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Key Points:

- PM source apportionment in Shanghai is simulated using a regional model
- Regional transport plays an important role in haze formation in Shanghai
- Controlling emission from vehicle and industrial process is vital

Supporting Information:

- Text S1
- Table S1
- Table S2
- Figure S1
- Figure S2
- Figure S3
- Figure S4
- Readme

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Source apportionment of fine particulate matter during autumn haze episodes in Shanghai, China

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Abstract Understanding the origin of fine particulate matter is essential to proposing proper strategies for heavy haze mitigation in Shanghai, China. In this study we used the Particulate Matter Source Apportionment Technology in Comprehensive Air Quality Model with Extensions to quantify the impacts of emissions on the concentrations of fine particulate matter ($PM_{2.5}$) and its important components in Shanghai during heavy haze episodes in late autumn (6–22 November 2010). The factors considered here are regions of Shanghai and its surrounding areas, long-range regional transport, and different local emission categories. The results indicate that industrial process is the dominant local contributor to total $PM_{2.5}$ mass in the whole city except that at the urban center vehicle emission contributes slightly more. In addition, industrial process and vehicle emission are the major local contributors for nitrate in Shanghai, although at urban core the contribution from vehicle emission is remarkably larger. Generally, both local contribution and regional transport contribution could dominate a severe haze event in late autumn. However, the dominant contributor could either be local emission or regional transport, usually depending on the meteorological conditions. Therefore, particular attentions should be paid to the emission control in the upwind adjacent provinces, as well as in local areas, for developing effective strategies to reduce $PM_{2.5}$ pollution in Shanghai.

1. Introduction

Air quality has noticeably deteriorated due to the dramatic increase of anthropogenic pollutants emission with rapid economic development and urbanization in China in the recent decades [*Zhang et al.*, 2009; *Huang et al.*, 2011; *Saikawa et al.*, 2011; *Yang et al.*, 2011]. The high particulate matter pollution level in China has raised many concerns over the past decade [*Andreae et al.*, 2008; *Chan and Yao*, 2008; *Chang et al.*, 2009; *Fang et al.*, 2009; *Gao et al.*, 2009; *Feng et al.*, 2006; *Li et al.*, 2007; *Fu et al.*, 2012; *Xu et al.*, 2011; *Huang et al.*, 2012a, 2012d]. Elevated levels of $PM_{2.5}$ (i.e., particulate matter with an aerodynamic diameter less than 2.5 μm) have been linked to negative impacts on human health [*Gurjar et al.*, 2010; *Huang et al.*, 2012c]. In order to improve air quality in China, the Chinese government issued a new ambient air quality standard in March 2012 that will go into effect nationwide by 2016 while major cities including Shanghai must regularly monitor additional pollutants including $PM_{2.5}$ and O_3. For the first time, $PM_{2.5}$ is included in the ambient air quality standard in China. The limits are 75 μg m^{-3} for 24 h average concentration and 35 μg m^{-3} for annual concentration. However, air quality levels of many cities in China do not meet the new standard yet.

Shanghai is the leading city of Yangtze River Delta region (YRD) with a population well over 20 million. Its annual average $PM_{2.5}$ concentrations in the last 5 years reached 40–50 μg m^{-3} which is nearly 3 times the annual standard in the USA. In Shanghai, there are more than 100 haze days annually in the recent 5 years. The high $PM_{2.5}$ episodes in Shanghai are widely recognized as one of its major air pollution issues in recent years [*Fu et al.*, 2008b; *Li et al.*, 2013; *He et al.*, 2012; *Wang et al.*, 2012a]. Developing an emission control strategy is highly desired to improve air quality in Shanghai.

Factors which should be considered when proposing emission regulations in Shanghai include the relative importance of the source origin of the pollutants (e.g., the fractions from local and regional sources) and the understanding of the fractions of the pollutant concentrations in receptors of interest with different emission categories. Understanding the relationship between emission source origins and the ambient PM concentrations is vital to establish effective control strategies. However, developing a completely reliable source-receptor relationship remains a challenging task [*Seinfeld and Pandis*, 2006].

Some studies of long-range transport impacting a receptor region have been conducted using Lagrangian dispersion models [*Ravetta et al*., 2007; *Trickl et al*., 2003; *Forster et al*., 2004; *Owen et al*., 2006; *Prata et al*., 2007], but Lagrangian dispersion models have difficulties in accurately quantifying source contributions due to simplified chemistry mechanisms used or even neglect chemistry. Another type of models, three-dimensional Eulerian chemical transport models (CTMs), has been a potential tool to help address these source-receptor questions since they simulate all the necessary physical and chemical processes that impact air pollution levels in the domain.

Several approaches based on three-dimensional Eulerian CTMs have been developed for source apportionments. The most simple is the Brute force method (BFM) which is to run a model, repeat it with perturbed emissions, and compare the two simulation results. Unfortunately, the sum of all source contributions will not equal the simulated concentrations in the base case whenever the model response is nonlinear [*Koo et al*., 2009]. Compared to the BFM, Decoupled Direct Method (DDM) developed by *Dunker* [1980, 1981] is an efficient and accurate alternative for sensitivity analysis [*Dunker et al*., 2002]. Some studies have used DDM to provide sensitivities of air pollutants [*Napelenok et al*., 2007; *Napelenok et al*., 2006; *Cohan et al*., 2005]. This approach focuses specifically on the sensitivities (responses to small or moderate emission changes) rather than the contributions from the source regions or categories. Another approach, Source Oriented External Mixture developed by *Kleeman and Cass* [2001] simulates each tagged species separately through every modeled atmospheric process (physical and chemical). It is potentially accurate but is computationally very demanding [*Koo et al*., 2009].

