Science of the Total Environment 601–602 (2017) 1476–1487

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Source apportionment of fine particulate matter in China in 2013 using a source-oriented chemical transport model

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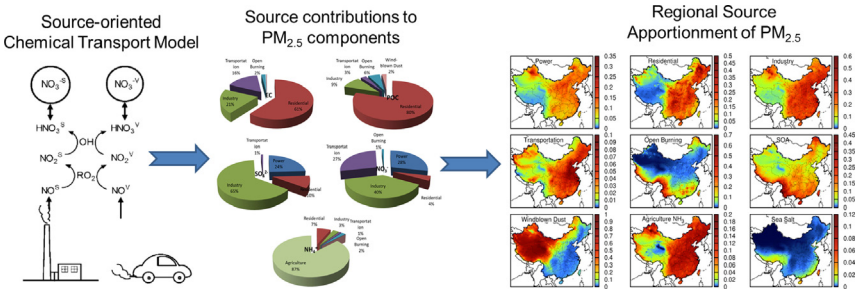
HIGHLIGHTS

- Source contributions to primary $PM_{2.5}$ and SNA were explicitly tracked.
- Residential and industrial sources are the two major anthropogenic sources for primary $PM_{2.5}$ and also for total $PM_{2.5}$.
- Industries, agriculture, power plants, and transportation are important sources for SNA annually.
- Residential emissions are an important source of SNA in winter.
- Large variations of $PM_{2.5}$ sources exist among different regions/provinces.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 14 March 2017
Received in revised form 2 May 2017
Accepted 2 June 2017
Available online xxxx

Editor: D. Barcelo

Keywords:
Source contributions
Primary particulate matter
Secondary inorganic aerosols
Source oriented model
Province

ABSTRACT

China has been suffering high levels of fine particulate matter ($PM_{2.5}$). Designing effective $PM_{2.5}$ control strategies requires information about the contributions of different sources. In this study, a source-oriented Community Multiscale Air Quality (CMAQ) model was applied to quantitatively estimate the contributions of different source sectors to $PM_{2.5}$ in China. Emissions of primary $PM_{2.5}$ and gas pollutants of SO_2, NO_x, and NH_3, which are precursors of particulate sulfate, nitrate, and ammonium (SNA, major $PM_{2.5}$ components in China), from eight source categories (power plants, residential sources, industries, transportation, open burning, sea salt, windblown dust and agriculture) were separately tracked to determine their contributions to $PM_{2.5}$ in 2013. Industrial sector is the largest source of SNA in Beijing, Xi'an and Chongqing, followed by agriculture and power plants. Residential emissions are also important sources of SNA, especially in winter when severe pollution events often occur. Nationally, the contributions of different source sectors to annual total $PM_{2.5}$ from high to low are industries, residential sources, agriculture, power plants, transportation, windblown dust, open burning and sea salt. Provincially, residential sources and industries are the major anthropogenic sources of primary $PM_{2.5}$, while industries, agriculture, power plants and transportation are important for SNA in most provinces. For total $PM_{2.5}$, residential and industrial emissions are the top two sources, with a combined contribution of 40–50% in most provinces. The contributions of power plants and agriculture to total $PM_{2.5}$ are about 10%, respectively. Secondary organic aerosol accounts for about 10% of annual $PM_{2.5}$ in most provinces, with higher contributions in southern

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http://dx.doi.org/10.1016/j.scitotenv.2017.06.019

provinces such as Yunnan (26%), Hainan (25%) and Taiwan (21%). Windblown dust is an important source in western provinces such as Xizang (55% of total $PM_{2.5}$), Qinghai (74%), Xinjiang (59%). The large variation in sources of $PM_{2.5}$ across China suggests that $PM_{2.5}$ mitigation programs should be designed separately for different regions/provinces.

1. Introduction

Airborne particulate matter (PM) has raised great concerns because of its adverse effects on human health and the ecosystem and its important roles in climate change (Menon et al., 2008; Pöschl, 2006; Pui et al., 2014). Fine particulate matter (with aerodynamic diameter ≤ 2.5 μm, $PM_{2.5}$) can cause cancers, cardiovascular and pulmonary diseases, as well as a variety of adverse effects including acute cytotoxicity and immunotoxicity (Gao et al., 2015; Pope et al., 2009). In addition, $PM_{2.5}$ affects solar radiation by absorbing and scattering light, which in turn can lead to degradation of visibility in the atmosphere (Cheung et al., 2005). PM in the atmosphere can be directly emitted from sources such as wildfires, combustion, windblown dust, and sea salt (i.e., primary PM) or formed from emitted gases through atmospheric chemistry and gas-to-particle conversion processes (i.e., secondary PM). PM level is strongly influenced by meteorological conditions and emissions (Steiner et al., 2006; Tai et al., 2010). China has been suffering high levels of PM pollution due to large amounts of fossil fuel consumption associated with rapid economic growth, industrialization, and urbanization in the past a few decades. It is estimated that NO_x emissions in China have increased by 70% from 1995 to 2004 (Zhang et al., 2007), SO_2 by ~60% from 2000 to 2006 (Lu et al., 2011) and organic carbon (OC) by ~30% from 2000 to 2010 (Lu et al., 2011). Consequently, the annual $PM_{2.5}$ mass concentration reached up to 60–140 μg m^{-3} at 21 out of 31 Chinese provincial capital cities in 2013 (Wang et al., 2014d), which was 2–4 times of the National Ambient Air Quality Standards (NAAQS) of China for annual $PM_{2.5}$ (35 μg m^{-3}).

To effectively control $PM_{2.5}$, it is necessary to understand the contributions of different sources. Source apportionment of $PM_{2.5}$ needs to be conducted in different regions of China, as Wang et al. (2008) have demonstrated that the predominant sources of $PM_{2.5}$ vary across different city types. Many studies have used the measurement based statistical receptor models, such as the chemical mass balance (CMB) and the positive matrix factorization (PMF) models, to investigate the contribution of various sources to $PM_{2.5}$ in various regions of China (for examples: Gao et al., 2014; Hu et al., 2014b; Li et al., 2016; Liu et al., 2015; Song et al., 2007; Song et al., 2006a; Song et al., 2006b; Tao et al., 2014; Wang et al., 2016b; Yu et al., 2013; Zhang et al., 2013). These studies have provided valuable information in identifying the important sources of $PM_{2.5}$. However, most of these studies focused on one or a few sites in a single city during measurement periods (often a few weeks in different seasons), making it difficult to have a complete picture of the source contributions of $PM_{2.5}$ at regional scale over long time period. More importantly, the statistical receptor models have limited capability to resolve source contributions to secondary PM. Secondary inorganic aerosols (sulfate (SO_4^{2-}), nitrate (NO_3^-) and ammonium (NH_4^+), SNA) and secondary organic aerosols (SOA) were treated as a separate "source category" in these studies. Ambient measurements have revealed that secondary PM accounts for a large fraction of the total $PM_{2.5}$ in China (Huang et al., 2014). SNA were identified to be the predominant components of $PM_{2.5}$ in China, making up to 30–40% of the total $PM_{2.5}$ mass annually (Cao et al., 2012; Yang et al., 2011), and even >50% of $PM_{2.5}$ mass during some heavy pollution episodes (Zheng et al., 2015). Considering the high contributions of SNA and SOA to total $PM_{2.5}$, it is very important to determine the contributions of different sources to SNA and SOA concentrations to develop efficient emission control strategies.

