Interannual variations of tropospheric ozone in eastern China: the key role of transport

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We quantify the interannual variations (IAVs) of tropospheric O_3 over China for years 1986-2012 by using the global three-dimensional Goddard Earth Observing System chemical transport model (GEOS-Chem). With variations in both meteorological parameters and emissions, simulated seasonal mean surface-layer O_3 concentrations over North China (NC, 110-120˚E, 32-42˚N), South China (SC, 110-120˚E, 22-32˚N), and Sichuan Basin (SCB, 102-110˚E, 27-33˚N) show large IAVs; the deviations from the mean are in the range of -7.0% to +7.5%, -6.0% to +6.0%, and -9.6% to +7.0% over NC, SC, and SCB, respectively. The IAVs in surface-layer O_3 by variations in meteorological fields are simulated to be larger than those by variations in anthropogenic emissions. Process analyses are performed to identify key meteorological parameters that influence the IAVs of O_3. Over NC and SC, transport flux and chemical production are found to be the first and second important processes that drive the IAVs of O_3 throughout the year, with relative contributions of, respectively, 46-52% and 28-34% over NC and 59-63% and 16-21% in SC. Over SCB, transport is the most dominant process that leads to the IAVs of O_3, with high relative contributions of 58-87% throughout the year. Our results have important implications for the effectiveness of short-term air quality control strategies in China.

Keywords: Tropospheric ozone, Transport, Interannual variations, Eastern China, Process analysis

Quantifying nonlinear regional contributions to ozone and fine particles using an updated response surface modeling technique

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Besides local sources, regional emission sources can have substantial contributions to ozone and fine particle concentrations. However, responsiveness of such regional contributions to secondary pollutants is still challenging to simulate. In this study, we used a novel extended response surface modeling (ERSM) technique to quantify the nonlinear regional contributions to ozone and fine particles. The new ERSM (ERSM2.0) was updated from a previous version of ERSM (ERSMv1.0) by adding an explicit representation of interregional effects among multiple regions. The new ERSMv2.0 model was successfully applied in a case study in Beijing-Tianjin-Hebei region in China, exhibiting a better performance in reproducing the nonlinearity in the response of ozone and $PM_{2.5}$ to precursors compared to the previous versions of ERSMv1.0.
The new ERSMv2.0 model presents abilities in attributing the regional contributions to either 1) directly transporting the pollutant from source area to receptor area, or 2) transporting precursors from source area and produce the secondary pollutant at receptor area. In the case study of Beijing-Tianjin-Hebei, we apportioned the ozone and $PM_{2.5}$ responses to three components, i.e., local chemistry, regional transport and indirect effects. Results suggest that in most cases, the apportion of $PM_{2.5}$ formed locally in the receptor region is more than that of regional transport of $PM_{2.5}$ formed outside the receptor region. The opposite way is found for ozone that the regional sources contribute more than formed at local. Besides, the contribution from regional sources becomes larger during more polluted episodes for both ozone and $PM_{2.5}$, suggesting the importance of joint controls on regional sources for reducing the highest pollution level.

Keywords: response surface model, regional contribution, ozone, fine particles

Source Contributions to Secondary Organic Aerosol in China: Comparison of MEIC and REAS2

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A source-oriented version of the Community Multiscale Air Quality Model (CMAQ) was developed to simulate the formation of secondary organic aerosol (SOA) and determine the contributions of different anthropogenic and biogenic sources to the predicted SOA in China in the entire year of 2013. The SOA module in the source-oriented CMAQ model is based on a vanilla version of the SOA module, which tracks the formation and equilibrium partitioning of semi-volatile products from precursors of long chain alkanes, aromatic compounds, monoterpenes and sesquiterpenes and from reactive surface uptake of glyoxal, methylglyoxal, isoprene epoxydiols (IEPOX) and methacrylic acid epoxide (MAE). The source-oriented version tracks precursors and their oxidation products from different sources simultaneously to determine their contributions to SOA. To explore the uncertainty in SOA concentration and source contributions due to different inventories, two popular emission inventories, the Multi-resolution Emission Inventory for China (MEIC) developed by the Tsinghua University and the Regional Emission inventory in Asia version 2 (REAS2) developed by the National Institute for Environmental Studies in Japan. While the total monthly emissions of precursor species in China from the two inventories are close to each other, leading to similar levels of SOA, emission rates from different source sectors are quite different. For example, for the aromatic compounds, REAS2 suggests that solvent utilization and transportation sectors are the two most important sources but MEIC shows majority of the emissions are from industry sources. These differences in emissions lead to different estimations of source contributions to SOA, particularly in the winter when biogenic contributions are much smaller in most part of China. Summer time SOA is mostly due to biogenic emissions of isoprene, thus the difference in anthropogenic contributions are limited to a small extend. The results from this analysis suggests that significant uncertainties still exist in current emission estimations for China.

