

## 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<sub>3</sub> 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<sub>3</sub> 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<sub>3</sub> 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<sub>3</sub>. 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<sub>3</sub> 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<sub>3</sub>, 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<sub>2.5</sub> 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<sub>2.5</sub> concentration maximum of 180  $\mu\text{g m}^{-3}$  is found to be mainly triggered by the long-range transport from the northern regions, contributing to PM<sub>2.5</sub> 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<sub>2.5</sub> peak of 231.0  $\mu\text{g m}^{-3}$ , 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<sub>2.5</sub> 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 CO<sub>2</sub> 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 CO<sub>2</sub> 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 SO<sub>2</sub> and CO<sub>2</sub> 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, SO<sub>4</sub>, BC and elements, such as S, V, and Fe. The primary contribution of ship emissions to harbor PM<sub>2.5</sub> concentrations will be determined after all data were processed.

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

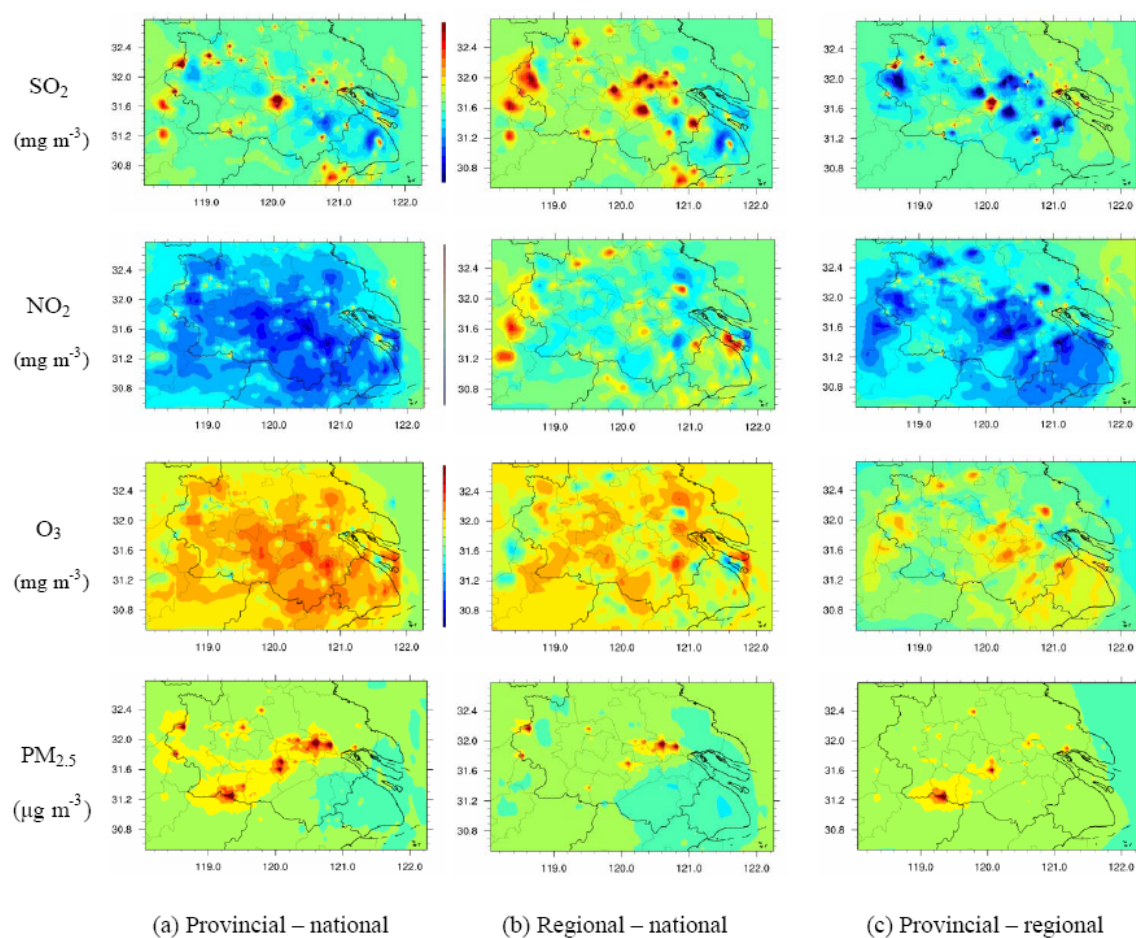
## Development of a high-resolution emission inventory and its evaluation and application through air quality modeling for Jiangsu China