Chemical transport models (CTMs) have been developed and applied to estimate the contributions of different sources to $PM_{2.5}$ in China in recent years, including the brute force method (Wang et al., 2014c), the tracer-based Particulate Source Apportionment Technology in the CAMx model (Li et al., 2015b; Lu and Fung, 2016), and the source-oriented CTM method (Hu et al., 2015b; Wang et al., 2014a; Zhang et al., 2012a). Wang et al. (2014c), Li et al. (2015b), and Wang et al. (2014a) investigated the source contributions to $PM_{2.5}$ during the severe haze episode of January 2013 in the southern Hebei province in north China, in the Yangtze River Delta (YRD) region in eastern China, and in Shaanxi province in northwest China, respectively. Lu and Fung (2016) and Zhang et al. (2012b) studied the source contributions to sulfate and nitrate in the Pearl River Delta (PRD) region and over the entire China, respectively, and differences in winter and summer source contributions were contrasted. Hu et al. (2015b) also investigated the seasonal variations in source contributions of primary $PM_{2.5}$ over the entire China. These studies either investigated the sources of $PM_{2.5}$ in certain regions during representative pollution episodes or investigated the sources of a certain fraction of $PM_{2.5}$ in China. A systematic analysis of the source contribution to $PM_{2.5}$ and its components over a full year long period at the individual province level over the entire China has not been reported.

In this study, a source-oriented CTM based on the Community Multiscale Air Quality (CMAQ) modeling system was applied to quantify the contributions of different source sectors to $PM_{2.5}$ as well as its primary and secondary inorganic components. Simulations of the entire year of 2013 were conducted to determine the source contributions to $PM_{2.5}$ in each individual province of China. The results of this study will improve our understanding of $PM_{2.5}$ sources in China and provide insights on designing effective control strategies.

2. Method

The host model for the source-oriented CTM is CMAQ v5.0.1 (http://www.cmascenter.org/cmaq/), originally developed by the U.S. Environment Protection Agency (EPA) (Byun and Ching, 1999). To improve the capability of the model in predicting secondary $PM_{2.5}$ formation (including both SNA and SOA), a few changes have been made to the original CMAQ v5.0.1, including heterogeneous formation of sulfate and nitrate (Ying et al., 2014a), isoprene epoxydiol and methacrylic acid epoxide from isoprene oxidation and SOA formation from surface controlled reactive uptake of dicarbonyls, isoprene epoxydiol and methacrylic acid epoxide (Li et al., 2015a; Ying et al., 2015). The source-oriented CMAQ model is applied to simulated the air quality in China in the entire year of 2013 over East Asia, covering the entire mainland China with a horizontal resolution of 36 × 36 km. Details of the modeling domain, emission inputs, meteorological inputs, initial and boundary conditions have been described in previous studies (Hu et al., 2016a; Hu et al., 2017) and therefore are not repeated here.

The modified source-oriented CMAQ model tracks primary PM and SNA from different sources separately through the calculation of all atmospheric processes including emissions, transport, gas and particle phase chemistry, gas-to-particle conversion, deposition. The algorithms for tracking primary PM and SNA were first developed in the University of California, Davis/California Institute of Technology (UCD/CIT) model (Held et al., 2005; Hu et al., 2014a; Mysliwiec and Kleeman, 2002; Ying and Kleeman, 2006; Ying et al., 2008; Ying et al., 2009; Zhang

and Ying, 2010) and implemented into the CMAQ model afterwards (Hu et al., 2015b; Wang et al., 2014b; Ying et al., 2014b; Zhang et al., 2014b; Zhang et al., 2012a). The major features of the algorithms are summarized here and more details can be found in the above references.

The source-oriented CMAQ model tracks primary particles emitted from different sources by adding artificial non-reactive tracers (which emission rates are 0.001% of the total primary particle emissions) to represent total primary mass from these sources (Hu et al., 2015b). This small ratio is to ensure the particle size and mass will not be significantly altered to affect the chemistry and deposition processes. The mass concentration of primary PM from a given source then can be calculated by scaling the simulated artificial tracer concentration from that source with a factor of 1×10^5. Source specific emission profiles are used to estimate the concentrations of primary PM components (such as elemental carbon (EC) and primary organic carbon (POC)) using the following equation:

$$C_{i,j} = T_i \times A_{i,j} \quad (E1)$$

where $C_{i,j}$ represents the concentration of the jth primary PM component from the ith source. T_i represents the concentration of the total primary PM from the ith source, and $A_{i,j}$ represents the ratio of the jth component in the total primary PM from the ith source. The source specific profiles for EC and POC are provided in Hu et al. (2015b).

The source-oriented CMAQ model determines the source contributions to sulfate, nitrate, and ammonium by tracking SO_2, NO_x and NH_3 from different sources separately through the gas chemistry, aerosol chemistry and gas-to-particle conversion processes (Zhang et al., 2012a). The SAPRC11 photochemical mechanism and the aerosol module is expanded in this study so that SO_2, NO_x and NH_3 and their chemical reaction products leading to SNA formation are labeled with the source-identity. For example, nitrate formation from the gas phase reaction of NO_2 with hydroxyl radical (OH) in the original CMAQ model is as follows:

$$NO_2 + OH \rightarrow HNO_3(g) \leftrightarrow NO_3^- \quad (R1)$$

where $HNO_3(g)$ and NO_3^- represent the gas phase nitric acid and particulate nitrate, respectively. In the source-oriented model, if there are two sources (X1 and X2) for NO_2, NO_2 is expanded to two species NO_2_X1 and NO_2_X2. Thus, R1 can be rewritten as:

$$NO_2\_X1 + OH \rightarrow HNO_3\_X1 \leftrightarrow NO_3^-\_X1 \quad (R2)$$

$$NO_2\_X2 + OH \rightarrow HNO_3\_X2 \leftrightarrow NO_3^-\_X2 \quad (R3)$$

The concentration of NO_2_X1 and NO_2_X2 therefore contains the contribution of NO_2 emissions from source X1 and source X2 to nitrate formation. A similar treatment is conducted for all reactions involved in the formation of SNA.

In this study, emissions of primary PM, SO_2, NO_x and NH_3 were grouped into eight source categories: power plants, residential sources, industries, transportation, open burning, sea salt, windblown dust, and agriculture to track the contribution of each source to $PM_{2.5}$ in China. It should be noted that SOA is treated as a separate "source category" in this study, due to large uncertainties associated with emissions of its precursors, i.e. volatile organic compounds (VOCs), and incomplete SOA formation pathways, although the source-oriented SOA algorithm has been developed and applied to study the sources of SOA in the United States (Hu et al., 2016b; Zhang and Ying, 2011). SOA source apportionment study for China using the source-oriented model is undergoing and will be provided in a separate manuscript.