The PM source apportionment technology (PSAT) [*Wagstrom et al*., 2008] is expected to be high in efficiency and flexibility to apportion primary PM, secondary PM, and gaseous precursors of secondary PM among different source categories and source regions [*ENVIRON*, 2011]. *Wagstrom and Pandis* [2011a, 2011b] applied PSAT to study the contributions from local emission and different long-range transport to the targeted PM components and to quantify the impacts of different regions on fine particulate matter in the Eastern United States. PSAT also performs better at estimating the impact on PM concentrations when removing all the emissions rather than a fraction of the emissions from a source [*Koo et al*., 2009].

As a useful tool for source apportionment, PSAT has been used in several studies about air pollution issues in China. The emission activities influence PM concentration in the Pearl River Delta region has been analyzed using PSAT tool [*Wu et al*., 2013]. *Li et al*. [2013] and *Huang et al*. [2012b] applied PSAT tool to investigate the contribution to SO_2 (sulfur dioxide) from emission sources in Tangshan and Beijing, Northern China, respectively. However, a study about source contribution using PSAT tool has never been conducted so far in Shanghai and YRD. Although there are a few studies on PM modeling using three-dimensional Eulerian CTMs in Shanghai [*Li et al*., 2011; *Wang et al*., 2012b], the identification of source regions or source categories of PM has not been considered in their studies. The source-receptor relationship and the contributions of emission categories and regions to ambient fine particular matter concentration in Shanghai are not well understood.

In this study, Comprehensive Air Quality Model with Extensions (CAMx) with PSAT tool was applied to study the contributions of different emission regions including Shanghai, its surrounding areas (i.e., Zhejiang and Jiangsu Provinces within the finest modeling domain), and the long-range (more than about 150 km away) regional transportation to the fine particular matter concentrations during late autumn severe haze episodes in Shanghai, China. The contributions of eight local emission categories are assessed quantitatively. These results can provide general information for the decision makers that propose proper strategy to alleviate $PM_{2.5}$ pollution in megacity Shanghai.

2. Methodology

2.1. Model Setup and Inputs

The daily average observed concentration of $PM_{2.5}$ at Shanghai Academy of Environmental Sciences (SAES) in Shanghai during 6–22 November 2010 in late autumn reaches 90.6 $\mu g\ m^{-3}$, which obviously fails to meet the new national ambient air quality standard in China. Due to such heavy haze episodes occurring frequently in late autumn as well as in winter in recent years, understanding the source contributions of high $PM_{2.5}$ concentrations during such heavy haze episodes is crucial to proposing proper strategies for heavy haze mitigation in Shanghai. Therefore, we chose 17 day period (6–22 November 2010) including several heavy haze episodes as a case in this study to carry out the source apportionment simulation of $PM_{2.5}$ using CAMx (version 5.40) with PSAT tool. Four components are used as the representatives of $PM_{2.5}$ species: elemental carbon as a primary nonvolatile PM species; sulfate, nitrate, and ammonium as the secondary PM species. A spin-up period of 5 days (1–5 November) is used to minimize the influence of initial conditions. For the initial and the outermost boundary conditions, low or zero background concentrations are used for all species. The Carbon Bond 05 chemical mechanism (CB05) [*Yarwood et al*., 2005] is used in the CAMx model. The outermost (first) domain with a horizontal grid spacing of 81 km covers the entire area of China, Japan, Korea, parts of India, and Southeast Asia. The second domain with a horizontal grid spacing of 27 km covers eastern China, and the third domain whose horizontal grid spacing is 9 km covers the Yangtze River Delta (YRD) and the Shandong province. The fourth (finest) domain with 3 km horizontal grid resolution covers the entire area of Shanghai and parts of Zhejiang province and Jiangsu province. All the grids have 14 layers vertically extending from the surface to an altitude of about 19 km above the ground, with the first layer thickness of about 40 m. The outer three domains are specified together in a single run with two-way grid nesting, which means that pollutant concentration information propagates into and out of the three grid nests during the model's iterative calculation. The outputs from the third domain with 9 km horizontal grid spacing are used to provide boundary condition for the finest domain with 3 km horizontal grid spacing by one-way nesting. The PSAT tool is only used for the finest domain.

The meteorological fields were created using the fifth generation Pennsylvania State University/National Center for Atmospheric Research Mesoscale Model (MM5, version 3.7) [*Grell et al*., 1995]. The meteorological inputs, including horizontal wind components, temperature, pressure, water vapor, vertical diffusivity, clouds, and rainfall, were all generated by the MM5CAMx program based on the output of MM5 simulation. The nested horizontal domains for MM5 were the same as those for CAMx. The vertical resolution used in MM5 consists of 34 sigma layers up to 50 mbar. The one-way nesting simulations are performed with the following physics options: the Noah land-surface models, the Grell cumulus scheme, and the mixed-phase microphysics. The National Centers for Environmental Protection $1.0 \times 1.0°$ global reanalysis data were used.

The regional East Asian emission inventory is provided by Intercontinental Chemical Transport Experiment - Phase B (INTEX-B) [*Zhang et al*., 2009; *Streets et al*., 2003a, 2003b; *Fu et al*., 2008a]. For biogenic volatile organic carbon (VOC) emissions, this study uses the natural VOC emission inventory of the Global Emissions Inventory Activity 1990 (http://geiacenter.org). The emission inventories for the third and the fourth (finest) domains are updated to the year 2010 based on the emission calculation for YRD in 2007 [*Huang et al*., 2011] and the new energy consumption data in 2010.

