncertainty in determining the burden of disease using this approach, discuss different methods and data sets, and explain sources of discrepancies among values in the literature. For this purpose we primarily use the MODIS satellite observations in concert with the GEOS-Chem chemical transport model. We contrast results in the United States

and China for the years 2004–2011. Using the Burnett et al. (2014) integrated exposure response function, we estimate that in the United States, exposure to PM2.5 accounts for approximately 2 % of total deaths compared to 14 % in China (using satellite-based exposure), which falls within the range of previous estimates. The difference in estimated mortality burden based solely on a global model vs. that derived from satellite is approximately 14 % for the US and 2 % for China on a nationwide basis, although regionally the differences can be much greater. This difference is overshadowed by the uncertainty in the methodology for deriving PM2.5 burden from satellite observations, which we quantify to be on the order of 20 % due to uncertainties in the AOD-to-surface-PM2.5 relationship, 10 % due to the satellite observational uncertainty, and 30 % or greater uncertainty associated with the application of concentration response functions to estimated exposure.

The contrasting roles of water and dust in controlling daily variations in radiative heating of the summertime Saharan heat low

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The summertime Sahara heat low (SHL) is a key component of the West African monsoon (WAM) system. Considerable uncertainty remains over the relative roles of water vapour and dust aerosols in controlling the radiation budget over the Sahara and therefore our ability to explain variability and trends in the SHL, and in turn, the WAM. Here, new observations from Fennec supersite-1 in the central Sahara during June 2011 and June 2012, together with satellite retrievals from GERB, are used to quantify how total column water vapour (TCWV) and dust aerosols (from aerosol optical depth, AOD) control day-to-day variations in energy balance in both observations and ECWMF reanalyses (ERA-I). The data show that the earth-atmosphere system is radiatively heated in June 2011 and 2012. Although the empirical analysis of observational data cannot completely disentangle the roles of water vapour, clouds and dust, the analysis demonstrates that TCWV provides a far stronger control on TOA net radiation, and so the net heating of the earth-atmosphere system, than AOD does. In contrast, variations in dust provide a much stronger control on surface heating, but the decreased surface heating associated with dust is largely compensated by increased atmospheric heating, and so dust control on net TOA radiation is weak. Dust and TCWV are both important for direct atmospheric heating. ERA-I, which assimilated radiosondes from the Fennec campaign, captures the control of TOA net flux by TCWV, with a positive correlation ($r = 0.6$) between observed and modelled TOA net radiation, despite the use of a monthly dust climatology in ERA-I that cannot capture the daily variations in dustiness. Variations in surface net radiation, and so the vertical profile of radiative heating, are not captured in ERA-I,

since it does not capture variations in dust. Results show that ventilation of the SHL by cool moist air leads to a radiative warming, stabilising the SHL with respect to such perturbations. It is known that models struggle to capture the advective moistening of the SHL, especially that associated with mesoscale convective systems. Our results show that the typical model errors in Saharan water vapour will lead to substantial errors in the modelled TOA energy balance (tens of W m−2), which will lead to errors in both the SHL and the WAM.

The importance of vehicle emissions as a source of atmospheric ammonia in the megacity of Shanghai

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Agricultural activities are a major source contributing to NH3 emissions in Shanghai and most other regions of China; however, there is a long-standing and ongoing controversy regarding the contributions of vehicle-emitted NH3 to the urban atmosphere. From April 2014 to April 2015, we conducted measurements of a wide range of gases (including NH3) and the chemical properties of PM2.5 at hourly resolution at a Shanghai urban supersite. This large data set shows NH3 pollution events, lasting several hours with concentrations 4 times the annual average of 5.3 μg m−3, caused by the burning of crop residues in spring. There are also generally higher NH3 concentrations (mean ± 1 σ) in summer (7.3 ± 4.9 μg m−3; n = 2181) because of intensive emissions from temperature-dependent agricultural sources. However, the NH3 concentration in summer was only an average of 2.4 μg m−3 or 41 % higher than the average NH3 concentration of other seasons. Furthermore, the NH3concentration in winter (5.0 ± 3.7 μg m−3; n = 2113) was similar to that in spring (5.1 ± 3.8 μg m−3; n = 2198) but slightly higher, on average, than that in autumn (4.5 ± 2.3 μg m−3; n = 1949). Moreover, other meteorological parameters like planetary boundary layer height and relative humidity were not major factors affecting seasonal NH3 concentrations. These findings suggest that there may be some climate-independent NH3 sources present in the Shanghai urban area. Independent of season, the concentrations of both NH3 and CO present a marked bimodal diurnal profile, with maxima in the morning and the evening. A spatial analysis suggests that elevated concentrations of NH3 are often associated with transport from regions west–northwest and east–southeast of the city, areas with dense road systems. The spatial origin of NH3and the diurnal concentration profile together suggest the importance of vehicle-derived NH3 associated with daily commuting in the urban environment. To further examine vehicular NH3 emissions and transport, sampling of the NH3 concentration was performed in (from the entrance to the exit of the tunnel) and out (along a roadside transect spanning 310 m perpendicular to the tunnel) of a heavily trafficked urban tunnel during the spring of 2014. NH3 concentrations in