Keywords: Secondary Organic Aerosol, Source Apportionment, Emission Inventory, CMAQ

Source Tagging Modeling Study of Heavy Haze Episodes under Complex Regional Transport Processes over Wuhan Megacity, Central China

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This study carried on the Nested Air Quality Prediction Modeling System (NAQPMS) coupled with an on-line source-tagging module, aims at delving into the role of regional transport in the occurrence of two strong haze episodes over Wuhan in October 2014 and quantitatively assessing the contributions from potential local and regional sources to $PM_{2.5}$ concentration. Validation of predictions based on observations shows modeling system good skills in reproducing key meteorological and chemical features. Different types of the regional transport were found to play key roles in the formation of the two haze episodes. The first short-time haze episode occurring on 12 October under strong northerly wind, with hourly $PM_{2.5}$ concentration maximum of 180 $\mu g m^{-3}$ is found to be mainly triggered by the long-range transport from the northern regions, contributing to $PM_{2.5}$ concentration by 60.6% versus a total of 32.7% for Wuhan local and nearby sources. For the second episode extending over 15-20 October under stable regional large-scale synoptic condition and weak winds and with hourly $PM_{2.5}$ peak of 231.0 $\mu g m^{-3}$, both the long-range transport from far regions and the short-range transport from Wuhan Megalopolis are the main causes of the haze episode and account for 24.8% and 29.2% of the $PM_{2.5}$ concentration respectively. Therefore, the regional transport acts as a crucial driver of haze pollution over Wuhan through not only long-range transfer of pollutants, but also short-range aerosol movement associated with complex interaction and stagnation under specific meteorological conditions. The present findings incontestably call for enhancing enforcement of strategic environmental assessment with commitment of stakeholders at both local and regional scales.

Keywords: Regional chemical transport, Wuhan, Central China, Haze pollution, Source tagging method

Shipping emissions and its contribution to port air quality by an ongoing field campaigns (SEISO-Bohai)

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East Asia has the most rapidly growing shipping emissions of both CO2 and traditional air pollutants, but the least in-depth analysis. Recent study shows that shipping emissions in East Asia accounted for 16% of global shipping CO2 in 2013, leading to large adverse health impacts with 14,500-37,500 premature deaths per year. Wintertime air quality was measured in port area as part of the Shipping Emission and Impacts by Switching Oil in Bohai Bay SEISO-Bohai campaign during December 2016-January 2017. SEISO-Bohai is a field campaign focusing on shipping emission and its contribution to air quality research. It was taken place at a site in a port of Bohai Bay. The site is located close enough to the ships while far from other sources. The particle and gas inlets were installed on the roof of a measurement container at 2.8 m above ground level. Inside the container, the particle speciation was measured with a Single Particle Aerosol Mass Spectrometer. The organic gases were measured by SIFT-MS, a high-sensitivity instrument with fast time response that employs a quadrupole mass spectrometer to measure volatile organic compounds. PM samples were collected by mid-volume samplers. The pre-fired quartz microfiber filters was used to determine organic and elemental carbon. The quartz microfiber filters were conditioned for 24 h before the sampling, then weighted by a five-digit microbalance. Meteorology conditions were continuously monitored with 1 min time resolution, including temperature and relative humidity at 4.8m height, wind speed and direction and pressure at station level. Carbon monoxide, the tracer for combustion emissions, was measured by vacuum ultra-violet fluorescence.
Port area reflects a mixture of regional background and nearby shipping and truck emissions. Both the strong regional influence of haze event and shipping emission influence were observed. Shipping emission flumes provide basic idea to check the compliance of sulfur content of shipping fuel for several container vessels. The measured SO2 and CO2 concentrations show that the fuel sulfur contents may be lower than previous estimation. A profile of PM from shipping exhaust was built, with major species of OM, SO4, BC and elements, such as S, V, and Fe. The primary contribution of ship emissions to harbor PM2.5 concentrations will be determined after all data were processed.