The PSAT algorithm was used to track the source contributions from 4 regions and 8 emission categories to both primary and secondary inorganic particulate matters in the finest domain that focused on Shanghai. This algorithm is based on the principle that molecules of the same species react and are transported identically regardless of their source [*Wagstrom et al*., 2008]. PSAT works in parallel with CAMx to allow tracking of source contributions through all the processes represented in the model without interfering with the CTM calculations. One fundamental assumption in PSAT is that each primary and secondary PM species should be apportioned to the primary gas or PM precursor. For example, the secondary portion of particulate sulfate is apportioned to its precursor SO_x among certain source regions/categories, while the primary portion of particulate sulfate is apportioned to the same species among source regions/categories. For some secondary particulate species such as particulate nitrate, the apportion procedure is complex because several more chemical reactions perform between gaseous precursors and the resulting PM. Hence, a single tracer can track one of the primary PM species whereas secondary PM species require several tracers. Apportionment changes during transport are calculated based on fluxes between cells and the known apportionment in each cell. The parallel treatment and lack of disturbance of the original model calculations result in an algorithm that allows the computationally efficient tracking of numerous sources simultaneously. Comparisons of apportionment predicted by PSAT with that predicted by more computationally expensive benchmark methods showed agreement within a few percent for secondary aerosol species [*Wagstrom et al*., 2008].

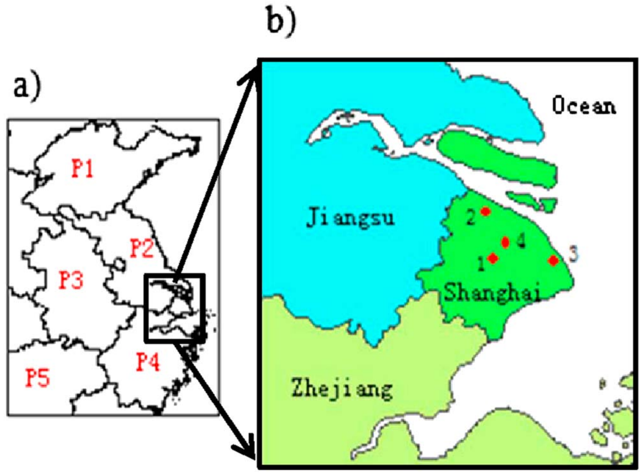

Figure 1. (a) The third and fourth (finest) domains for CAMx simulation. The main provinces in third domain are labeled in red codes: P1-Shandong Province, P2-Jiangsu Province, P3-Anhui Province, P4-Zhejiang Province, and P5-Jiangxi Province; (b) the source regions and receptor sites in the finest domain. Locations of receptors are labeled in numbers: 1-SAES (deputy urban site), 2-Baoshan (industrial site), 3-Nanhui (rural site), and 4-Jing'an (urban center site).

2.2. Source Regions and Emission Categories

Shanghai is situated at the eastern tip of the Yangtze River Delta and halfway along China's eastern coastline; that is, it is adjacent to the East China Sea. Around Shanghai, there are two provinces including Zhejiang and Jiangsu provinces. In the innermost nested domain, four source regions comprised of Jiangsu, Zhejiang, Shanghai, and Ocean were chosen as shown in Figure 1. It should be noted that here Jiangsu and Zhejiang as names of source regions in this study denote only small geographical parts of Jiangsu and Zhejiang provinces, respectively. Eight emission categories are agriculture source, industrial process which is a kind of ground-level point sources, industrial boilers, natural source, boilers in power plants, residential sources, vehicle emission, and volatile sources. The industrial boilers as an emission category in this study represent all industrial boilers except for those in power plants. The volatile sources are comprised of gas station, oil tanks and depots, coating application, garbage disposal and sewage treatment stations, and residential emission of ammonia. In addition, the road dust emission has been incorporated in vehicles emission categories. Eight emission categories over four source regions produce 32 separate source groupings, so that all sources of primary and secondary inorganic particulate matter precursors are accounted, and the corresponding reaction tracers allow these pollutants from multiple source groups to be tracked simultaneously within a single CAMx simulation. The CAMx boundary conditions and initial conditions are always tracked as separate source groupings.

The receptor sites are SAES, Baoshan, Nanhui, and Jing'an as shown in Figure 1. Jing'an site is located in the urban core of Shanghai; SAES site, as one of the Shanghai deputy centers, is located in the southwest of the urban area and close to the Wujing industrial area. Baoshan and Nanhui sites are located in Shanghai industrial area and the rural area, respectively.

3. Results and Discussions

3.1. Predicted $PM_{2.5}$ Concentrations

The average predictions over the entire period of simulation (6–22 November 2010) for total $PM_{2.5}$ mass, sulfate, nitrate, ammonium, elemental carbon, and ozone are depicted in Figure 2. The highest concentrations for most PM species are observed over the densely populated areas including the urban areas of Shanghai, Hangzhou, Suzhou, Jiaxing, Nantong, Huzhou, Wuxi, and Changzhou, while the lowest concentrations of ozone are observed over the same areas, due to the impact of a large amount of nitrogen oxides from the vehicle exhausts and the industrial emissions in urban areas. Figure 2 also shows that one common feature for the total $PM_{2.5}$ mass, sulfate, nitrate, ammonium, and elemental carbon in the entire simulation domain is the general decrease from the west to the east, indicating that there is a notable transport impact for those species from west regions including Zhejiang and Jiangsu provinces to Shanghai during the simulation period. Nevertheless, compared with the other species, transport impact is weak for elemental carbon since it is a primary and stable species.