the tunnel exit were over 5 and 11 times higher than those in the tunnel entrance and in the ambient air, respectively. Based on the derived mileage-based NH3 emission factor of 28 mg km−1, a population of 3.04 million vehicles in Shanghai produced around 1300 t NH3 in 2014, which accounts for 12 % of total NH3 emissions in the urban area. Collectively, our results clearly show that vehicle emissions associated with combustion are an important NH3 source in Shanghai urban areas and may have potential implications for PM2.5 pollution in the urban atmosphere.

Source apportionment vs. emission inventories of non-methane hydrocarbons (NMHC) in an urban area of the Middle East: local and global perspectives

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We applied the positive matrix factorization model to two large data sets collected during two intensive measurement campaigns (summer 2011 and winter 2012) at a sub-urban site in Beirut, Lebanon, in order to identify NMHC (non-methane hydrocarbons) sources and quantify their contribution to ambient levels. Six factors were identified in winter and five factors in summer. PMF-resolved source profiles were consistent with source profiles established by near-field measurements. The major sources were traffic-related emissions (combustion and gasoline evaporation) in winter and in summer accounting for 51 and 74 wt %, respectively, in agreement with the national emission inventory. The gasoline evaporation related to traffic source had a significant contribution regardless of the season (22 wt % in winter and 30 wt % in summer). The NMHC emissions from road transport are estimated from observations and PMF results, and compared to local and global emission inventories. The PMF analysis finds reasonable differences on emission rates, of 20–39 % higher than the national road transport inventory. However, global inventories (ACCMIP, EDGAR, MACCity) underestimate the emissions up to a factor of 10 for the transportation sector. When combining emission inventory to our results, there is strong evidence that control measures in Lebanon should be targeted on mitigating the NMHC emissions from the traffic-related sources. From a global perspective, an assessment of VOC (volatile organic compounds) anthropogenic emission inventories for the Middle East region as a whole seems necessary as these emissions could be much higher than expected at least from the road transport sector.

Validation of the Swiss methane emission inventory by atmospheric observations and inverse modeling

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Source: *Atmos. Chem. Phys., 16, 3683-3710, 2016*
http://www.atmos-chem-phys.net/16/3683/2016/doi:10.5194/acp-16-3683-2016

Atmospheric inverse modelling has the potential to provide observation-based estimates of greenhouse gas emissions at the country scale, thereby allowing for an independent validation of national emission inventories. Here, we present a regional-scale inverse modelling study to quantify the emissions of methane (CH4) from Switzerland, making use of the newly established CarboCount-CH measurement network and a high-resolution Lagrangian transport model. In our reference inversion, prior emissions were taken from the "bottom-up" Swiss Greenhouse Gas Inventory (SGHGI) as published by the Swiss Federal Office for the Environment in 2014 for the year 2012. Overall we estimate national CH4 emissions to be 196 ± 18 Gg yr−1 for the year 2013 (1σ uncertainty). This result is in close agreement with the recently revised SGHGI estimate of 206 ± 33 Gg yr−1 as reported in 2015 for the year 2012. Results from sensitivity inversions using alternative prior emissions, uncertainty covariance settings, large-scale background mole fractions, two different inverse algorithms (Bayesian and extended Kalman filter), and two different transport models confirm the robustness and independent character of our estimate. According to the latest SGHGI estimate the main CH4 source categories in Switzerland are agriculture (78 %), waste handling (15 %) and natural gas distribution and combustion (6 %). The spatial distribution and seasonal variability of our posterior emissions suggest an overestimation of agricultural CH4 emissions by 10 to 20 % in the most recent SGHGI, which is likely due to an overestimation of emissions from manure handling. Urban areas do not appear as emission hotspots in our posterior results, suggesting that leakages from natural gas distribution are only a minor source of CH4 in Switzerland. This is consistent with rather low emissions of 8.4 Gg yr−1 reported by the SGHGI but inconsistent with the much higher value of 32 Gg yr−1implied by the EDGARv4.2 inventory for this sector. Increased CH4 emissions (up to 30 % compared to the prior) were deduced for the north-eastern parts of Switzerland. This feature was common to most sensitivity inversions, which is a strong indicator that it is a real feature and not an artefact of the transport model and the inversion system. However, it was not possible to assign an unambiguous source process to the region. The observations of the CarboCount-CH network provided invaluable and independent information for the validation of the national bottom-up inventory. Similar systems need to be sustained to provide independent monitoring of future climate agreements.