Keywords: shipping emission, China, air quality, field campaigns, SEISO-Bohai

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

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

Keywords: emission inventory, air quality modeling, China

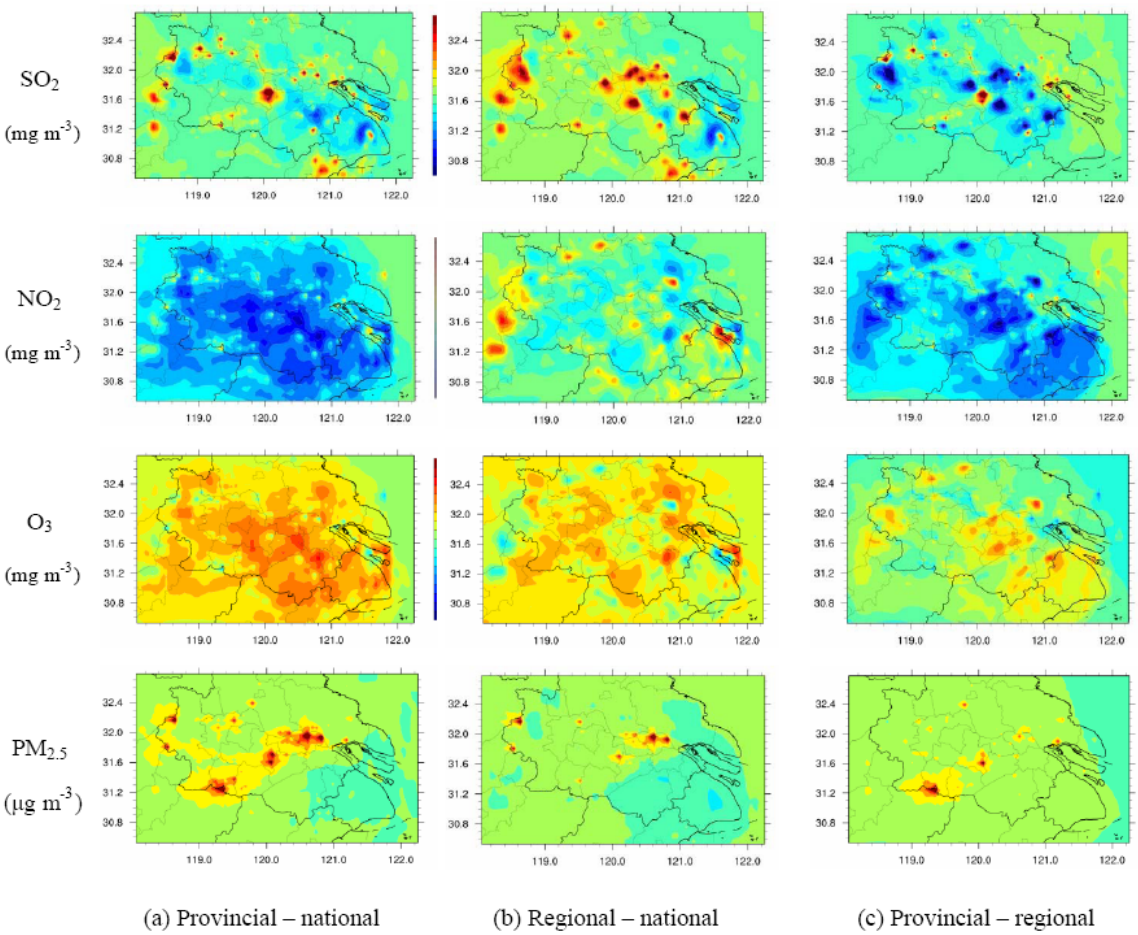

Figure The differences in the monthly means of simulated SO_2, NO_2, $PM_{2.5}$, and O_3 concentrations using different emission inventories: (a) provincial–national; (b) regional–national; and (c) provincial–regional. The black star A and triangle B referred to the locations of grids with maximum SO_2 emissions in provincial and regional inventory.