3. Results

3.1. Model evaluation

The capability of CTMs in predicting the concentrations, temporal and spatial variations of $PM_{2.5}$ and chemical composition of $PM_{2.5}$ needs to be evaluated before confidence can be made in the source contribution estimation from the models. Previous studies have reported the capability of the source-oriented CMAQ model in predicting air quality in the United States (Zhang et al., 2014a) and in China (Wang et al., 2014b; Zhang et al., 2012b). Recently, the source-oriented CMAQ model has been applied to simulate air quality in China for the entire year of 2013 (Hu et al., 2016a; Hu et al., 2017). The capability of the model to predict O_3 and $PM_{2.5}$ mass concentrations has been evaluated with observation data from 422 sites in 60 major cities in China (Hu et al., 2016a). EC and OC in $PM_{2.5}$ as well as VOCs, have been evaluated with available observations at multiple sites in different seasons (Hu et al., 2017). Mean fractional bias (MFB), mean fractional error (MFE), mean normalized bias (MNB), and mean normalized error (MNE) of hourly concentrations were calculated to verify model performance. Details of the model performance were discussed in the above references, therefore only a brief summary is described here. Predicted O_3 and $PM_{2.5}$ are in good agreement with observed O_3 and $PM_{2.5}$, with the overall model performance within the performance criteria for O_3 (MNB ≤ ±15% and MNE ≤ 35%, suggested by the U.S. EPA (2007)) and for $PM_{2.5}$ (MFB and MFE as a function of the $PM_{2.5}$ concentrations, generally MFB ≤ ±60% and MFE ≤ 75% when $PM_{2.5} \geq 5\ \mu g\ m^{-3}$, suggested by Boylan and Russell (2006)). Model predictions of EC, OC and VOCs also generally agree with observations at a few available measurement sites, and capture the observed temporal variations. Model performance is better in the more economically developed regions such as the Yangtze River Delta (YRD) and the North China Plain (NCP) compared to the less developed regions such as the northwest China, due to more accurate emission inventories in the more developed regions.

Ambient measurement studies have revealed that SNA account for a large fraction of $PM_{2.5}$ total mass in China (Quan et al., 2014; Zhang et al., 2014c), especially during extreme haze pollution events (Huang et al., 2014). Fig. 1 shows the predicted spatial distribution of SNA to $PM_{2.5}$ ratio in 2013. Overall, SNA is the major PM component in eastern China, accounting for over 50% of $PM_{2.5}$ in large areas in the east, confirming the importance of apportioning sources of SNA for accurate estimation of source contributions of $PM_{2.5}$ in China. An evaluation of the model performance on SNA was conducted in the present study using data that span the entire year of 2013. Such a long-term

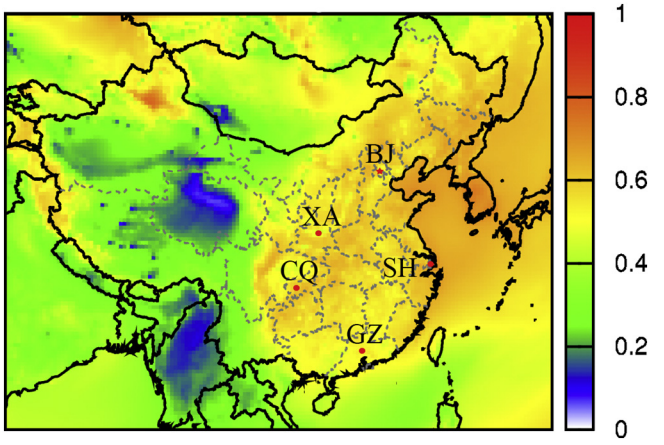

Fig. 1. SNA/$PM_{2.5}$ ratios in 2013. The red dots show the locations of BJ (Beijing), SH (Shanghai), GZ (Guangzhou), XA (Xi'an), and CQ (Chongqing), the 5 megacities shown in Fig. 5. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

comparison of predicted vs. observed the SNA concentrations in China has not been reported in previous studies.

SO_4^{2-}, NO_3^- and NH_4^+ measured at five locations during various sampling periods in 2013 were used to evaluate the model performance on SNA. Table S1 lists the information of measurement sites and sampling periods. Daily $PM_{2.5}$ samples (from 08:00 h to 08:00 h the next day) were collected at the five sites using the medium-volume samplers (TH-150CIII, 100 L min^{-1}, Tianhong Co., Wuhan, China) onto 90-mm quartz fiber filters (Whatman QM/A, Maidstone, UK). Concentrations of NH_4^+ were measured by Dionex-600 and concentrations of NO_3^- and SO_4^{2-} were measured by Dionex-2100 Ion Chromatograph (Dionex Inc., Sunnyvale, CA, USA). Detailed information about the sampling, sample treatment, and lab analysis have been included in Xu et al. (2016). Fig. 2 shows predicted and observed SO_4^{2-} concentrations at the five sites. High SO_4^{2-} concentrations over 40 μg m^{-3} are frequently observed and large daily variations of the observed SO_4^{2-} concentrations occur at all the sites. The model captures the temporal variations of SO_4^{2-} at each monitoring site, but generally under-predicts the concentrations with negative MFB values. Especially, the model tends to underpredict the high peaks of SO_4^{2-} concentrations. The model performance of SO_4^{2-} is similar among the different sites. The MFB values (ranging from −0.56 to −0.29) all within ±60%, meeting the performance criteria, and the MFE values (ranging from 0.62 to 0.80) are around the criteria boundary of 75%.

Figs. 3 and 4 show the comparison of predicted and observed daily average NO_3^- and NH_4^+ concentrations at the five sites, respectively. Large daily variations of NO_3^- and NH_4^+ are also observed. The model generally reproduces the observed temporal variations in the observations at all the monitoring sites. The MFB values of NO_3^- (ranging from −0.47 to 0.19) and NH_4^+ (ranging from −0.44 to −0.01) are slightly better than those of SO_4^{2-} and the MFE values (ranging from 0.58 to 0.83 for NO_3^-, and 0.65 to 0.82 for NH_4^+) are similar to those of SO_4^{2-}.

3.2. Seasonal and regional source apportionment of $PM_{2.5}$

Contributions of different sources to the primary and secondary components of $PM_{2.5}$ were separately calculated and used to estimate the source contributions to total $PM_{2.5}$. Since the source apportionment of primary $PM_{2.5}$ of China in 2013 has been recently reported in a previous work (Hu et al., 2015b), the main focuses of this paper are on the source contributions to SNA and the total $PM_{2.5}$.