In situ measurements and modeling of reactive trace gases in a small biomass burning plume

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http://www.atmos-chem-phys.net/16/3813/2016/doi:10.5194/acp-16-3813-2016

An instrumented NASA P-3B aircraft was used for airborne sampling of trace gases in a plume that had emanated from a small forest understory fire in Georgia, USA. The plume was sampled at its origin to derive emission factors and followed ∼ 13.6 km downwind to observe chemical changes during the first hour of atmospheric aging. The P-3B payload included a proton-transfer-reaction time-of-flight mass spectrometer (PTR-ToF-MS), which measured non-methane organic gases (NMOGs) at unprecedented spatiotemporal resolution (10 m spatial/0.1 s temporal). Quantitative emission data are reported for CO2, CO, NO, NO2, HONO, NH3, and 16 NMOGs (formaldehyde, methanol, acetonitrile, propene, acetaldehyde, formic acid, acetone plus its isomer propanal, acetic acid plus its isomer glycolaldehyde, furan, isoprene plus isomeric pentadienes and cyclopentene, methyl vinyl ketone plus its isomers crotonaldehyde and methacrolein, methylglyoxal, hydroxy acetone plus its isomers methyl acetate and propionic acid, benzene, 2,3-butanedione, and 2-furfural) with molar emission ratios relative to CO larger than 1 ppbV ppmV−1. Formaldehyde, acetaldehyde, 2-furfural, and methanol dominated NMOG emissions. No NMOGs with more than 10 carbon atoms were observed at mixing ratios larger than 50 pptV ppmV−1 CO. Downwind plume chemistry was investigated using the observations and a 0-D photochemical box model simulation. The model was run on a nearly explicit chemical mechanism (MCM v3.3) and initialized with measured emission data. Ozone formation during the first hour of atmospheric aging was well captured by the model, with carbonyls (formaldehyde, acetaldehyde, 2,3-butanedione, methylglyoxal, 2-furfural) in addition to CO and CH4 being the main drivers of peroxy radical chemistry. The model also accurately reproduced the sequestration of NOx into peroxyacetyl nitrate (PAN) and the OH-initiated degradation of furan and 2-furfural at an average OH concentration of 7.45 ± 1.07 × 106 cm−3 in the plume. Formaldehyde, acetone/propanal, acetic acid/glycolaldehyde, and maleic acid/maleic anhydride (tentatively identified) were found to be the main NMOGs to increase during 1 h of atmospheric plume processing, with the model being unable to capture the observed increase. A mass balance analysis suggests that about 50 % of the aerosol mass formed in the downwind plume is organic in nature.

Significant increase of surface ozone at a rural site, north of eastern China

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http://www.atmos-chem-phys.net/16/3969/2016/doi:10.5194/acp-16-3969-2016