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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<sub>2</sub>, NO<sub>x</sub>, CO, NH<sub>3</sub>, volatile organic compounds (VOCs), total suspended particulates (TSP), PM<sub>10</sub>, PM<sub>2.5</sub>, black carbon (BC), organic carbon (OC), and CO<sub>2</sub>. 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<sub>x</sub> emissions in our provincial inventory was more consistent with summer tropospheric NO<sub>2</sub> 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<sub>2</sub>, NO<sub>2</sub>, O<sub>3</sub> and PM<sub>2.5</sub>. 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<sub>2</sub> 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<sub>2.5</sub> and O<sub>3</sub> 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<sub>2.5</sub> concentrations in Nanjing. The O<sub>3</sub> formation was VOCs-limited in southern Jiangsu, and the concentrations were negatively correlated with NO<sub>x</sub> emissions in urban areas owing to the accumulated NO<sub>x</sub> from transportation. The sensitivity analysis also indicated that regional transport might contribute significantly to suburban O<sub>3</sub> 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 1** The differences in the monthly means of simulated  $\text{SO}_2$ ,  $\text{NO}_2$ ,  $\text{PM}_{2.5}$ , and  $\text{O}_3$  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  $\text{SO}_2$  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 (O<sub>3</sub>) 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 PM<sub>2.5</sub> by reducing SO<sub>2</sub> and industrial smoke and dust emissions. Recent monitoring data shows that declining trends in PM<sub>2.5</sub> in China, reflecting the effectiveness of PM<sub>2.5</sub> 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 O<sub>3</sub>, particles scatter and absorb the sunlight, reduce the UV radiation and consequently suppress O<sub>3</sub> 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 O<sub>3</sub> formation.

To investigate the impact of PM and SO<sub>2</sub> emission controls in China on the ground level O<sub>3</sub> formation, we used Community Multi-scale Air Quality Model (CMAQv5.0.1) to simulated O<sub>3</sub> concentrations under different PM and SO<sub>2</sub> 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 O<sub>3</sub> is predicted to increase with the reduction in the SO<sub>2</sub> and PM emissions in the NCP area and its downwind ocean areas. The maximum 1h peak O<sub>3</sub> concentrations in the three months are over 10 ppbv in the center and east China. The largest increase of 45 ppbv in 1h O<sub>3</sub> concentration is over the east China sea, downwind of Shanghai and Jiangsu province. Moreover, the PM and SO<sub>2</sub> controls also may greatly increase the 1h O<sub>3</sub> concentrations (~15-25 ppbv) in Korea and Japan, even though the seasonal average increase of 8h maximum O<sub>3</sub> 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 ( $O_{3-INTFLOW}$ ) decreased, whereas ozone contributions from other source regions change little during this period.

Keywords: urbanization, black carbon, ozone chemistry

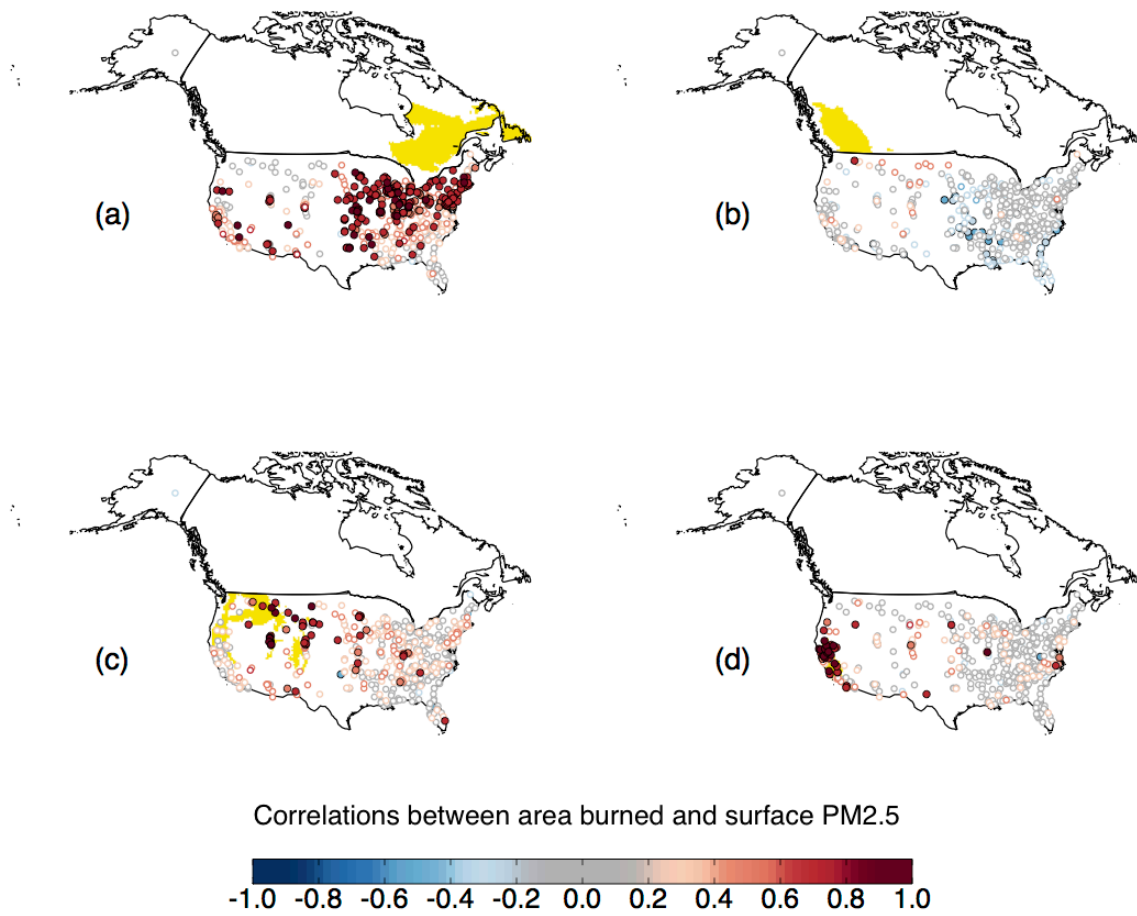
## Increasing wildfire emissions worsen air quality of U.S. megacities by the 2050s