According to the average concentrations at ground level over the entire domain for the entire period of simulation, nitrate accounts for 17.6% of total $PM_{2.5}$ mass, followed by sulfate (11.6%), ammonium (9.3%), and EC (4.9%). In addition, it should be noted that the average ratio of nitrate to sulfate is 1.5 over the entire simulation domain during the time period. In comparison, the ratio varied between 0.1 and 0.7 with an annual average of 0.4 in Shanghai during 1999 and 2000 [*Yao et al.*, 2002]. During 2006 and 2007, the ratio was

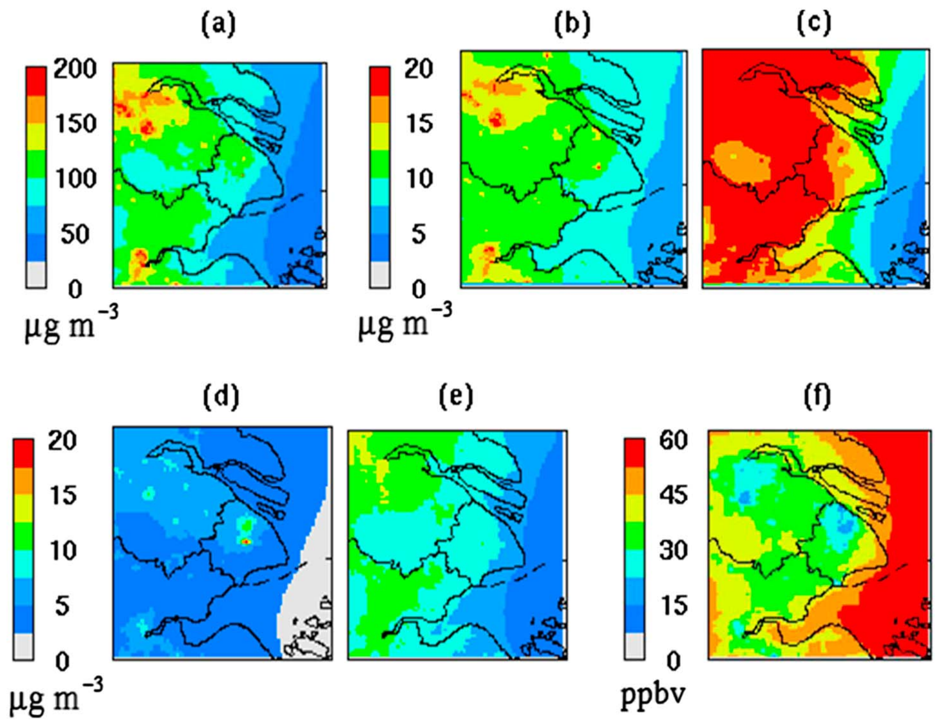

Figure 2. Average predicted ground-level concentrations of (a) $PM_{2.5}$ mass, (b) particulate sulfate, (c) particulate nitrate, (d) particulate elemental carbon, (e) particulate ammonium, and (f) ozone in the innermost nested domain for 6–22 November 2010.

about 0.4 in Lin'an, a rural area close to Shanghai [*Zhang et al.*, 2012]. The main reason for sharp increase of the average nitrate-to-sulfate ratio in recent years is the high emission intensity of NO_x from industrial sources and vehicles whose population increases sharply, followed by the decrease of SO_2 emission due to the increasing application of desulfurization devices in power plants and some other industrial factories during the eleventh 5 year plan period [*SEPB*, 2011]. Figure 2 also shows one particular high PM concentration located in Wujing industrial area that is attributed to high emission intensity of particles from a coking plant. Regarding the spatial distribution in the innermost nested grid as shown in Figure 2, it can be seen that $PM_{2.5}$ mass and sulfate show a similar spatial pattern. In each area with high population density and intense industrial activities, the elevated concentrations are observed for both $PM_{2.5}$ mass and sulfate, but the opposite is the case in rural area. Based on the results of PSAT, the PFN (other Fine Particulate) is on average making up more than 40% of the total $PM_{2.5}$ mass and more than 70% of the remaining $PM_{2.5}$ mass excluding sulfate, nitrate, ammonium, and elemental carbon. PFN species is fairly generic but its source apportionment has also been done using PSAT. The PFN and particulate sulfate have the same biggest local contributor that is industrial process emission based on the results of PSAT. Therefore, particulate sulfate and the total $PM_{2.5}$ mass have a similar spatial distribution as show in Figure 2. Considering the decrease of particulate sulfate and sulfur dioxide emission from vehicles due to great improvement of emission standard, industrial sources probably can be regarded as the dominant contributor to the $PM_{2.5}$ mass and sulfate at ambient environment in areas with densely populated and intense industrial activities.

In Figure 2, we also note that there is such a large contribution coming from the western boundary to the innermost nested grid. To better understand what is related with the high contribution on the boundary, a map with much larger scale is presented in Figure S3 (in the supporting information). A region much larger than the finest grid suffered high pollution level of each pollutant for the simulated period as shown in Figure S3. As mentioned above, there are several densely populated cities surrounding Shanghai and within the innermost nested grid as shown in Figure 2. Moreover, there are some densely populated cities including Nanjing, Ningbo, Yangzhou, Zhenjiang, Shaoxing, Taizhou, and Zhoushan within the third grid but outside the finest grid. These cities and the high densely populated cities within the finest grid as mentioned above are almost contiguous, constituting the Yangtze Delta Megalopolis located in the Yangtze River Delta (YRD) region.