Unintentional Regional Ozone Increase in the Western Pacific Due to Particulate Matter Controls in China

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China has been suffering severe particulate matter (PM) and ozone (O3) pollution due to large amount of fossil fuel consumption associated with rapid economic growth, industrialization, and urbanization during the past four decades. Air pollution control measures have recently been taken into action to reduce the levels of air pollutants, primarily focusing on PM2.5 by reducing SO2 and industrial smoke and dust emissions. Recent monitoring data shows that declining trends in PM2.5 in China, reflecting the effectiveness of PM2.5 control measures. However, ozone, the major pollutant of photo chemical smog which also causes serious damages to public health and agricultural products, is becoming more serious meanwhile. Although particles are not the precursors of O3, particles scatter and absorb the sunlight, reduce the UV radiation and consequently suppress O3 formation. Declining trends in radiation has been found in observations for the last half century, and studies indicate that serious PM pollution is responsible for some of the radiation reduction. Theoretically, the current PM controls lower the PM concentrations, and thus would increase the radiation and enhance O3 formation.

To investigate the impact of PM and SO2 emission controls in China on the ground level O3 formation, we used Community Multi-scale Air Quality Model (CMAQv5.0.1) to simulated O3 concentrations under different PM and SO2 emission control scenarios. We simulated 2013 summer (June, July, and August) with a 36-km horizontal resolution East Asia region. We used the online photolysis rate module to calculate the effects of scattering and absorbing aerosols in modulating photolysis rates. The results show that seasonal average 8h peak O3 is predicted to increase with the reduction in the SO2 and PM emissions in the NCP area and its downwind ocean areas. The maximum 1h peak O3 concentrations in the three months are over 10 ppbv in the center and east China. The largest increase of 45 ppbv in 1h O3 concentration is over the east China sea, downwind of Shanghai and Jiangsu province. Moreover, the PM and SO2 controls also may greatly increase the 1h O3 concentrations (~15-25 ppbv) in Korea and Japan, even though the seasonal average increase of 8h maximum O3 is not significant (~0.5-1.5ppbv).

Keywords: ozone, air quality modeling, emission control

Impact of urban land surface and black carbon on ozone chemistry

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Urbanization affects air quality mainly through two ways. First, the meteorological fields and surface properties changed by urban land surfaces alter the spatial distribution, chemical reaction conditions, and deposition of primary and secondary air pollutants. Second, urbanization processes increase local human activities and hence increase anthropogenic emissions of air pollutants. Both types of effects have crucial and complex impacts on air qualities in urban and surrounding areas. In this study, the impact of upstream urban land surface forcing on downstream photochemistry and the impacts of black carbon (BC) on surface ozone were conducted by numerical simulations in summer season over megacities of Yangtze River Delta.

The results show that the upstream urban Shanghai has a significant impact on the boundary layer structures and circulations over Kunshan and further affects Kunshan’ s O_3 air quality by redistributing O_3 and its precursors. Horizontal transport of O_3 and its precursors, from Shanghai to Kunshan, are suppressed in the lower boundary layer but are strengthened in the upper boundary layer because of strong urban heat island circulation. As a result, O_3 chemical production is a little decreased in the lower boundary layer of Kunshan (~2 ppbv) but is increased significantly in the upper boundary layer (~40 ppbv).

With the impact of BC, surface ozone concentration reduced by more than 10% in the morning (10:00~12:00). High contribution of vertical mixing process demonstrates that the depression of boundary layer caused by BC takes important responsibility for the ozone reduction. With the application of online ozone source apportionment method coupled in the WRF-Chem, the changes of surface ozone source-receptor relationship are listed. It shows that, with the impact of BC, the contributions of ozone from Jiangsu (JS), Anhui (AH) and outside the model domain ($O_{3\text{-}INFLOW}$) decreased, whereas ozone contributions from other source regions change little during this period.