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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 PM<sub>2.5</sub> concentrations at 1775 U.S. sites. Composite analyses show distinct responses of surface PM<sub>2.5</sub> concentrations to wildfire episodes (area burned at >90<sup>th</sup> 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 PM<sub>2.5</sub> concentrations at U.S. megacities with at least half million population. Simulations show that present-day wildfires enhance summer mean PM<sub>2.5</sub> concentrations by  $> 0.5 \mu\text{g m}^{-3}$  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 PM<sub>2.5</sub> enhancement is as high as  $1\text{-}3 \mu\text{g m}^{-3}$  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 importance of the photochemical pathways that transform  $\text{NO}_x$  to  $\text{HNO}_3$  and (2) the relative contributions of local vs. long-distance  $\text{NO}_x$  emissions to dry vs. wet deposition in various environmental settings. To address these uncertainties we determined the  $\Delta^{17}\text{O}$  values of wet and dry deposited  $\text{HNO}_3$  in 2009 at two sites along the western coast of northern Japan, downwind of the East Asian continent where  $\text{NO}_x$  emissions have increased approximately four-fold during the past forty years. At a remote site, nitrate  $\Delta^{17}\text{O}$  values in wet and dry deposition showed similar seasonal variation, ranging between ~23 and 30‰. These results suggest that both forms of deposition experienced similar photochemical reactions during their formation, with  $\text{O}_3$  as the dominant oxidant in winter and a combination of  $\text{O}_3$  and OH in summer. In contrast, at an urban site, nitrate  $\Delta^{17}\text{O}$  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  $\text{NO}_x$  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<sub>2.5</sub> 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<sub>2.5</sub> 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<sub>2.5</sub> with other criteria pollutants and meteorological parameters were studied. PM<sub>2.5</sub> showed a stronger co-relation with NO<sub>2</sub> in winter and SO<sub>2</sub> in monsoon. PM<sub>2.5</sub> 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<sub>2.5</sub> in Delhi, Lucknow and Bangalore.

Keywords: India, long range transport, back trajectory, PM<sub>2.5</sub>

## A study of local circulation and their effect on air quality: a case study over Veracruz city, Mexico

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Veracruz city Harbor is the most important in Mexico. Recently an expansion project has result land cover changes, a population and vehicular fleet increase and major increment on emission rates. Therefore the meteorological condition information that favor or not the dispersion of pollutants is crucial to decision making. There are few studies about the pollution dispersion in this region; however the role of local circulations, the mixing height variability and particle trajectories emitted have not been examined yet. In this work, meteorological surface stations, buoys and the North America Regional Reanalysis data were used to characterize daily and seasonal variability of winds, temperature humidity. The CALMET model was also used to assess meteorological conditions for events with medium to high PM<sub>10</sub> concentrations. The results show that a strong katabatic wind originates from radiational cooling of air atop the Central Mexican Plateau reaches the coast during the night transporting particles to Xalapa and Veracruz cities area. Also during daylight the sea breeze carries particles inland and at night the emissions are trapped in a shallow boundary layer near the coastline. Additionally PM<sub>10</sub> concentration maxima occur during cold surges events due to wind erosion.

Keywords: dispersion of pollutants, local circulation, CALMET model

## Regional contributions to primary and secondary inorganic components of particulate matter in India

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Source-oriented versions of the Community Multiscale Air Quality (CMAQ) model were used to estimate the contributions of different sources and regions to primary and secondary inorganic components (including elemental carbon, organic carbon, sulfate, nitrate, and ammonia) of particulate matter (PM) in India using Emission Database for Global Atmospheric Research (EDGAR) and Weather Research & Forecasting (WRF) model for meteorological inputs. The whole year 2015 was simulated with emissions grouped to seven sectors and nine regions. Seasonal variations in contributions of different sources and regions to major cities in Delhi were analyzed. Results indicate that while residential burning was the dominant source of PM during winter, open burning dominated during pre-monsoon. The source-region analysis indicates that Haryana-Punjab, Rajasthan, Haryana-Punjab and Uttar Pradesh regions were contributing to major fractions of primary and secondary inorganic PM in Delhi during winter, pre-monsoon and post-monsoon seasons.

Keywords: Particulate matter, Source apportionment, Regional transport, India

## Regional transport of ozone and its precursors to Southeast Louisiana

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The Community Multi-scale Air Quality (CMAQ) model with modified photochemical mechanism is used to investigate the contributions of regional transport to ozone (O<sub>3</sub>) and its precursors to Southeast Louisiana in summer months from 2006 to 2015. Contributions from eight different source sectors and regions to the 8 hour average daytime O<sub>3</sub> concentrations will be determined. The source types including residential wood combustion, on-road transportation, oil and gas, off-road, electric generating utilities (EGU), open burning, industry and other sources. After the local sources are quantified, transport of upwind sources is determined. Contributions of different source regions to direct O<sub>3</sub> concentrations or its precursors will be obtained. Under favorable transport conditions, the maximum contribution to 1 hour O<sub>3</sub> from each region will also be evaluated. Changes of the contributions of regional transport by comparing different years will show the effectiveness of previous control measures. The results would provide valuable information on controlling local and regional emissions of O<sub>3</sub> precursors for improve O<sub>3</sub> air quality in Southeast Louisiana.