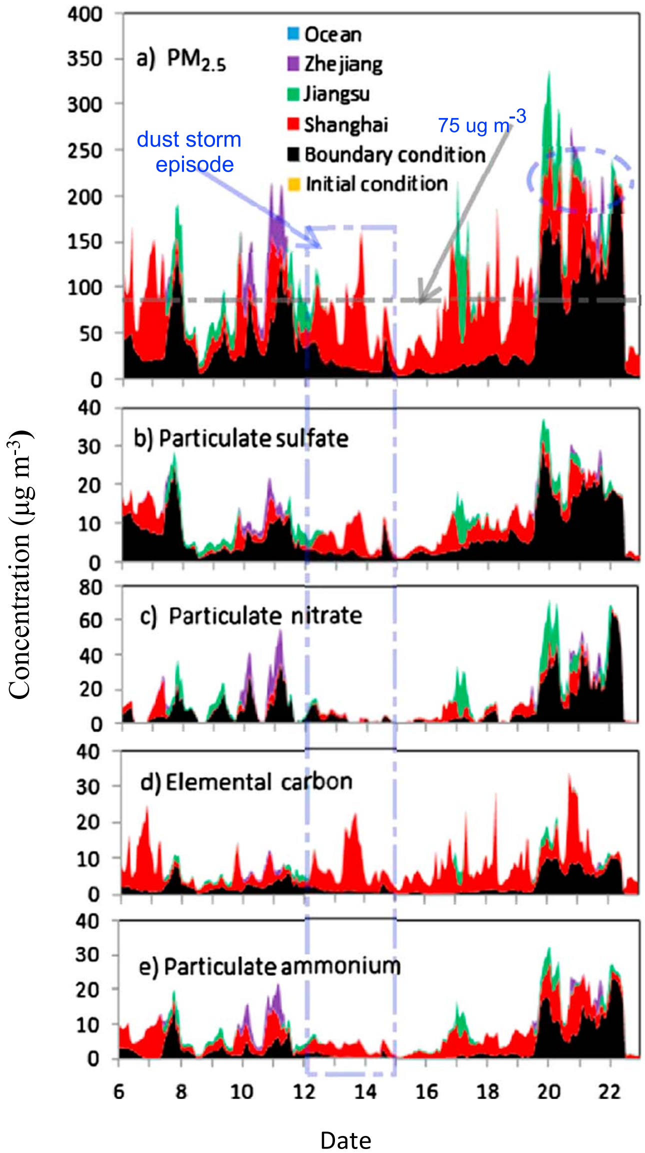

Figure 3. Time series of contributions from different source regions to (a) $PM_{2.5}$ mass, (b) particulate sulfate, (c) particulate nitrate, (d) elemental carbon, and (e) particulate ammonium at the SAES site for 6–22 November 2010 (LT)

Yangtze Delta Megalopolis as one of the biggest city cluster in China has massive industrial activities leading to high emission intensity of air pollutants. Moreover, Shanghai only accounts for 1% of the whole area of Yangtze Delta Megalopolis, and air quality over such small region is very easy to be affected by a large amount of emission in surrounding cities. Therefore, it is not difficult to understand the large contribution of $PM_{2.5}$ and its species coming from boundaries, especially from the western boundary, to the innermost nested grid. Examination of the averaged map of $PM_{2.5}$ and its species helps us to understand that the regional transport plays an important role in severe haze episodes in Shanghai, even in YRD.

3.2. Contributions From Local and Surrounding Areas

3.2.1. Variation With Time

The temporal contributions from different regions to the concentrations of total $PM_{2.5}$ mass, sulfate, nitrate, ammonium, and elemental carbon at the SAES site during 6–22 November 2010 are shown, respectively, in Figure 3. Their concentrations at the SAES site are grouped into the six categories, four of which vary with time, and the concentrations from initial condition and ocean region can be ignored due to their low concentrations. The boundary condition of the finest domain can be roughly taken as the contribution from long-range (more than about 150 km away) transportation. Most circumstances whose daily average concentrations of $PM_{2.5}$ are higher than 75 μg m^{-3} were observed as haze day. The contribution of transport from outside Shanghai is dominant in the concentrations of sulfate and nitrate at almost all times except that it is small for sulfate during dust storm episode of the twelfth to thirteenth. In comparison with nitrate and sulfate, the contributions of transport to ammonium concentration are not so high. It is notable that the $PM_{2.5}$ concentrations are relatively low and cover about only one quarter of PM_{10} concentrations based on observations during dust storm episode. For elemental carbon as a primary and stable species, the contributions from local emission are much more variable with time than those for sulfate, nitrate, and ammonium. Overall, we can conclude that regional and long-range regional transports play an important role in the formation of severe hazes in Shanghai.

Shanghai topography allows polluted air entrainment into Shanghai by westerly wind, while clean air is blown into Shanghai by northeast or southeast wind. High relative humidity is helpful to hygroscopic growth for fine particles. Figure S4 (in the supporting information) shows that northwesterly or southwesterly wind, low wind speed, and high relative humidity occurred simultaneously for some days

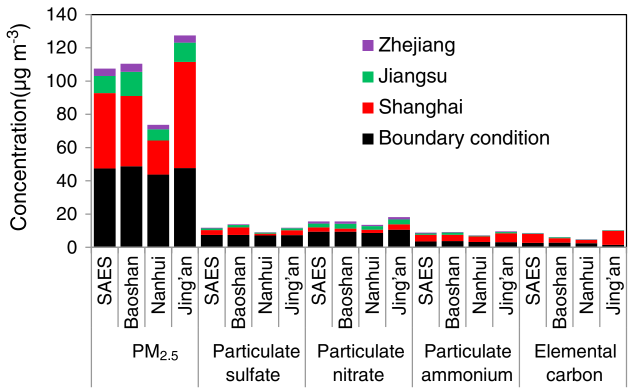

Figure 4. Average concentrations of total $PM_{2.5}$ mass, sulfate, nitrate, ammonium, elemental carbon, and their corresponding contributions from source regions at the SAES (deputy urban), Baoshan (industrial area), Nanhui (rural area), and Jing'an (urban center) sites, respectively, for 6–22 November 2010.