Fig. 5 shows the seasonal source contribution to SNA in five representative megacities in different regions, which are Beijing (NCP),

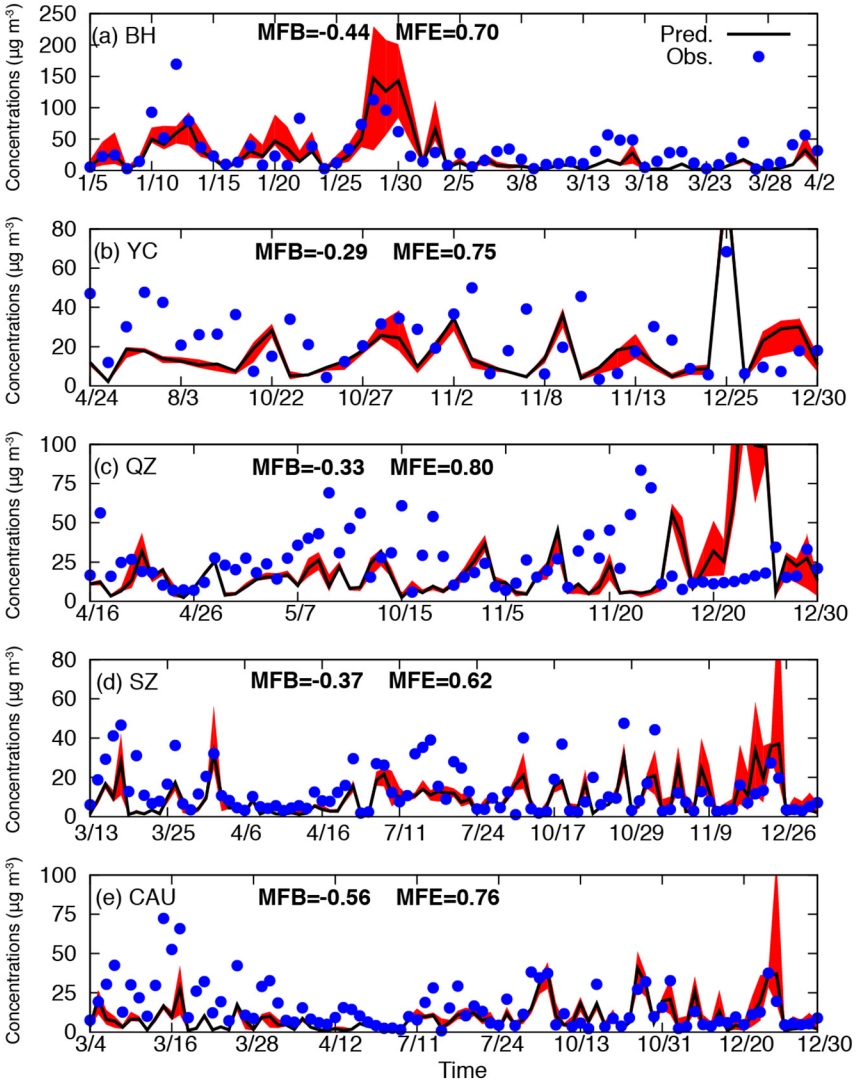

Fig. 2. Observed and model simulated SO_4^{2-} concentrations at 5 sites. Shaded areas represent the minimum and maximum of the concentrations within 3 × 3 grid cells that surround the observation stations (Same in Figs. 3 and 4).

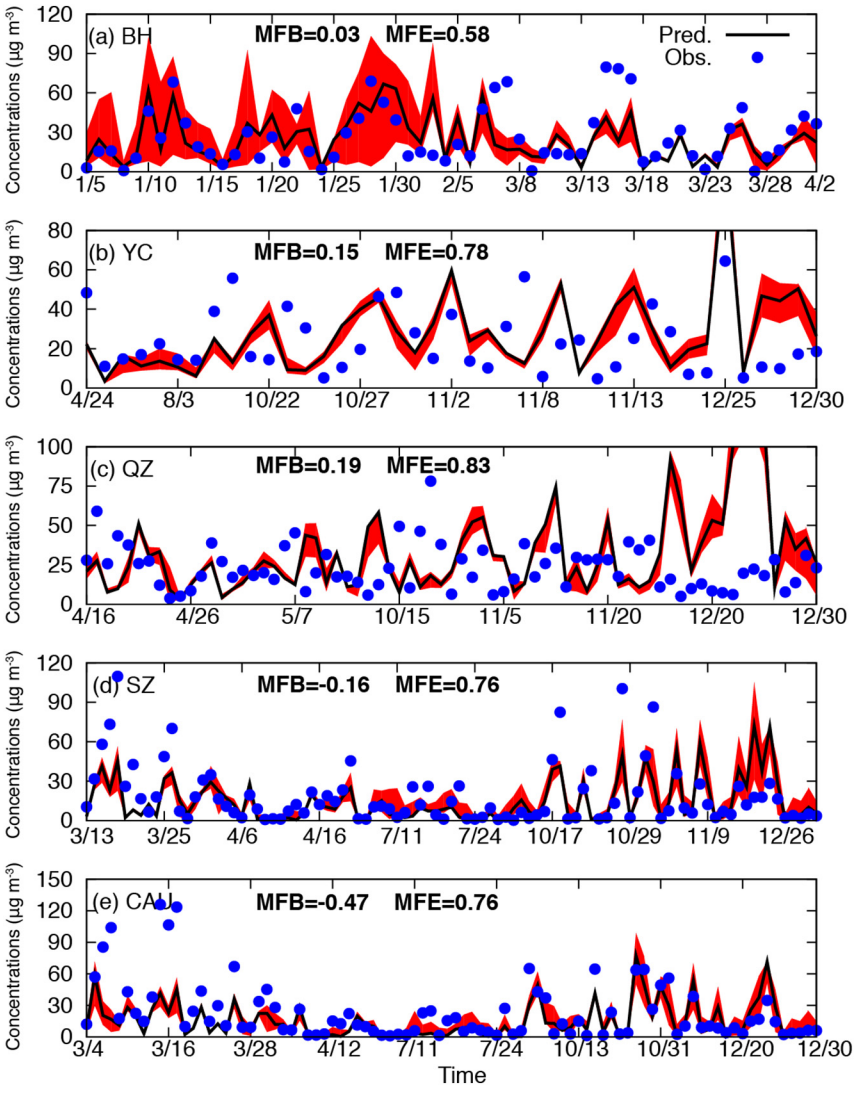

Fig. 3. Observed and model simulated NO_3^- concentrations at 5 sites.

Shanghai (YRD), Guangzhou (RPD), Xi'an (the Guanzhong Plain) and Chongqing (the Sichuan Basin). They are featured with high population densities (together accounting for >8% of the total population of China, population data is shown in Table S2) and severe air pollution problems (Hu et al., 2015a). Source contributions to SO_4^{2-} and NO_3^- in the four seasons are shown in Fig. S1 and Fig. S2, respectively. Results for NH_4^+ are not shown since agriculture is the single dominant source based on the current inventory. SNA concentrations at all the five cities have a similar seasonal pattern, with the lowest values in the summer and the highest values in the winter. The source contributions to SNA at Beijing, Xi'an and Chongqing are similar, which are dominated by industrial emissions in all seasons, with higher relative contributions in summer and fall. This can be seen in SO_4^{2-} and NO_3^- as well (Figs. S1 and S2), with industrial emissions having the highest contributions to the both components across all cities in the entire year. Agriculture and power plants are the 2nd and 3rd source of SNA, contributing to about 25% and 20% respectively in all cities with little variations among all seasons. Residential emissions are also an important source of SNA, especially in the winter. The importance of residential emissions in the NCP region has been demonstrated by Liu et al. (2016). This study shows that residential emissions also need to be considered for improving air quality in other regions.