Keywords: Ozone, Regional transport, Photochemical mechanism, CMAQ, Southeast Louisiana



## A modeling study of effective radiative forcing and climate response due to increased methane concentration

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An atmospheric general circulation model BCC\_AGCM2.0 and observation data from ARIS were used to calculate the effective radiative forcing (ERF) due to increased methane concentration since pre-industrial times and its impacts on climate. The ERF of methane from 1750 to 2011 was  $0.46 \text{ W m}^{-2}$  by taking it as a well-mixed greenhouse gas, and the inhomogeneity of methane increased its ERF by about  $0.02 \text{ W m}^{-2}$ . The change of methane concentration since pre-industrial led to an increase of  $0.31 \text{ }^{\circ}\text{C}$  in global mean surface air temperature and  $0.02 \text{ mm d}^{-1}$  in global mean precipitation. The warming was prominent over the middle and high latitudes of the Northern Hemisphere (with a maximum increase exceeding  $1.4 \text{ }^{\circ}\text{C}$ ). The precipitation notably increased (maximum increase of  $1.8 \text{ mm d}^{-1}$ ) over the ocean between  $10^{\circ}\text{N}$  and  $20^{\circ}\text{N}$  and significantly decreased (maximum decrease  $>-0.6 \text{ mm d}^{-1}$ ) between  $10^{\circ}\text{S}$  and  $10^{\circ}\text{N}$ . These changes caused a northward movement of precipitation cell in the Intertropical Convergence Zone (ITCZ). Cloud cover significantly increased (by approximately 4%) in the high latitudes in both hemispheres, and sharply decreased (by approximately 3%) in tropical areas.

Keywords: Methane, Effective radiative forcing, Climate change

## Observationally constrained simulation of aerosol optical properties over East Asia

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Aerosol chemical composition over East Asia, a mixture of natural dust, sea salt, biomass burning, and air pollutants, is more diverse than other regions. There are still large uncertainties to simulate the aerosol composition and its associated optical properties in aerosol models. The aerosol optical properties simulated by a flexible high resolution global to regional air quality model (NICAM-Chem) are evaluated using the space-based and ground-based observations. The aerosol processes over East Asia including emission, transport, and deposition are compared between multiple aerosol models, and the general similarities and differences are found. Based on Local Ensemble Transform Kalman filter (LETKF) method, the aerosol assimilation system for the NICAM-Chem is further developed to improve the model performances. Assimilation leads to significantly positive effect on the simulated AOD field, improving agreement with all of the 12 AERONET sites over the Eastern Asia based on both the correlation coefficient and the root mean square difference (assimilation efficiency). Meanwhile, better agreement of the Ångström Exponent (AE) field is achieved for 8 of the 12 sites due to the assimilation of AOD only.

Keywords: Aerosol optical properties, LETKF, Aerosol assimilation

## Correlation patterns and seasonal scaling behaviors of PM<sub>2.5</sub> concentration in China

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Recently China has been suffering from air pollution. Aerosols or particulate matters are an important component of the atmosphere, transporting under complex meteorological conditions. Here the data of PM<sub>2.5</sub> observations provided by the ministry of environmental protection, is first studied by a complex networks approach. We calculate the cross-correlation function for different seasons. The seasonal scaling behaviour of the probability distribution function of correlation can be observed. We report the two types of correlations, which correspond to the local and long-range interactions respectively. The local interaction is mainly caused by free expansion or transmission by wind. And a whole picture about the direction of transmission of PM<sub>2.5</sub> is given in China for different seasons. The long-range interaction is correlated with atmospheric waves.

## A review on the local and inter-regional contributions to primary and secondary PM<sub>2.5</sub> pollution in key regions of China

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Rapid economic growth and urbanization in China lead to increased primary air pollutants emissions and secondary particulate matter formation from power generation, industries, transportation as well as residential sectors. Primary and Secondary PM<sub>2.5</sub> can be formed by local emissions and also can be transported over longer distances. Understanding the contributions of local and regional transport contributions to primary and secondary PM<sub>2.5</sub> in key regions of China is necessary for designing effective emission control programs to reduce PM<sub>2.5</sub> pollution in these regions. In this study, we reviewed the studies on the local and regional transport contributions of PM<sub>2.5</sub> in China published in literature based on various methods including ambient measurements, trajectory analysis, and air quality modeling, etc. Contributions of different source regions to primary and secondary PM<sub>2.5</sub> will be summarized quantitatively for the key regions under representative pollution episodes. The meteorological conditions that affect the formation, transport and gas-to-particle partitioning of PM<sub>2.5</sub> will be analyzed.