(i.e., 20th to 22th). Under such meteorological conditions, severe hazes are observed with a characteristic of extremely high $PM_{2.5}$ concentrations (Figure 3). Nevertheless, there is almost no contribution from surrounding areas to the total $PM_{2.5}$ and each of the four PM species during the dust storm episode since the air masses blew into Shanghai from ocean.

3.2.2. Variation by Location

Figure 4 shows a notable difference between the urban center site (Jing'an) and the rural site (Nanhui) for the concentrations of total $PM_{2.5}$ mass and its components for the simulated period. The ratios of urban center (Jingan) to rural site (Nanhui) for total $PM_{2.5}$ mass, sulfate, nitrate, ammonium, and elemental carbon are around 1.73, 1.30, 1.34, 1.32, and 2.20, respectively. Compared with these secondary species, the high ratio of elemental carbon reveals that elemental carbon concentrations at urban site and rural site are both greatly affected by local emission. In other words, the transport contribution is low for elemental carbon, compared with sulfate and nitrate. The average concentrations of nitrate are all higher than those for ammonium at these sites. Elemental carbon concentrations at these sites are all lower than those of ammonium, except for those at the urban center site (Jing'an). Figure 4 also shows that the average concentration of total $PM_{2.5}$ mass resulted from local at the urban site is as high as about 3 times of that at the rural site. The local contributions are dominantly responsible for the variation of total $PM_{2.5}$ mass concentration among these sites. Figure 4 also suggests that the emission intensity associated with $PM_{2.5}$ is very high in the whole Shanghai except the rural area.

Average percentage of contribution from each source region to total $PM_{2.5}$ mass and its four species at each of the four sites for the simulation period is respectively shown in Figure 5. It can be seen that average local contribution to the total $PM_{2.5}$ mass is about 50% at the urban center site while it is only 28% at the rural site, suggesting that there is a large amount of anthropogenic emission in the urban area. In Figure 5, it also can be seen that contribution from long-range transport is notable to total $PM_{2.5}$ mass at each of the four sites. In addition, average contribution from Jiangsu to $PM_{2.5}$ at the industrial area (Baoshan) site is much larger than that at any other sites, and contribution from Zhejiang is much smaller to the total $PM_{2.5}$ mass than those from Jiangsu at each of the four sites. It is notable that local contribution dominates the elemental carbon concentration with a percentage of 81% in the urban area, indicating that the emission sources have been concentrated in the urban area for elemental carbon as a stable primary species. In comparison, local contributions to sulfate at these sites are all relatively low; in particular at the rural site, it is only 12%. However, its contribution from long-range transport is dominant in each of these sites. The main reason for those is that the oxidation of SO_2 and the condensation of sulfate both require some time. Most particulate sulfate results from a slow process rather than direct emission of particulate sulfate. As a result, local contributions to sulfate are much smaller than those to the stable primary species such as elemental carbon. On the contrary, the regional

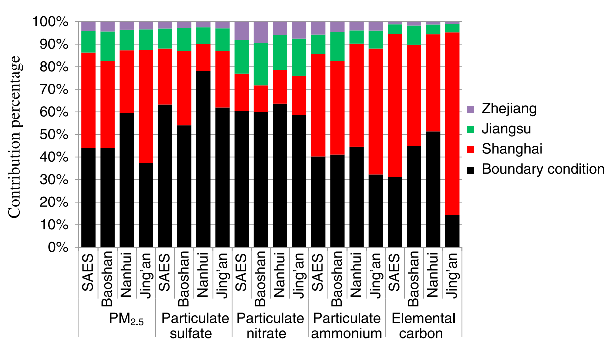

Figure 5. Average percentages of contribution from each source region to total $PM_{2.5}$ mass and sulfate, nitrate, ammonium, and elemental carbon at the SAES (deputy urban), Baoshan (industrial area), Nanhui (rural area), and Jing'an (urban center) sites, respectively, for 6–22 November 2010.

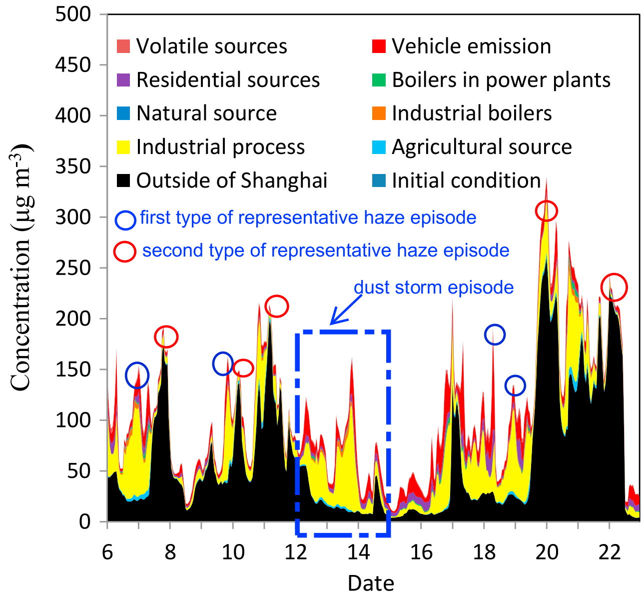

Figure 6. Time series of contributions from local emission categories and outside Shanghai to total $PM_{2.5}$ mass concentration at the SAES site for 6–22 November 2010 (LT).

transport contributions from Zhejiang and Jiangsu to sulfate are slightly higher than those to elemental carbon for the same reason.