Ozone pollution in eastern China has become one of the top environmental issues. Quantifying the temporal trend of surface ozone helps to assess the impacts of the anthropogenic precursor reductions and the likely effects of emission control strategies implemented. In this paper, ozone data collected at the Shangdianzi (SDZ) regional atmospheric background station from 2003 to 2015 are presented and analyzed to obtain the variation in the trend of surface ozone in the most polluted region of China, north of eastern China or the North China Plain. A modified Kolmogorov–Zurbenko (KZ) filter method was performed on the maximum daily average 8 h (MDA8) concentrations of ozone to separate the contributions of different factors from the variation of surface ozone and remove the influence of meteorological fluctuations on surface ozone. Results reveal that the short-term, seasonal and long-term components of ozone account for 36.4, 57.6 and 2.2 % of the total variance, respectively. The long-term trend indicates that the MDA8 has undergone a significant increase in the period of 2003–2015, with an average rate of 1.13 ± 0.01 ppb year−1 (R2 = 0.92). It is found that meteorological factors did not significantly influence the long-term variation of ozone and the increase may be completely attributed to changes in emissions. Furthermore, there is no significant correlation between the long-term O3 and NO2 trends. This study suggests that emission changes in VOCs might have played a more important role in the observed increase of surface ozone at SDZ.

Atmospheric constraints on the methane emissions from the East Siberian Shelf

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Source*: Atmos. Chem. Phys., 16, 4147-4157, 2016*
http://www.atmos-chem-phys.net/16/4147/2016/doi:10.5194/acp-16-4147-2016

Subsea permafrost and hydrates in the East Siberian Arctic Shelf (ESAS) constitute a substantial carbon pool, and a potentially large source of methane to the atmosphere. Previous studies based on interpolated oceanographic campaigns estimated atmospheric emissions from this area at 8–17 TgCH4 yr−1. Here, we propose insights based on atmospheric observations to evaluate these estimates. The comparison of high-resolution simulations of atmospheric methane mole fractions to

continuous methane observations during the whole year 2012 confirms the high variability and heterogeneity of the methane releases from ESAS. A reference scenario with ESAS emissions of 8 TgCH4 yr−1, in the lower part of previously estimated emissions, is found to largely overestimate atmospheric observations in winter, likely related to overestimated methane leakage through sea ice. In contrast, in summer, simulations are more consistent with observations. Based on a comprehensive statistical analysis of the observations and of the simulations, annual methane emissions from ESAS are estimated to range from 0.0 to 4.5 TgCH4 yr−1. Isotopic observations suggest a biogenic origin (either terrestrial or marine) of the methane in air masses originating from ESAS during late summer 2008 and 2009.

Stratospheric ozone changes under solar geoengineering: implications for UV exposure and air quality

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http://www.atmos-chem-phys.net/16/4191/2016/doi:10.5194/acp-16-4191-2016

Various forms of geoengineering have been proposed to counter anthropogenic climate change. Methods which aim to modify the Earth's energy balance by reducing insolation are often subsumed under the term solar radiation management (SRM). Here, we present results of a standard SRM modelling experiment in which the incoming solar irradiance is reduced to offset the global mean warming induced by a quadrupling of atmospheric carbon dioxide. For the first time in an atmosphere–ocean coupled climate model, we include atmospheric composition feedbacks for this experiment. While the SRM scheme considered here could offset greenhouse gas induced global mean surface warming, it leads to important changes in atmospheric composition. We find large stratospheric ozone increases that induce significant reductions in surface UV-B irradiance, which would have implications for vitamin D production. In addition, the higher stratospheric ozone levels lead to decreased ozone photolysis in the troposphere. In combination with lower atmospheric specific humidity under SRM, this results in overall surface ozone concentration increases in the idealized G1 experiment. Both UV-B and surface ozone changes are important for human health. We therefore highlight that both stratospheric and tropospheric ozone changes must be considered in the assessment of any SRM scheme, due to their important roles in regulating UV exposure and air quality.

Understanding the recent trend of haze pollution in eastern China: roles of climate change

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In this paper, the variation and trend of haze pollution in eastern China for winter of 1960–2012 were analyzed. With the overall increasing number of winter haze days in this period, the 5 decades were divided into three sub-periods based on the changes of winter haze days (WHD) in central North China (30–40° N) and eastern South China (south of 30° N) for east of 109° E mainland China. Results show that WHD kept gradually increasing during 1960–1979, remained stable overall during 1980–1999, and increased fast during 2000–2012. The author identified the major climate forcing factors besides total energy consumption. Among all the possible climate factors, variability of the autumn Arctic sea ice extent, local precipitation and surface wind during winter is most influential to the haze pollution change. The joint effect of fast increase of total energy consumption, rapid decline of Arctic sea ice extent and reduced precipitation and surface winds intensified the haze pollution in central North China after 2000. There is a similar conclusion for haze pollution in eastern South China after 2000, with the precipitation effect being smaller and spatially inconsistent.