Fig. 6 shows the predicted regional distribution of seasonal averaged source contributions to SNA in China. SNA from power plants are widely spread, especially in the NCP, Sichuan Basin, and the Northeast Plain. The contribution of power sector to SNA is >20 μg m^{-3} in winter in these regions but is much less in summer. Industrial sector has a similar spatial pattern as the power sector but with greater contributions. The contribution of industrial sector is up to 20 μg m^{-3} even in summer in NCP. Residential sector shows very strong seasonal and spatial variations. It becomes a significant source of SNA in winter, with the maximum contribution of >20 μg m^{-3} in NCP and the Northeast Plain. Agriculture sector also has the similar seasonal variations due to higher SO_4^{2-} and NO_3^- formed in the winter. Contribution of agriculture dominates in eastern China where most of the agricultural activities are located. Open burning in South Asia countries in spring, fall and winter can affect the SNA concentrations in the southern provinces near the sources. More details on regional distribution of seasonal source apportionment of SO_4^{2-} and NO_3^- are shown in Fig. S3 and S4, respectively.

Fig. 7 shows the regional distribution of annual average total $PM_{2.5}$ mass concentrations from all the sources. The ranking of source contributions from the largest to the smallest is industries, residential, agriculture, power, transportation, SOA, windblown dust, open burning and sea salt. The results clearly demonstrate that $PM_{2.5}$ from industrial, residential, power, transportation and agriculture are similarly high in the more populous and economically developed eastern China. Contributions from SOA and opening burning are high in southern China, while $PM_{2.5}$ from windblown is more concentrated in the western China.

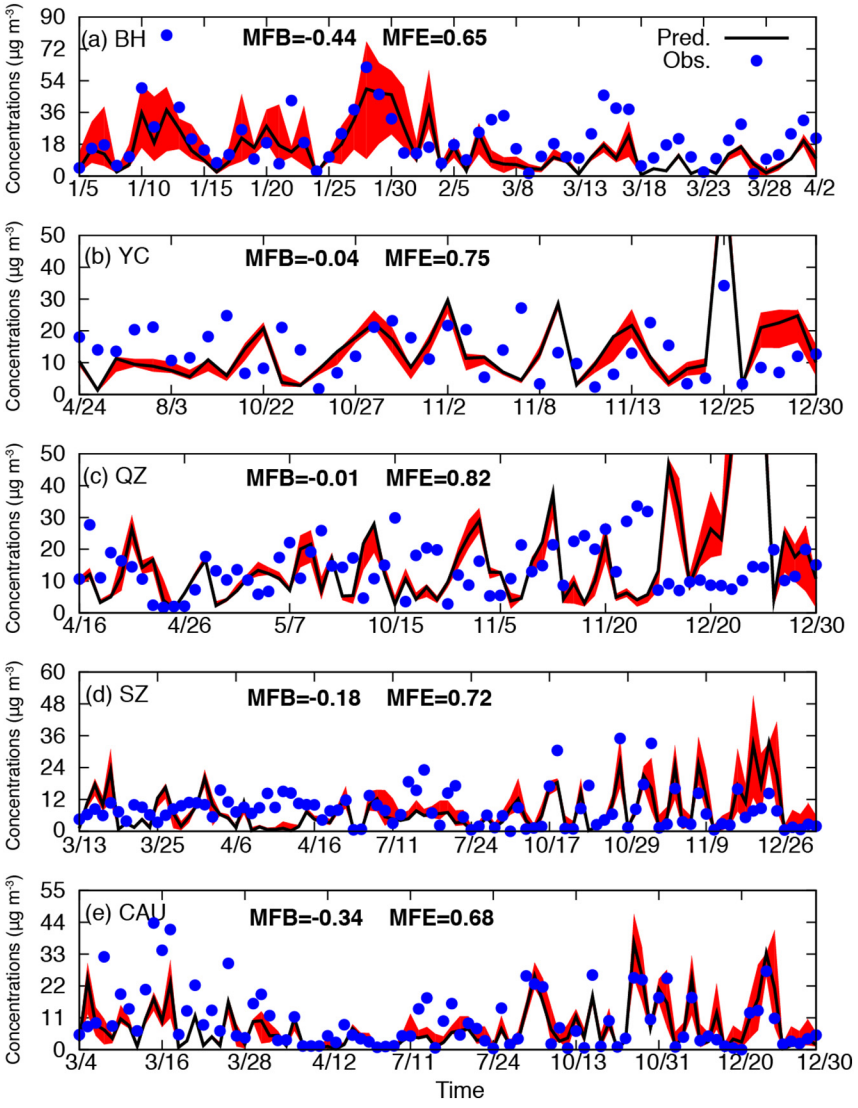

Fig. 4. Observed and model simulated NH_4^+ concentrations at 5 sites.

3.3. Provincial source apportionment of $PM_{2.5}$

Source contributions to the primary $PM_{2.5}$ were estimated over the entire year of 2013 for all the provinces in China, by averaging the results of all the grid cells in each province, as shown in Table 1. The population and the region of each province is shown in Table S3. Residential and industrial sources are the two major anthropogenic source sectors of primary $PM_{2.5}$ in most provinces, but with large variations. Residential sector contributes over 35% of the primary $PM_{2.5}$ in provinces with high population density such as Beijing, Tianjin, Chongqing, Hebei, Jiangsu, Anhui, Shandong, and Henan. It has the largest contribution of 46% in Anhui. However, <5% of primary $PM_{2.5}$ from residential sources is found in Xizang (Tibet), Qinghai, and Xinjiang where few people reside in most of the regions there. Industrial emissions contribute >30% of the primary $PM_{2.5}$ in Tianjin, Shanghai, Hebei, Jiangsu, Zhejiang, and Shandong provinces, most of which are in the most developed regions of eastern China, but the its contribution in Xizang, Qinghai and Xinjiang is negligible. Windblown dust emissions are also important for the primary $PM_{2.5}$ almost in all the provinces. It is the dominant source of primary $PM_{2.5}$ in the provinces in western China, i.e., Xizang, Gansu, Qinghai, Xinjiang, Neimeng, and Ningxia. The primary $PM_{2.5}$ from power plants and transportation sources are marginal, accounting for <5% in most of the provinces. Seasonal open biomass burning emissions are important for primary $PM_{2.5}$ in provinces in southern China such as Fujian, Jiangxi, Hunan, Guangdong, Guangxi, Yunnan, and Taiwan. Sea salt is found to have some impacts in the two island provinces of China, 7% in Hainan and 11% in Taiwan. The provincial average source contributions to the primary $PM_{2.5}$ components of EC and POC are shown in Tables S4 and S5, respectively.