Keywords: PM<sub>2.5</sub>, local emission, regional transport

# A Modeling Study of Emission Control Strategies in Urban Cities in the Yangtze River Delta, China

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With fast advances in economy, most eastern Chinese cities are experiencing severe air pollution and in an urgent demand of stringent emission control strategies. The Community Multi-scale Air Quality model (CMAQ) and the Weather Research & Forecasting model (WRF) were applied to study the air quality and emission control strategies in two urban cities, i.e. Shanghai and Nanjing in the Yangtze River Delta (YRD), China. Multi-resolution Emission Inventory for China (MEIC) and the Model of Emissions of Gases and Aerosols from Nature (MEGAN) were used for anthropogenic and biogenic emissions, respectively. We evaluated model's performances against seasonal observations of  $O_3$ ,  $NO_x$ ,  $SO_2$ ,  $PM_{2.5}$  and  $PM_{10}$  at 10 monitoring sites in Shanghai and 11 monitoring sites in Nanjing during 2015. We further compared detailed  $PM_{2.5}$  composition from the model and measured data at an urban monitoring site as an additional constraint. The model can well reproduce the spatial and temporal distribution of these chemical compounds. We then designed emission control strategies for  $PM_{2.5}$  in Shanghai and Nanjing based on the modeling results. Sensitivity tests showed that long-range transport is mainly responsible for  $PM_{2.5}$  pollution in both cities. Therefore, a collaborative emission control strategy in Nanjing/Shanghai and their surrounding regions is needed to effectively improve air quality. We also performed several sensitivity tests to study the response of  $PM_{2.5}$  to different total controlled emission reductions as well as major primary emitted  $PM_{2.5}$  precursors. This information is very useful for the government in policy making in the future.

Keywords: Emission control, CMAQ, WRF, China

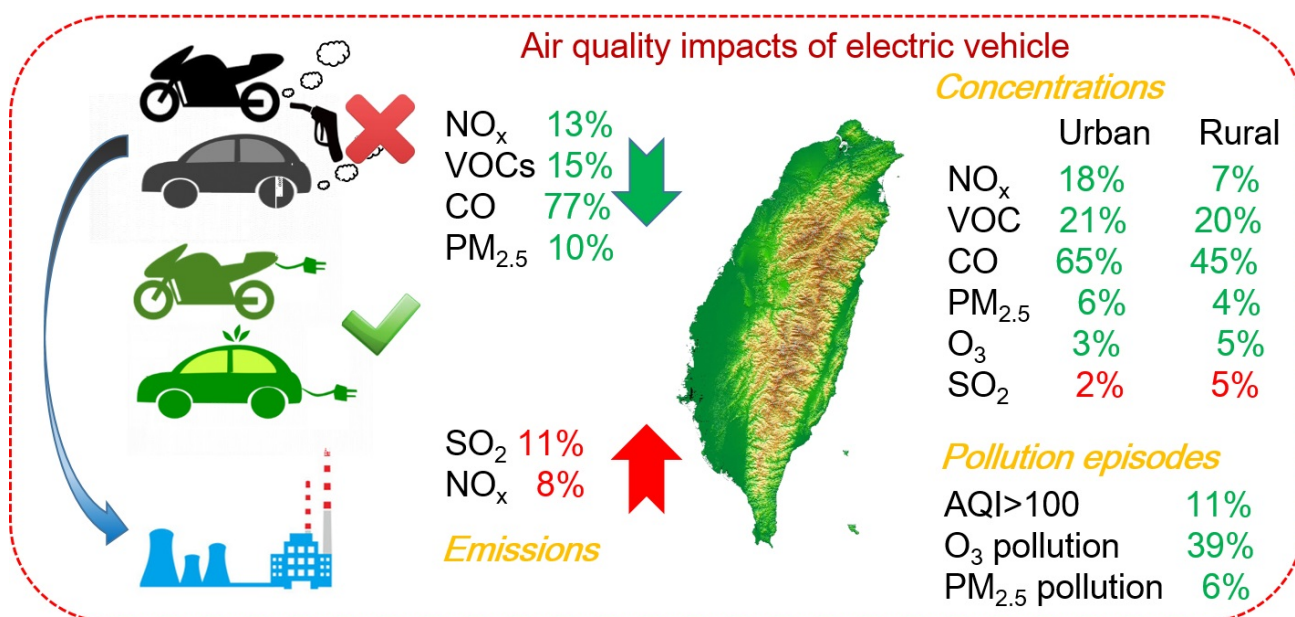
## Potential impacts of electric vehicles on air quality in Taiwan

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The prospective impacts of electric vehicle (EV) penetration on the air quality in Taiwan were evaluated using an air quality model with the assumption of an ambitious replacement of current light-duty vehicles under different power generation scenarios. With full EV penetration (i.e., the replacement of all light-duty vehicles), CO, VOCs, NO<sub>x</sub> and PM<sub>2.5</sub> emissions in Taiwan from a fleet of 20.6 million vehicles would be reduced by 1500, 165, 33.9 and 7.2 Gg yr<sup>-1</sup>, respectively, while electric sector NO<sub>x</sub> and SO<sub>2</sub> emissions would be increased by up to 20.3 and 12.9 Gg yr<sup>-1</sup>, respectively, if the electricity to power EVs were provided by thermal power plants. The net impacts of these emission changes would be to reduce the annual mean surface concentrations of CO, VOCs, NO<sub>x</sub> and PM<sub>2.5</sub> by about 260, 11.3, 3.3 ppb and 2.1 μg m<sup>-3</sup>, respectively, but to increase SO<sub>2</sub> by 0.1 ppb. Larger reductions tend to occur at time and place of higher ambient concentrations and during high pollution events. Greater benefits would clearly be attained if clean energy sources were fully encouraged. EV penetration would also reduce the mean peak-time surface O<sub>3</sub> concentrations by up to 7 ppb across Taiwan with the exception of the center of metropolitan Taipei where the concentration increased by ~2 ppb. Furthermore, full EV penetration would reduce annual days of O<sub>3</sub> pollution episodes by ~40% and PM<sub>2.5</sub> pollution episodes by 6–10%. Our findings offer important insights into the air quality impacts of EV and can provide useful information for potential mitigation actions.