As shown in Figure 5, the contributions from surrounding areas and long-range transport to nitrate at all the sites are high (about 23–29% and 59–64%, respectively), suggesting that the regional and super regional transports are the very important sources for high nitrate concentration in Shanghai. Likewise, high emission intensity of precursors of nitrate in Shanghai could obviously contribute to the adjacent downwind provinces. Therefore, emission controlling of the precursors of particulate nitrate in Shanghai is very important, due to its contribution to particulate nitrate in both local Shanghai and the adjacent downwind provinces.

Ammonium is usually associated with nitrate, sulfate, or some other anion and incorporated into the aerosol. Once ammonia releases into the atmosphere, it can return to the surface as either gaseous ammonia or ammonium. Atmospheric ammonia is in dynamic equilibrium with growing vegetation, and the amount of ammonium in $PM_{2.5}$ is quite associated with local ammonia emission. Consequently, local contribution percentages to ammonium are much higher than those to either nitrate or sulfate at each of these sites as shown in Figure 5.

3.3. Contributions From Local Emission Categories

Figure 6 shows time series of contributions from the local emission categories and regional transport to the total $PM_{2.5}$ mass at the SAES site for the studied period. Regional transport contribution represents as the contribution from emission outside Shanghai. By combining Figure 6 and Figure S4 (in the supporting information), it can be found that $PM_{2.5}$ concentration in Shanghai can usually meet the new ambient air quality standard when wind speed exceeds 4 m s^{-1}. However, in the condition of rather weak wind (less than about 0.5 m s^{-1} of wind speed), the $PM_{2.5}$ pollution is usually heavy in late autumn. For example, in the nighttime of 6 November 2010, $PM_{2.5}$ concentration is more than 140 μg m^{-3} and about 80% of it comes from local contribution. Moreover, its biggest local contributor is industrial processes, followed by vehicle emission. The high $PM_{2.5}$ pollution episode in the condition of rather weak wind speed can be regarded as the first type of the representative haze episode. If effective measures of controlling local emission can be taken, the occurrence of such haze episodes will be greatly reduced.

The second type of representative haze episode which can be found by combining Figure 6 and Figure S4 (in the supporting information) is the severe $PM_{2.5}$ pollution which is usually formed when the air masses over Shanghai comes from the upwind adjacent provinces with a moderate wind speed (about 2 m s^{-1}) during late autumn. For example, the night of the seventh, $PM_{2.5}$ concentration is extremely high which is more than 170 μg m^{-3}; moreover, it should be noted that regional transport contribution is dominant, accounting for about 85% of $PM_{2.5}$ mass, suggesting that the regional transport plays a very important role in the formation of such haze episodes. Therefore, effective emission control in upwind adjacent Zhejiang and Jiangsu provinces can greatly reduce the occurrence of the second type of representative haze episodes in Shanghai. In addition, from Figure 6, we also can see that the second type of representative haze episode occurs more frequently than the first type, and the second type is usually severer than the first type during the studied period.

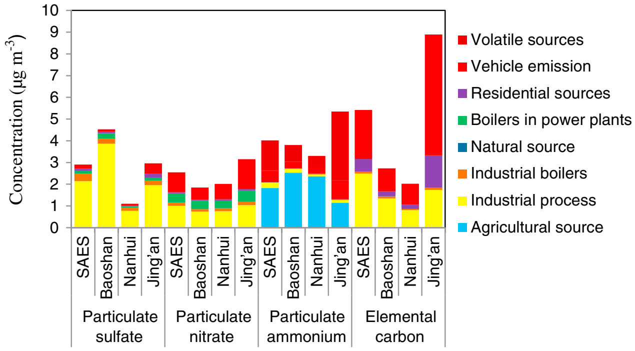

Figure 7. Average mass concentration of sulfate, nitrate, ammonium, and elemental carbon contributed from local emission categories at the SAES (deputy urban), Baoshan (industrial area), Nanhui (rural area), and Jing'an (urban center) sites, respectively, for 6–22 November 2010.

In general, both local emission and regional transport can cause a severe haze event in late autumn. However, the dominant contributor can either be local emission or regional transport, usually depending on the meteorological conditions. Therefore, particular attention should be paid to emission control in the upwind adjacent provinces, as well as in local, in order to alleviate Shanghai ambient $PM_{2.5}$ concentration.

Figure 7 shows the average mass concentrations of sulfate, nitrate, ammonium, and elemental carbon contributed from each local emission category at each of the four sites for 6–22 November 2010. For each of these species, its local contribution represents the total contribution from eight local emission categories. The local contributions to nitrate at these sites are much more uniform compared to sulfate and elemental carbon as shown in Figure 7. For ammonium in the rural area, the contribution from local agriculture source is high and the contribution from local mobile source is low. But the opposite is the case in the urban area. In addition, the contribution to ammonium from vehicle sources and volatile sources (e.g., residential ammonia emission, garbage disposal, and sewage treatment sites) is high at the urban site, due to the high population density. In general, compared to sulfate and elemental carbon, there are relatively small differences of local contributions to ammonium concentration among these sites as shown in Figure 7. It is notable that the local contribution to sulfate at industrial site (Baoshan) is much higher than that at each of the other sites, due to densely concentrated industrial process sources at the industrial site (Baoshan). It is also noted that the local contribution to elemental carbon at urban center site (Jingan) is very high, especially for the contribution from local vehicle emission, due to a large amount of moving vehicles at urban center in Shanghai. The ratio of elemental carbon concentration at the urban center site (Jingan) to the elemental carbon concentration at the rural site (Nanhui), which resulted from local contributions, is nearly 4.4, demonstrating that the elemental carbon emissions are mainly in the urban area.