The provincial average source contributions to SNA are very different from those to the primary $PM_{2.5}$, as shown in Table 2. The individual results of SO_4^{2-}, NO_3^- and NH_4^+ can be found in Tables S6–S8, respectively. Industrial emissions, contributing about 40% of SNA, >60% of SO_4^{2-} and 30% of NO_3^-, are the largest source for all the provinces except for Xizang. Emissions from agriculture, power plants and transportation, which only contribute to a minor fraction of the primary $PM_{2.5}$, contribute about 25%, 20%, and 10% of SNA in most provinces. In contrast, residential emissions, which are the most important source of the primary $PM_{2.5}$, generally contribute to <10% of SNA. Agriculture emissions are the single dominant source of NH_4^+. Power emissions are important for SO_4^{2-} and NO_3^-, contributing ~20% and 30%, respectively. Transportation is an important source of NO_3^-, contributing ~30% of NO_3^-; but it contributes a negligible fraction of SO_4^{2-}. Open burning is a minor source of SNA in all provinces except for Xizang where SO_2

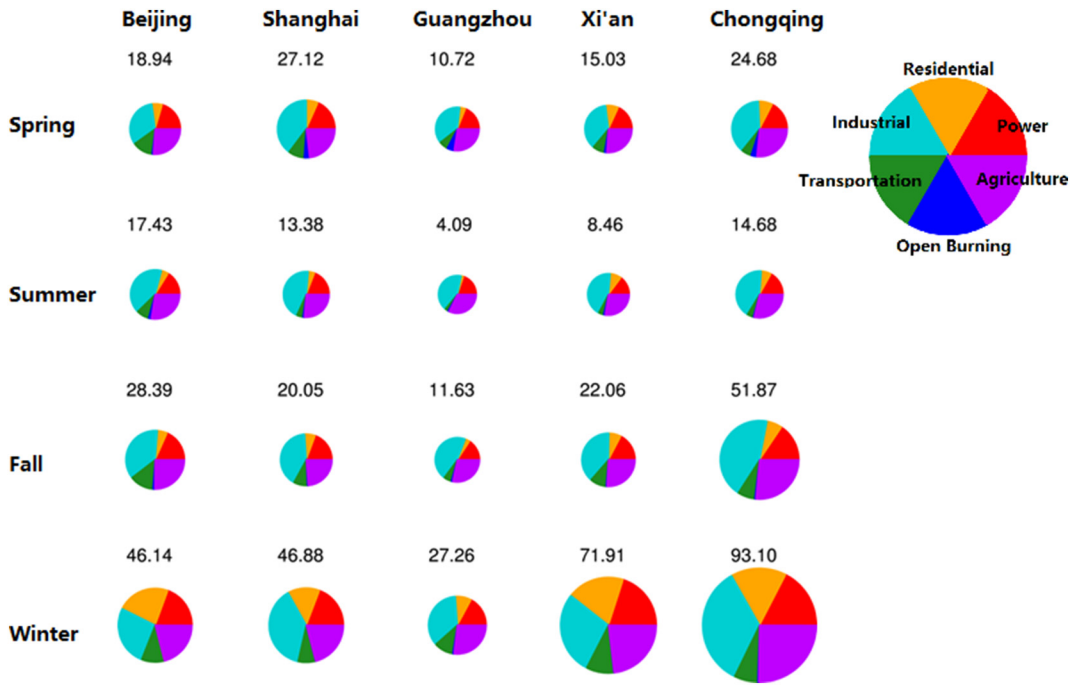

Fig. 5. Seasonal source contributions to SNA at 5 cities. The numbers illustrate the annual average SNA concentrations. Units are μg/m^3.

and NO_x emissions from other sources including industrial, power plants and transportation are very low.

After explicitly resolving the source contributions to the primary $PM_{2.5}$ and SNA (SOA treated as a separate source), the source contributions to the total $PM_{2.5}$ mass then were determined, as shown in Table 3. Residential and industrial emissions are the top two sources of the total $PM_{2.5}$, with a combined contribution of 40–50% in most provinces. The contributions from power plants and agriculture are similar, accounting for about 10%. SOA accounts for 10% of the total $PM_{2.5}$ in most provinces, with more contributions in southern provinces such as Yunnan (26%), Hainan (25%), Taiwan (21%), Guangxi (18%), and Guangdong (17%) on the annual basis. Windblown dust is important for the total $PM_{2.5}$ in western provinces such as Xizang (55%), Qinghai (74%), Xinjiang (59%), Gansu (28%), and Neimeng (22%). Open burning is important in southern provinces, with contributions >10% in Fujian, Jiangxi, Hunan, Guangdong, Guangxi, Yunnan, Xizang, Hainan, and Taiwan.

4. Discussion

Although the capability of the source-oriented CTM method in predicting the source contributions to air pollutants has been demonstrated in previous studies for different regions and episodes, the predicted source contributions in the present study are influenced by several factors. First, accurate emissions are required for better estimation of source contributions to $PM_{2.5}$ and its components. However, large uncertainties exist in the emission data (such as activity levels, emission source fractions and emission factors) (Akimoto et al., 2006; Lei et al., 2011). For example, Huang et al. (2011) evaluated an emission inventory of the YRD region and found the overall uncertainties for SO_2, NOx, CO, PM_{10}, $PM_{2.5}$, VOCs and NH_3 were ±19.1%, ±27.7%, ±47.1%, ±117.4%, ±167.6%, ±133.4% and ±112.8%, respectively. The uncertainties in the emissions of SO_2, NO_x and NH_3 would affect the accuracy of the estimation of source contributions to SNA. In addition, the uncertainties of emissions vary in different regions. The emission estimates are generally more accurate in the NCP, YRD and PRD regions, due to accumulation of more air quality research for years. Therefore, the estimation of source contributions in the provinces located in these regions are expected to be more accurate, compared to the provinces located in northwest and southwest China.

Second, the source apportionment of SNA is also affected by the representation of their chemical transformation processes. Recent studies suggested that SO_2 can be oxidized by NO_2 to form SO_4^{2-} in aerosol water with sufficient NH_3 during severe haze pollution in China (Cheng et al., 2016; Wang et al., 2016a). This process is not included in the model when the simulations described in this study were conducted. The model performance and the source contributions may be changed with this new SO_4^{2-} formation pathway, and the impact needs to be quantified when the detailed mechanisms are implemented in CTMs in the future.

The provincial average source contributions are calculated as the average of all the grids in each province, therefore they are different the from source contributions of the major cities in that province. The $PM_{2.5}$ pollution is a regional problem and it is meaningful to consider its sources in a province or even at a regional scale rather than at a city scale when designing $PM_{2.5}$ mitigation programs. Although the model predictions of $PM_{2.5}$ in this study using a 36 km horizontal resolution generally agree with observations, finer grid resolutions are recommended for source apportionment studies in different regions/provinces.

For designing effective air pollution control strategies, it is also important to understand the contributions from local emission sources vs. regional transport sources. Some studies have suggested that regional and long-distance transport can contribute significantly during heavy PM pollution events (Fu et al., 2016; Hu et al., 2015b; Jiang et al., 2015; Liang et al., 2016; Ying et al., 2014b). Further studies are recommended to investigate the drivers of the regional and long-distance transport for different regions, and to quantify the contributions of different sources during the transport processes.