Keywords: urbanization, black carbon, ozone chemistry

Increasing wildfire emissions worsen air quality of U.S. megacities by the 2050s

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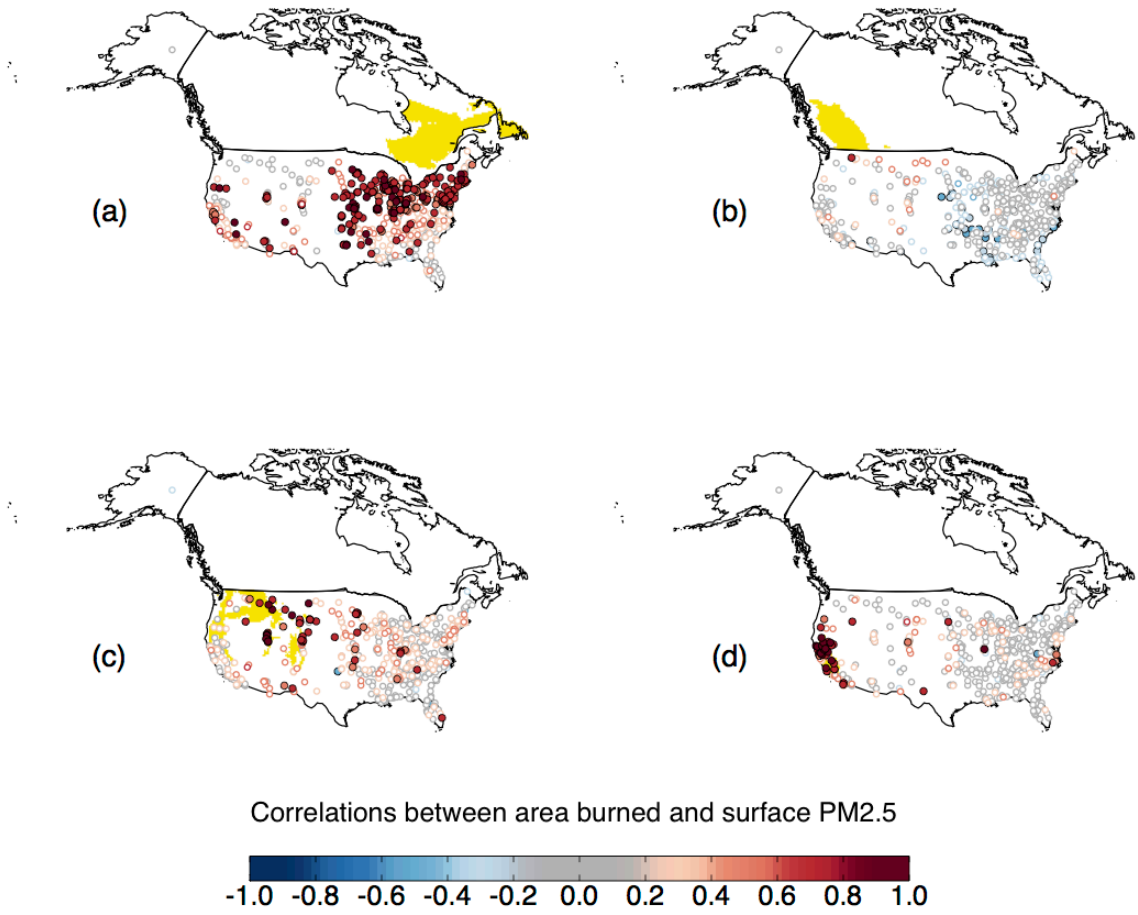
Wildfire emissions can adversely affect air quality locally and downwind. Here, we apply combined chemistry transport modeling (CTM) and multiple observational datasets to quantify the impact of North American wildfire emissions on the air quality of U.S. megacities. First, we analyze records of fire reports over U.S. and Canada and ground measurements of PM2.5 concentrations at 1775 U.S. sites. Composite analyses show distinct responses of surface PM2.5 concentrations to wildfire episodes (area burned at >90^{th} percentile). Western and central cities are mostly influenced by fire events in the western U.S. In contrast, northeastern cities are easily affected by the long-range transport of pollution emitted by Canadian wildfires. Then, we perform sensitivity experiments with the GEOS-Chem CTM and calculate changes in surface PM2.5 concentrations at U.S. megacities with at least half million population. Simulations show that present-day wildfires enhance summer mean PM2.5 concentrations by > 0.5 μg m^{-3} at Chicago, Detroit, San Jose, San Francisco, Seattle, and Portland, the first two of which are influenced by Canadian fire emissions. By the midcentury, fire-induced summer PM2.5 enhancement is as high as 1-3 μg m^{-3} at these megacities, following the largely increased area burned in a warmer climate. The worse air quality due to wildfire emissions pose an emerging threat to the public health in U.S. megacities.