Summertime ozone formation in Xi'an and surrounding areas, China

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http://www.atmos-chem-phys.net/16/4323/2016/doi:10.5194/acp-16-4323-2016

In this study, the ozone (O3) formation in China's northwest city of Xi'an and surrounding areas is investigated using the Weather Research and Forecasting atmospheric chemistry (WRF-Chem) model during the period from 22 to 24 August 2013, corresponding to a heavy air pollution episode with high concentrations of O3 and PM2.5. The model generally performs well compared to measurements in simulating the surface temperature, relative humidity, and wind speed and direction, near-surface O3 and PM2.5 mass concentrations, and aerosol constituents. High aerosol concentrations in Xi'an and surrounding areas significantly decrease the photolysis frequencies and can reduce O3 concentrations by more than 50 μg m−3 (around 25 ppb) on average. Sensitivity studies show that the O3 production regime in Xi'an and surrounding areas is complicated, varying from NOx to VOC (volatile organic compound)-sensitive chemistry. The industrial emissions contribute the most to the O3concentrations compared to biogenic and other anthropogenic

sources, but neither individual anthropogenic emission nor biogenic emission plays a dominant role in the O3 formation. Under high O3 and PM2.5 concentrations, a 50 % reduction in all the anthropogenic emissions only decreases near-surface O3 concentrations by about 14 % during daytime. The complicated O3 production regime and high aerosol levels pose a challenge for O3 control strategies in Xi'an and surrounding areas. Further investigation regarding O3 control strategies will need to be performed, taking into consideration the rapid changes in anthropogenic emissions that are not reflected in the current emission inventories and the uncertainties in the meteorological field simulations.

Hotspot of glyoxal over the Pearl River delta seen from the OMI satellite instrument: implications for emissions of aromatic hydrocarbons

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Source: *Atmos. Chem. Phys., 16, 4631-4639, 2016*
http://www.atmos-chem-phys.net/16/4631/2016/doi:10.5194/acp-16-4631-2016

The Pearl River delta (PRD) is a densely populated hub of industrial activity located in southern China. OMI (Ozone Monitoring Instrument) satellite observations reveal a large hotspot of glyoxal (CHOCHO) over the PRD that is almost twice as large as any other in Asia. Formaldehyde (HCHO) and NO2 observed by OMI are also high in the PRD but no more than in other urban/industrial areas of China. The CHOCHO hotspot over the PRD can be explained by industrial paint and solvent emissions of aromatic volatile organic compounds (VOCs), with toluene being a dominant contributor. By contrast, HCHO in the PRD originates mostly from VOCs emitted by combustion (principally vehicles). By applying a plume transport model to wind-segregated OMI data, we show that the CHOCHO and HCHO enhancements over the PRD observed by OMI are consistent with current VOC emission inventories. Prior work using CHOCHO retrievals from the SCIAMACHY satellite instrument suggested that emission inventories for aromatic VOCs in the PRD were too low by a factor of 10–20; we attribute this result in part to bias in the SCIAMACHY data and in part to underestimated CHOCHO yields from oxidation of aromatics. Our work points to the importance of better understanding CHOCHO yields from the oxidation of aromatics in order to interpret space-based CHOCHO observations in polluted environments.

Aura OMI observations of regional SO2 and NO2 pollution changes from 2005 to 2015

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The Ozone Monitoring Instrument (OMI) onboard NASA's Aura satellite has been providing global observations of the ozone layer and key atmospheric pollutant gases, such as nitrogen dioxide (NO2) and sulfur dioxide (SO2), since October 2004. The data products from the same instrument provide consistent spatial and temporal coverage and permit the study of anthropogenic and natural emissions on local-to-global scales. In this paper, we examine changes in SO2 and NO2 over some of the world's most polluted industrialized regions during the first decade of OMI observations. In terms of regional pollution changes, we see both upward and downward trends, sometimes in opposite directions for NO2and SO2, for different study areas. The trends are, for the most part, associated with economic and/or technological changes in energy use, as well as regional regulatory policies. Over the eastern US, both NO2 and SO2 levels decreased dramatically from 2005 to 2015, by more than 40 and 80 %, respectively, as a result of both technological improvements and stricter regulations of emissions. OMI confirmed large reductions in SO2 over eastern Europe's largest coal-fired power plants after installation of flue gas desulfurization devices. The North China Plain has the world's most severe SO2 pollution, but a decreasing trend has been observed since 2011, with about a 50 % reduction in 2012–2015, due to an economic slowdown and government efforts to restrain emissions from the power and industrial sectors. In contrast, India's SO2 and NO2 levels from coal power plants and smelters are growing at a fast pace, increasing by more than 100 and 50 %, respectively, from 2005 to 2015. Several SO2 hot spots observed over the Persian Gulf are probably related to oil and gas operations and indicate a possible underestimation of emissions from these sources in bottom-up emission inventories. Overall, OMI observations have proved valuable in documenting rapid changes in air quality over different parts of the world during last decade. The baseline established during the first 11 years of OMI is indispensable for the interpretation of air quality measurements from current and future satellite atmospheric composition missions.