5. Conclusions

In this study, the source-oriented CMAQ model was applied to quantify the contributions of different sources to the primary and secondary $PM_{2.5}$ components and the total $PM_{2.5}$ of China in the entire year of 2013. In addition to the good performances in predicting the total $PM_{2.5}$, OC and EC, the model can generally capture the temporal variations of observed SO_4^{2-}, NO_3^- and NH_4^+ at five monitoring sites. SNA are the major $PM_{2.5}$ components in China, accounting for over 50% of

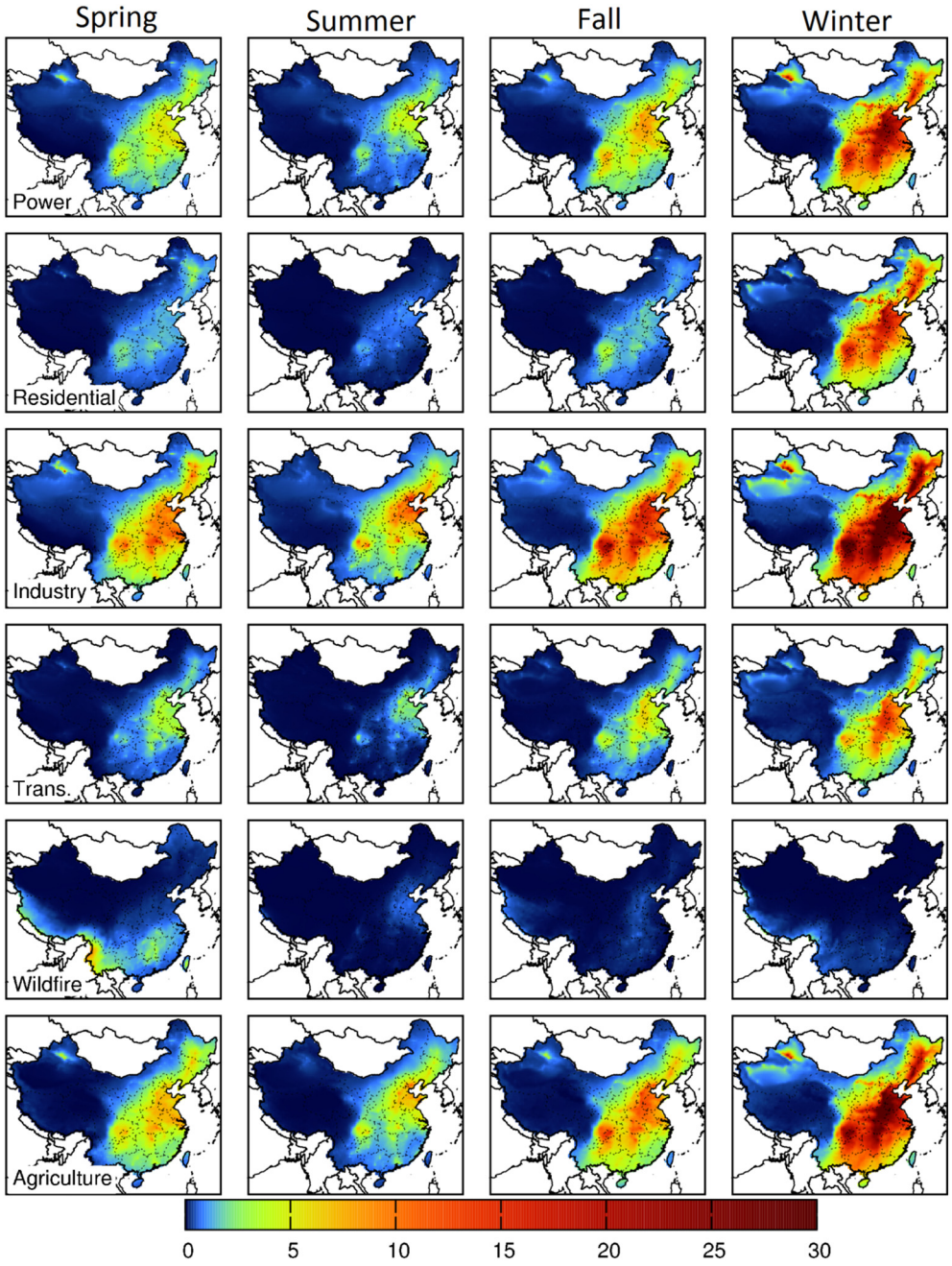

Fig. 6. Regional source apportionment of SNA in 2013. 'Trans.' in the figure represents the transportation section. Units are μg m^{-3}.

the total $PM_{2.5}$. SNA concentrations in the five major cities of Beijing, Shanghai, Guangzhou, Xi'an and Chongqing have a similar season pattern, with the lowest concentration in the summer and the highest in the winter. Industrial sector is the largest source of SNA in Beijing, Xi'an and Chongqing, followed by agriculture and power plants. The contribution from residential emissions is also very important for SNA, especially in winter when severe $PM_{2.5}$ pollution often occurs. For total $PM_{2.5}$, sources contributions from the high to low are industries, residential sources, agriculture, power, transportation, SOA, windblown dust, open burning, and sea salt on a national wide level. The provincial average source contributions were also estimated. Residential and industrial sources are the two major anthropogenic source sectors of primary $PM_{2.5}$, while industries, agriculture, power and transportation are important for SNA in most provinces. For total $PM_{2.5}$, residential and industrial emissions are the top two sources, with a combined contribution of 40–50% to total $PM_{2.5}$ in most provinces. The contributions of power plants and agriculture are approximately 10%. SOA also accounts for approximately 10% of total $PM_{2.5}$ in most provinces, with more contributions in southern provinces such as Yunnan (26%), Hainan (25%) and Taiwan (21%). Windblown dust is important for total $PM_{2.5}$ in western provinces such as such Xizang (55%), Qinghai (74%), Xinjiang (59%), Gansu (28%) and Neimeng (22%). The large variations of $PM_{2.5}$ sources among different regions/provinces should be considered when designing $PM_{2.5}$ mitigation programs. The results in this study enhance our understanding of $PM_{2.5}$ sources in China, and provide valuable information for designing cost-effective control programs in different regions.

Acknowledgement

This project is partly funded by the the National Natural Science Foundation of China (91544220 and 41275121), Natural Science Foundation of Jiangsu Province (BK20150904 and BK20151041), Jiangsu

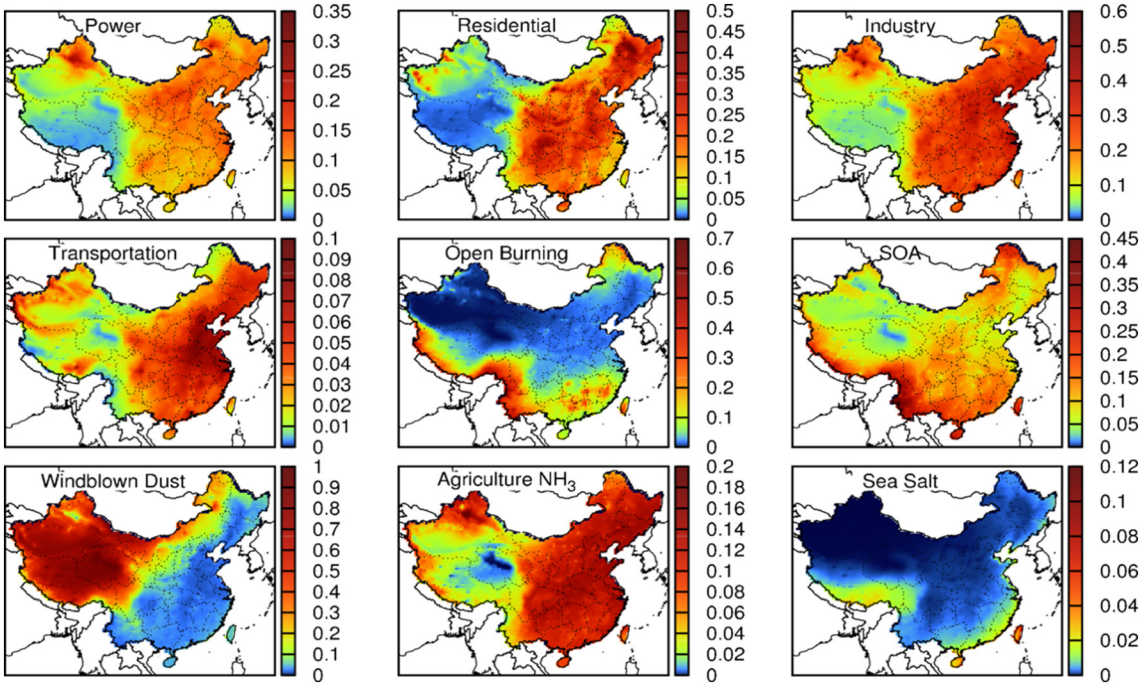

Fig. 7. Regional source apportionment (fractional contributions) of the total $PM_{2.5}$ mass concentration in 2013.