Keywords: megacity, wildfire, climate change, long-range transport, public health



Triple oxygen isotopes indicate that urbanization causes differences in the sources of nitrate between dry and wet atmospheric deposition

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The production of nitrogen oxides by human activities and to lesser extent natural processes significantly influences the chemical composition and reactivity of the troposphere, nitrogen deposition, and ultimately human and environmental health. However, significant uncertainties remain concerning (1) the relative important of the photochemical pathways that transform NO_x to HNO_3 and (2) the relative contributions of local vs. long-distance NO_x emissions to dry vs. wet deposition in various environmental settings. To address these uncertainties we determined the $\Delta^{17}O$ values of wet and dry deposited HNO_3 in 2009 at two sites along the western coast of northern Japan, downwind of the East Asian continent where NO_x emissions have increased approximately four-fold during the past forty years. At a remote site, nitrate $\Delta^{17}O$ values in wet and dry deposition showed similar seasonal variation, ranging between ~23 and 30‰. These results suggesting that both forms of deposition experienced similar photochemical reactions during their formation, with O_3 as the dominant oxidant in winter and a combination of O_3 and OH in summer. In contrast, at an urban site, nitrate $\Delta^{17}O$ values in wet deposition were larger (range of 24-31‰) than those in dry deposition (range of 19-25‰), particularly during the winter. These results suggest an important role of an alternative photochemical pathway for the formation of dry deposition in urban environments: oxidation of NO by peroxy radicals that originate from reactive hydrocarbons. Wet deposition at the urban site likely originates from long-distance transport, whereas most dry deposition likely originates from local NO_x emissions. These results illustrate the value of stable isotope tracers for assessing the sources, transport distances, and sinks of dry and wet atmospheric deposition.

Keywords: nitrate, isotopes, atmospheric deposition, Hokkaido, urban, rural

Characterization and regional transport of $PM_{2.5}$ in different Indian metropolises

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Indian cities are facing severe air pollution with 18 cities among the top 50 most polluted cities in the world as reported by WHO in 2016. This paper studies the seasonal variations of $PM_{2.5}$ from June 2015 to May 2016 in seven Indian metropolises including New Delhi and Lucknow in north India, Patna in east India, Bangalore, Chennai and Hyderabad in south India, and Mumbai in west India. Correlations of $PM_{2.5}$ with other criteria pollutants and meteorological parameters were studied. $PM_{2.5}$ showed a stronger co-relation with NO_2 in winter and SO_2 in monsoon. $PM_{2.5}$ concentrations were the highest during winter and lowest in monsoon except in Chennai where highest concentrations were in monsoon. Cities in northern and eastern India had higher concentrations than other cities. Three days back trajectory was obtained at heights of 500 m to determine the transport of regional sources. Cluster analysis using k-means clustering algorithm was performed for each city. A concentration weighted trajectory analysis was carried out to understand potential regions of higher concentrations. In winter, potential regions are north-west for Delhi, Lucknow and Patna, south-east for Chennai, Hyderabad, and Mumbai, and north-east for Mumbai. The differences between local and regional sources on hours with extreme concentrations were also identified. In winter, significant long range transport is evident for $PM_{2.5}$ in Delhi, Lucknow and Bangalore.

Keywords: India, long range tansport, back trajectory, PM2.5