Photochemical age of air pollutants, ozone, and secondary organic aerosol in transboundary air observed on Fukue Island, Nagasaki, Japan

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http://www.atmos-chem-phys.net/16/4555/2016/doi:10.5194/acp-16-4555-2016

To better understand the secondary air pollution in transboundary air over westernmost Japan, ground-based field measurements of the chemical composition of fine particulate matter (≤ 1 μm), mixing ratios of trace gas species (CO, O3, NOx, NOy, i-pentane, toluene, and ethyne), and meteorological elements were conducted with a suite of instrumentation. The CO mixing ratio dependence on wind direction showed that there was no significant influence from primary emission sources near the monitoring site, indicating long- and/or mid-range transport of the measured chemical species. Despite the considerably different atmospheric lifetimes of NOy and CO, these mixing ratios were correlated (r2 = 0.67). The photochemical age of the pollutants, t[OH] (the reaction time × the mean concentration of OH radical during the atmospheric transport), was calculated from both the NOx/NOy concentration ratio (NOx/NOy clock) and the toluene/ethyne concentration ratio (hydrocarbon clock). It was found that the toluene / ethyne concentration ratio was significantly influenced by dilution with background air containing 0.16 ppbv of ethyne, causing significant bias in the estimation of t[OH]. In contrast, the influence of the reaction of NOx with O3, a potentially biasing reaction channel on [NOx] / [NOy], was small. The t[OH] values obtained with the NOx/NOy clock ranged from 2.9 × 105 to 1.3 × 108 h molecule cm−3 and were compared with the fractional contribution of the m⁄z 44 signal to the total signal in the organic aerosol mass spectra (f44, a quantitative oxidation indicator of carboxylic acids) and O3 mixing ratio. The comparison of t[OH] with f44 showed evidence for a systematic increase of f44 as t[OH] increased, an indication of secondary organic aerosol (SOA) formation. To a first approximation, the f44increase rate was (1.05 ± 0.03) × 10−9 × [OH] h−1, which is comparable to the background-corrected increase rate observed during the New England Air Quality Study in summer 2002. The similarity may imply the production of similar SOA component, possibly humic-like substances. Meanwhile, the comparison of t[OH] with O3 mixing ratio showed that there was a strong proportional relationship between O3 mixing ratio and t[OH]. A first approximation gave the increasing rate and background mixing ratio of ozone as (3.48 ± 0.06) × 10−7 × [OH] ppbv h−1 and 30.7 ppbv, respectively. The information given here can be used for prediction of secondary pollution magnitude in the outflow from the Asian continent.

Mercury dynamics and mass balance in a subtropical forest, southwestern China

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The mid-subtropical forest area in southwest China was affected by anthropogenic mercury (Hg) emissions over the past 3 decades. We quantified mercury dynamics on the forest field and measured fluxes and pools of Hg in litterfall, throughfall, stream water and forest soil in an evergreen broadleaved forest field in southwestern China. Total Hg (THg) input by the throughfall and litterfall was assessed at 32.2 and 42.9 μg m−2 yr−1, respectively, which was remarkably higher than those observed from other forest fields in the background of North America and Europe. Hg fluxes across the soil–air interface (18.6 mg m−2 yr−1) and runoff and/or stream flow (7.2 μg m−2 yr−1) were regarded as the dominant ways for THg export from the forest field. The forest field hosts an enormous amount of atmospheric Hg, and its reserves is estimated to be 25 341 μg m2. The ratio of output to input Hg fluxes (0.34) is higher compared with other study sites. The higher output / input ratio may represent an important ecological risk for the downstream aquatic ecosystems, even if the forest field could be an effective sink of Hg.