Keywords: Electric vehicle, air quality, AQI, CMAQ, Taiwan



## Investigating the feature and regional sources of urban PM<sub>2.5</sub> concentration over Central China in 2014

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In Central China, where Wuhan is a typical megacity, rapid economic growth has created numerous cities-clusters in recent years, which undoubtedly aggravates the regional and urban haze pollution in China attracting worldwide attention. In sight of recent works on observed particulate matter pollution, the feature and potential source regions of PM<sub>2.5</sub> concentration over Wuhan in the whole year remain highly uncertain. Based on analysis of observed data, the hazy days (average daily PM<sub>2.5</sub>  $75 \mu\text{g m}^{-3}$ ) in Wuhan accounted for 48% in 2014, and the annual concentration of PM<sub>2.5</sub> ( $84.1 \mu\text{g m}^{-3}$ ) kept the same level with that of Beijing in 2013. Especially, PM<sub>2.5</sub> value in January was twice of that in Beijing over the corresponding period. Air pollution was severest in winter over Wuhan with hazy days of 18-30days in different month, followed by spring and autumn. Though the air quality was the best in summer, the days of PM<sub>2.5</sub> concentration averagely exceeding  $75 \mu\text{g m}^{-3}$  ranged from 3 to 17days. We analyzed the impact of regional chemical transport of air pollutants throughout the year using a Nested Air Quality Prediction Model System (NAQPMS) with a source tagged tracer method. The monthly local contribution of Wuhan in winter is the smallest (less than 50%), which indicates regional transport is the dominant source of high PM<sub>2.5</sub> level. The local emissions play a determinant role in PM<sub>2.5</sub> formation in summer, while regional contribution could be 30%-40% approximately. The main reason is that prevailing strong winds in favor of regional transport from high-emission areas in winter, and strong local specificities and effective diffusion processes in vertical that decreasing PM<sub>2.5</sub> concentration in the local. Our results highlight the importance of the air pollutant transports in the formation of fine particulate matter over Wuhan. On a long term and durable perspective, regulating the regional trans-boundary environmental impact assessment in China appears to be an imperative for effectively mitigating urban PM<sub>2.5</sub> loading.

Keywords: PM2.5, Central China, regional transport, haze pollution, Wuhan

## Investigation of relationship between air pollution formation mechanism and synoptic pattern based on three-years observations in megacity Beijing, China

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Air pollution is a severe problem in China especially in winter season for the decade years. To investigate the formation mechanisms of air pollution in winter, the relationship between air pollution and synoptic pattern was explored by employed three years (2013-2015) high resolution PM<sub>2.5</sub> concentrations, synoptic charts, satellite images, radiosonde data, wind pattern observation, and HYSPLIT Trajectory Model in megacity Beijing, China. The results indicated that haze days (daily PM<sub>2.5</sub> concentration > 150  $\mu\text{g}/\text{m}^3$ ) in winter season Beijing is 36, 28 and 35 for 2013, 2014 and 2015, respectively. Consistent air pollution episodes always accompanied with the following synoptic patterns: 1) at 500 hPa, cold air forces were located in the north part and north china Plain was controlled by western wind; 2) at 850 hPa, warm advection frequently occurred above North China plain and Bohai bay, which favored stable synoptic pattern and transportations of air pollutants to Beijing; 3) On surface, Beijing was controlled by back of anticyclone, low pressure or uniform pressure situation, which accounted for 47.3%, 18.2% and 34.5% of the serious haze episodes, respectively. The above results also illustrated that air pollution episodes accompanied with anticyclone arose most frequently with maximum daily PM<sub>2.5</sub> concentration 258.8  $\mu\text{g}/\text{m}^3$ . The results will offer beneficial environmental implications for the air pollution forecasting.