The average percentages of contribution from each local emission category to total $PM_{2.5}$ mass, sulfate, nitrate, ammonium, and elemental carbon at the each of the four representative sites for

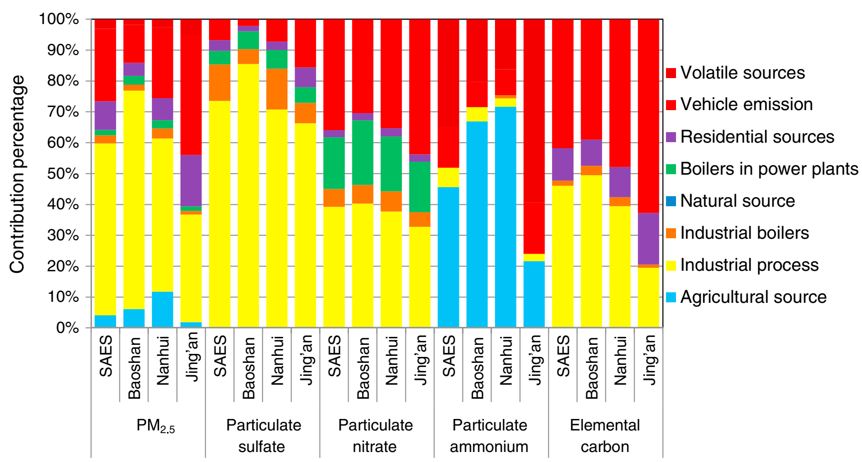

Figure 8. Average contribution percentages of each local emission category to total $PM_{2.5}$ mass, sulfate, nitrate, ammonium, and elemental carbon from local at the SAES (deputy urban), Baoshan (industrial area), Nanhui (rural area), and Jing'an (urban center) sites, respectively, for 6–22 November 2010.

thestudied period are illustrated in Figure 8. The total $PM_{2.5}$ mass concentrations attributed to local emission are dominated by industrial process source at these sites, except that at the urban center site (Jingan), the vehicle emission contribution is marginally higher than the industrial process contribution.

Figure 8 also shows that industrial process contribution remarkably dominates the sulfate concentration resulted from local by 66–85% at the whole Shanghai area. Therefore, controlling the local industrial process emission is an effective measure to reduce the sulfate concentration in Shanghai. As to nitrate concentrations attributed to local sources, industrial process contribution is marginally higher than vehicle emission contribution at these sites except at urban center site. In addition, emission from boilers in power plants is also a notable local source for particulate nitrate besides industrial process and vehicle emission. Compared to the total $PM_{2.5}$ mass and the other three species, there is no notable variation of contribution percentage of each local emission category to nitrate among these sites, also indicating that nitrate has a strong regional behavior.

4. Summary and Conclusions

In order to gain a better understanding of regional impact and the contribution of local emission categories during heavy haze episodes in Shanghai, we chose 6–22 November 2010 in late autumn as a case of heavy haze episodes in this work to carry out a source apportionment simulation of total $PM_{2.5}$ mass and its four species (nitrate, sulfate, ammonium, and elemental carbon) using CAMx with the PSAT tool. Our conclusions are listed below.

On average, about half $PM_{2.5}$ concentration at the urban center is attributed to emission outside Shanghai during the severe haze episodes in late autumn of 2010. Compared to elemental carbon, regional transport contributes much more to sulfate, nitrate, and ammonium in the whole Shanghai Municipality. Based on the source apportionment of total $PM_{2.5}$ mass and its species, emission controlling of $PM_{2.5}$ and the precursors associated with particulate secondary species in upwind adjacent provinces will be an effective measure to alleviate the total $PM_{2.5}$ and its secondary species pollution levels in Shanghai.

As to local contributions from multiple emission categories, industrial process is the dominant contributor to total $PM_{2.5}$ mass in the whole Shanghai except that at urban center, vehicle emission contributes slightly more. For nitrate, industrial process and vehicle emission are the major local contributors, although at urban center, the vehicle emission contribution is remarkably larger. The majority of sulfate concentration, which resulted from local contributions in the whole city, is attributed to industrial processes. For elemental carbon, at the urban center, vehicle emission is the biggest local contributor; however, industrial process contributes slightly more at the rest of the areas in Shanghai. For ammonium, the biggest local contributors at urban center are volatile sources comprised of gas station, oil tanks and depots, coating application, garbage disposal and sewage treatment stations, and residential emission of ammonia, but at the rest of the areas in Shanghai, the biggest local contributor is agricultural source. Overall, controlling vehicle emission and industrial process emission will be of significant benefit for reduction of total $PM_{2.5}$ mass, particulate sulfate, nitrate, and elemental carbon concentrations in Shanghai.

The serve hazes in Shanghai can be roughly classified into two types of representative haze episodes. The first type is the heavy $PM_{2.5}$ pollution with rather weak wind, which is dominantly attributed to local emission. Its biggest local contributors are industrial processes, and the second big local contributor is vehicle emission. The second type is the severe $PM_{2.5}$ pollution in the condition of wind blowing from adjacent provinces with moderate speed. The majority of $PM_{2.5}$ in this type of heavy haze episode is contributed by regional transport from upwind adjacent provinces. These results show that either local emission or regional transport can be the dominant contributor to a severe haze event in late autumn in Shanghai, and furthermore, which is the dominant contributor depends on the meteorological conditions.

This study highlights the important roles of both local emission and regional transport in formation of severe haze episode in late autumn in Shanghai. Particular attention should be paid to emission control in the upwind adjacent provinces, as well as in local, for developing effective strategies to reduce $PM_{2.5}$ pollution in Shanghai.

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