Characteristics of monsoon inversions over the Arabian Sea observed by satellite sounder and reanalysis data sets

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Source: *Atmos. Chem. Phys., 16, 4497-4509, 2016*
http://www.atmos-chem-phys.net/16/4497/2016/doi:10.5194/acp-16-4497-2016

Monsoon inversion (MI) over the Arabian Sea (AS) is one of the important characteristics associated with the monsoon activity over Indian region during summer monsoon season. In the present study, we have used 5 years (2009–2013) of temperature and water vapour measurement data obtained from satellite sounder instrument, an Infrared Atmospheric Sounding Interferometer (IASI) onboard MetOp satellite, in addition to ERA-Interim data, to study their characteristics. The lower atmospheric data over the AS have been examined first to identify the areas where MIs are predominant and occur with higher strength. Based on this information, a detailed study has been made to investigate their characteristics separately in the eastern AS (EAS) and western AS (WAS) to examine their contrasting features. The initiation and dissipation times of MIs, their percentage occurrence, strength, etc., has been examined using the huge database. The relation with monsoon activity (rainfall) over Indian region during normal and poor monsoon years is also studied. WAS ΔT values are $\sim$ 2 K less than those over the EAS, ΔT being the temperature difference between 950 and 850 hPa. A much larger contrast between the WAS and EAS in ΔT is noticed in ERA-Interim data set vis-à-vis those observed by satellites. The possibility of detecting MI from another parameter, refractivity N, obtained directly from another satellite constellation of GPS Radio Occultation (RO) (COSMIC), has also been examined. MI detected from IASI and Atmospheric Infrared Sounder (AIRS) onboard the NOAA satellite have been compared to see how far the two data sets can be combined to study the MI characteristics. We suggest MI could also be included as one of the semipermanent features of southwest monsoon along with the presently accepted six parameters

The vertical distribution of volcanic SO2 plumes measured by IASI

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Source: *Atmos. Chem. Phys., 16, 4343-4367, 2016*
http://www.atmos-chem-phys.net/16/4343/2016/doi:10.5194/acp-16-4343-2016

Sulfur dioxide (SO2) is an important atmospheric constituent that plays a crucial role in many atmospheric processes. Volcanic eruptions are a significant source of atmospheric SO2 and its effects and lifetime depend on the SO2 injection altitude. The Infrared Atmospheric Sounding Interferometer (IASI) on the METOP satellite can be used to study volcanic emission of SO2 using high-spectral resolution measurements from 1000 to 1200 and from 1300 to 1410 cm−1 (the 7.3 and 8.7 μm SO2 bands) returning both SO2 amount and altitude data. The scheme described in Carboni et al. (2012) has been applied to measure volcanic SO2 amount and altitude for 14 explosive eruptions from 2008 to 2012. The work includes a comparison with the following independent measurements: (i) the SO2 column amounts from the 2010 Eyjafjallajökull plumes have been compared with Brewer ground measurements over Europe; (ii) the SO2 plumes heights, for the 2010 Eyjafjallajökull and 2011 Grimsvötn eruptions, have been compared with CALIPSO backscatter profiles. The results of the comparisons show that IASI SO2 measurements are not affected by underlying cloud and are consistent (within the retrieved errors) with the other measurements. The series of analysed eruptions (2008 to 2012) show that the biggest emitter of volcanic SO2 was Nabro, followed by Kasatochi and Grímsvötn. Our observations also show a tendency for volcanic SO2 to reach the level of the tropopause during many of the moderately explosive eruptions observed. For the eruptions observed, this tendency was independent of the maximum amount of SO2 (e.g. 0.2 Tg for Dalafilla compared with 1.6 Tg for Nabro) and of the volcanic explosive index (between 3 and 5).