Keywords: air pollution, synoptic pattern, anticyclone, formation mechanisms



## Outstanding seasonality of the lower tropospheric ozone over central China observed by Ozone Monitoring Instrument (OMI)

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Recent study by Cooper et al. (2016) reviewed global ozone ( $O_3$ ) measurements, and showed a significant positive trend of the surface  $O_3$  over East Asia after 1990's. However, as the number of ground-based stations in East Asia is limited, a whole picture of the spatial distribution and its interannual variability of the boundary layer  $O_3$  over East Asia is not yet well captured by observations. On the other hand, recent technological advances have made it possible to observe atmospheric concentrations of  $O_3$  from space. However, almost 90 % of  $O_3$  is available in the stratosphere while the amount of  $O_3$  in the boundary layer is usually only several percentage of the total amount. Therefore, the vertical discrimination of  $O_3$  in the lower troposphere is a big challenge in satellite-borne measurements. In spite of the difficulty, substantial progress has been made on this problem. Liu et al. (2010) successfully derived the ozone profiles from the surface up to 60 km into 24 layers using the ultraviolet spectra observed by Ozone Monitoring Instrument (OMI). The lowermost layer corresponds to a layer from 0 km to about 2.5~3 km above the surface. Hayashida et al. (2015) examined the 24<sup>th</sup> layer of their products and assured the reliability of the  $O_3$  in the lower troposphere under enhanced  $O_3$  conditions. They reported  $O_3$  enhancement observed in Central and Eastern China (CEC), with Shandong as its center, and most notable in June in any given year. In this study, to reveal spatial and temporal variation of ozone distribution over CEC, we applied cluster analysis to the OMI  $O_3$  data over the regions. We focus the anomaly of ozone ( $DO_3$ ), which is defined as the difference from the a priori values ( $DO_3 = O_3[\text{retrieval}] - O_3[\text{a priori}]$ ). This analysis is effective to follow  $O_3$  enhancement under polluted condition, because our focus is the temporal  $O_3$  enhancement from the background level, i.e. climatological values. The  $DO_3$  values can be interpreted as an indicator of the ozone enhancement from the background level.

Before cluster analysis, we applied the screening as described in Hayashida et al. (2017) for all OMI retrievals during the period from October 2004 through December 2013 to remove any doubtful data that might be affected by the UT/LS ozone variability.

We divided all of the grids in the range of 25°- 40°N, and 100°-135°E into some clusters according to the similarity of the seasonal variation of  $DO_3$  at the 24<sup>th</sup> layer. The function used for the analysis is based on the complete linkage method for hierarchical clustering implemented in the statistical tool R (R Core Team, 2012). The number of the cluster was given from 4 to 11. By this analysis, we can distinguish the areas where  $DO_3$  has outstanding seasonality over the North China Plain and Sichuan basin (named as Cluster 1). The Cluster 1 corresponds to the areas of high  $NO_2$  concentration observed by satellite sensors. The values of  $DO_3$  as well as  $O_3$  in Cluster 1 show high in summer (in June in particular) and low in winter. We compared those clustered areas with the model simulations by Meteorological Research Institute -Chemistry Climate Model (MRI-CCM2) (Deushi and Shibata, 2011). The Cluster 1 corresponds to the areas of high chemical production rate in June in the model simulation. We also compared the results of cluster analysis with meteorological data. Along the coastal area,  $DO_3$  tends to drop to negative values (less than climatology) temporarily in August, which can be interpreted as the inflow of oceanic clean air into the inland area.

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Keywords: ozone, China, OMI

# High Resolution vehicular emissions inventory in Shanghai China: Application of REMI model

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Accelerated industrial development of Shanghai, China, lead to a large number of vehicles into the streets, generating critical levels of pollution that remains high nowadays. This is the biggest city in China, in commercial and industrial aspects, with more than 20 millions urban habitants, which produced an intense demand of public and private transport. Air pollutant concentrations persistently remains high with risk to health of population (<http://semc.gov.cn/aqi/home/English.aspx>, Environmental Service of Shanghai). This study presents a bottom-up vehicular emissions inventory with the R Emission Inventory (REMI) Package (Ibarra *et al*, 2017a) for the mega city of Shanghai. REMI is package wrote in R language that estimates vehicular emissions inventory considering exhaust, cold-start, evaporative, dust resuspension and wear emissions. The road network (Figure 1) of Open Street Map is used as input, identifying the type of street, to perform a spatial traffic interpolation as shown by Ibarra *et al* (2017b). This approach assumes high density of light duty vehicles in downtown, and in contrast, less density of trucks in downtown. The emission factors used are COmputer Programme to calculate Emissions from Road Transport (COPERT) with an euro equivalency as shown by Wang *et al* (2010). Nevertheless, REMI offer the option to use local emission factors or a merge between local and COPERT emission factors. The age distribution is very important, so it was assumed that all vehicles were in circulation till 40 years of use. REMI is also suitable for cities with limitation of data, as showed by Ibarra *et al* (2017b) because it interpolates traffic and assign it into the road network directly. REMI outputs consists in vehicular emission with high spatial and temporal resolution, with hourly emissions at street level. This study will consider a detailed perspective of vehicles, including the use of motorcycles and it will be compared with Wang *et al* (2008). The resulting estimation will give detailed pollutant for each road, hour of the day and day of the week, allowing investigation of vehicular emissions for the biggest city of China.