Influence of tropical cyclones on tropospheric ozone: possible implications

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Source*: Atmos. Chem. Phys., 16, 4837-4847, 2016*
http://www.atmos-chem-phys.net/16/4837/2016/doi:10.5194/acp-16-4837-2016

The present study examines the role of tropical cyclones in the enhancement of tropospheric ozone. The most significant and new observation reported is the increase in the upper-tropospheric (10–16 km) ozone by 20–50 ppbv, which has extended down to the middle (6–10 km) and lower troposphere (< 6 km). The descent rate of enhanced ozone layer during the passage of tropical cyclone is 0.8–1 km day−1, which is three times that of a clear-sky day (non-convective). Enhancement of surface ozone concentration by ∼ 10 ppbv in the daytime and 10–15 ppbv in the night-time is observed during a cyclone. Potential vorticity, vertical velocity and potential temperature obtained from numerical simulation, reproduces the key feature of the observations. A simulation study indicates the downward transport of stratospheric air into the troposphere. Space-borne observations of relative humidity indicate the presence of sporadic dry air in the upper and middle troposphere over the cyclonic region. These observations quantitatively constitute experimental evidence of redistribution of stratospheric ozone during cyclonic storms.

Source apportionment of ambient particle number concentrations in central Los Angeles using positive matrix factorization (PMF)

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In this study, the positive matrix factorization (PMF) receptor model (version 5.0) was used to identify and quantify major sources contributing to particulate matter (PM) number concentrations, using PM number size distributions in the range of 13 nm to 10 µm combined with several auxiliary variables, including black carbon (BC), elemental and organic carbon (EC/OC), PM mass concentrations, gaseous pollutants, meteorological, and traffic counts data, collected for about 9 months between August 2014 and 2015 in central Los Angeles, CA. Several parameters, including particle number and volume size distribution profiles, profiles of auxiliary variables, contributions of different factors in different seasons to the total number concentrations, diurnal variations of each of the resolved factors in the cold and warm phases, weekday/weekend analysis for each of the resolved factors, and correlation between auxiliary variables and the relative contribution of each of the resolved factors, were used to identify PM sources. A six-factor solution was identified as the optimum for the aforementioned input data. The resolved factors comprised nucleation, traffic 1, traffic 2 (with a larger mode diameter than traffic 1 factor), urban background aerosol, secondary aerosol, and soil/road dust. Traffic sources (1 and 2) were the major contributor to PM number concentrations, collectively making up to above 60 % (60.8–68.4 %) of the total number concentrations during the study period. Their contribution was also significantly higher in the cold phase compared to the warm phase. Nucleation was another major factor significantly contributing to the total number concentrations (an overall contribution of 17 %, ranging from 11.7 to 24 %), with a larger contribution during the warm phase than in the cold phase. The other identified

factors were urban background aerosol, secondary aerosol, and soil/road dust, with relative contributions of approximately 12 % (7.4–17.1), 2.1 % (1.5–2.5 %), and 1.1 % (0.2–6.3 %), respectively, overall accounting for about 15 % (15.2–19.8 %) of PM number concentrations. As expected, PM number concentrations were dominated by factors with smaller mode diameters, such as traffic and nucleation. On the other hand, PM volume and mass concentrations in the study area were mostly affected by sources with larger mode diameters, including secondary aerosols and soil/road dust. Results from the present study can be used as input parameters in future epidemiological studies to link PM sources to adverse health effects as well as by policymakers to set targeted and more protective emission standards for PM.

Detecting long-term changes in point-source fossil CO2 emissions with tree ring archives

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http://www.atmos-chem-phys.net/16/5481/2016/doi:10.5194/acp-16-5481-2016

We examine the utility of tree ring 14C archives for detecting long-term changes in fossil CO2 emissions from a point source. Trees assimilate carbon from the atmosphere during photosynthesis, in the process faithfully recording the average atmospheric 14C content in each new annual tree ring. Using 14C as a proxy for fossil CO2, we examine interannual variability over six years of fossil CO2 observations between 2004–2005 and 2011–2012 from two trees growing near the Kapuni Gas Treatment Plant in rural Taranaki, New Zealand. We quantify the amount of variability that can be attributed to transport and meteorology by simulating constant point-source fossil CO2 emissions over the observation period with the atmospheric transport model WindTrax. We compare model simulation results to observations and calculate the amount of change in emissions that we can detect with new observations over annual or multi-year time periods, given both the measurement uncertainty of 1ppm and the modelled variation in transport. In particular, we ask, what is the minimum amount of change in emissions that we can detect using this method, given a reference period of six years? We find that changes of 42 % or more could be detected in a new sample from one year at the same observation location or 22 % in the case of four years of new samples. This threshold is reduced and the method becomes more practical the more the size of the signal increases. For point sources 10 times larger than the Kapuni plant (a more typical size for power plants worldwide), it would be possible to detect sustained emissions changes on the order of 10 %, given suitable meteorology and observations.

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