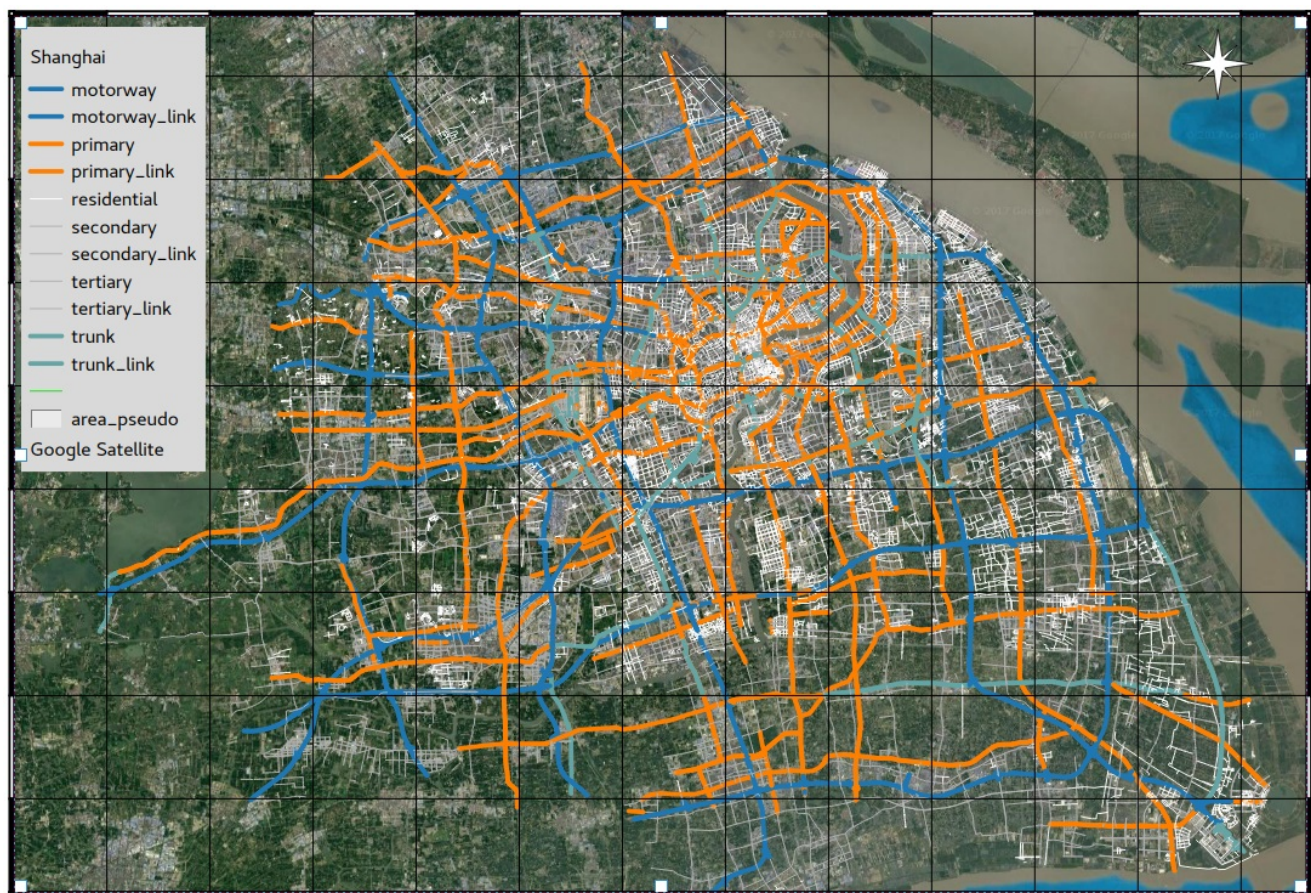
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Keywords: REMI, Emissions Inventory, Air Pollution



## Emissions of fine particulate nitrated phenols from the burning of five common types of biomass

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Nitrated phenols are among the major constituents of brown carbon and affect both climates and ecosystems. However, emissions from biomass burning, which comprise one of the most important primary sources of atmospheric nitrated phenols, are not well understood. In this study, the concentrations and proportions of 10 nitrated phenols, including nitrophenols, nitrocatechols, nitrosalicylic acids, and dinitrophenol, in fine particles from biomass smoke were determined under three different burning conditions (flaming, weakly flaming, and smoldering) with five common types of biomass (leaves, branches, corncob, corn stalk, and wheat straw). The total abundances of fine nitrated phenols produced by biomass burning ranged from 2.02 to 99.52  $\mu\text{g m}^{-3}$ . The compositions of nitrated phenols varied with biomass types and burning conditions. 4-nitrocatechol and methyl nitrocatechols were generally most abundant, accounting for up to 88–95% of total nitrated phenols in flaming burning condition. The emission ratios of nitrated phenols to  $\text{PM}_{2.5}$  increased with the completeness of combustion and ranged from 7 to 45  $\text{ng mg}^{-1}$  and from 239 to 1081  $\text{ng mg}^{-1}$  for smoldering and flaming burning, respectively. The ratios of fine nitrated phenols to organic matter in biomass burning aerosols were comparable to or lower than those in ambient aerosols affected by biomass burning, indicating that secondary formation contributed significantly to ambient levels of fine nitrated phenols. The emission factors of fine nitrated phenols from flaming biomass burning were approximately 0.75–11.07  $\text{mg kg}^{-1}$ . According to calculations based on corn and wheat production in 31 Chinese provinces in 2013, the total estimated emission of fine nitrated phenols from the burning of corncobs, corn stalks, and wheat straw was 670 t. This work highlights the apparent emission of methyl nitrocatechols from biomass burning and provides basic data for modeling studies.

Keywords: Nitrated phenols, emission, biomass burning, fine particulate matter, smoke