Distinguished Professor Project (2191071503201), Jiangsu Six Major Talent Peak Project (2015-JNHB-010), the Startup Fund for Talent at NUIST (2243141501008) and the Priority Academic Program Development of Jiangsu Higher Education Institutions (PAPD), and Jiangsu Province Innovation Platform for Superiority Subject of Environmental Science and Engineering (No. KHK1201). We would like to thank the computation resources from the Texas A&M Supercomputing Facility (http://sc.tamu.edu/) and the high performance computing resources provided by Louisiana State University (http://www.hpc.lsu.edu) for completing some of the model simulations reported in this study.

Table 1
Provincial average source contributions to the primary $PM_{2.5}$ in 2013.

Province	Power	Residential	Industry	Transportation	Open burning	SOA	Sea salt	Wind-blown dust	Agriculture
Beijing	0.03	0.42	0.27	0.04	0.03	–	0.00	0.21	–
Tianjin	0.03	0.44	0.32	0.04	0.03	–	0.00	0.14	–
Shanghai	0.07	0.19	0.42	0.05	0.06	–	0.02	0.19	–
Chongqing	0.02	0.44	0.21	0.02	0.08	–	0.00	0.23	–
Hebei	0.03	0.37	0.32	0.03	0.03	–	0.00	0.22	–
Shanxi	0.04	0.28	0.28	0.02	0.02	–	0.00	0.36	–
Neimeng	0.03	0.13	0.09	0.01	0.02	–	0.00	0.72	–
Liaoning	0.04	0.39	0.29	0.03	0.02	–	0.01	0.21	–
Jilin	0.04	0.42	0.23	0.03	0.03	–	0.00	0.24	–
Heilongjiang	0.03	0.43	0.15	0.02	0.06	–	0.00	0.30	–
Jiangsu	0.05	0.38	0.30	0.04	0.06	–	0.01	0.16	–
Zhejiang	0.06	0.20	0.30	0.04	0.14	–	0.03	0.23	–
Anhui	0.04	0.46	0.24	0.03	0.08	–	0.01	0.15	–
Fujian	0.05	0.17	0.23	0.03	0.24	–	0.03	0.25	–
Jiangxi	0.03	0.27	0.23	0.03	0.25	–	0.01	0.18	–
Shandong	0.04	0.43	0.30	0.05	0.03	–	0.01	0.15	–
Henan	0.03	0.43	0.27	0.04	0.04	–	0.00	0.18	–
Hunan	0.02	0.35	0.22	0.02	0.20	–	0.01	0.17	–
Hubei	0.03	0.43	0.24	0.02	0.07	–	0.00	0.20	–
Guangdong	0.04	0.26	0.21	0.02	0.20	–	0.03	0.23	–
Guangxi	0.03	0.28	0.22	0.02	0.24	–	0.01	0.21	–
Hainan	0.03	0.27	0.17	0.02	0.16	–	0.07	0.28	–
Sichuan	0.02	0.39	0.16	0.01	0.08	–	0.00	0.33	–
Yunnan	0.01	0.17	0.11	0.01	0.46	–	0.01	0.23	–
Xizang	0.00	0.00	0.00	0.00	0.04	–	0.00	0.95	–
Shaanxi	0.03	0.35	0.18	0.02	0.03	–	0.00	0.39	–
Gansu	0.01	0.13	0.07	0.01	0.01	–	0.00	0.77	–
Qinghai	0.00	0.02	0.01	0.00	0.00	–	0.00	0.96	–
Ningxia	0.03	0.20	0.12	0.01	0.02	–	0.00	0.63	–
Guizhou	0.03	0.39	0.18	0.01	0.14	–	0.01	0.25	–
Xinjiang	0.00	0.02	0.02	0.00	0.00	–	0.00	0.95	–
Taiwan	0.03	0.13	0.13	0.01	0.21	–	0.11	0.38	–

Table 2
Provincial average source contributions to secondary inorganic $PM_{2.5}$ (SNA) in 2013.

Province	Power	Residential	Industry	Transportation	Open burning	SOA	Sea salt	Wind-blown dust	Agriculture
Beijing	0.19	0.07	0.38	0.11	0.01	–	–	–	0.24
Tianjin	0.19	0.06	0.38	0.11	0.01	–	–	–	0.25
Shanghai	0.21	0.06	0.38	0.09	0.01	–	–	–	0.25
Chongqing	0.18	0.10	0.38	0.10	0.01	–	–	–	0.23
Hebei	0.20	0.07	0.38	0.10	0.01	–	–	–	0.25
Shanxi	0.23	0.07	0.38	0.08	0.01	–	–	–	0.23
Neimeng	0.22	0.09	0.37	0.05	0.01	–	–	–	0.25
Liaoning	0.19	0.07	0.41	0.08	0.01	–	–	–	0.24
Jilin	0.19	0.09	0.40	0.07	0.01	–	–	–	0.24
Heilongjiang	0.18	0.11	0.40	0.07	0.01	–	–	–	0.24
Jiangsu	0.21	0.06	0.36	0.12	0.01	–	–	–	0.24
Zhejiang	0.20	0.06	0.39	0.09	0.02	–	–	–	0.24
Anhui	0.20	0.07	0.36	0.13	0.01	–	–	–	0.23
Fujian	0.18	0.06	0.43	0.06	0.03	–	–	–	0.24
Jiangxi	0.18	0.07	0.38	0.10	0.04	–	–	–	0.23
Shandong	0.20	0.06	0.37	0.12	0.01	–	–	–	0.24
Henan	0.20	0.07	0.35	0.13	0.01	–	–	–	0.24
Hunan	0.18	0.08	0.37	0.11	0.02	–	–	–	0.23
Hubei	0.18	0.08	0.37	0.12	0.02	–	–	–	0.23
Guangdong	0.18	0.07	0.41	0.07	0.03	–	–	–	0.23
Guangxi	0.18	0.08	0.39	0.09	0.04	–	–	–	0.23
Hainan	0.17	0.07	0.45	0.03	0.04	–	–	–	0.23
Sichuan	0.17	0.10	0.39	0.09	0.01	–	–	–	0.23
Yunnan	0.17	0.10	0.36	0.05	0.10	–	–	–	0.22
Xizang	0.08	0.03	0.17	0.04	0.30	–	–	–	0.40
Shaanxi	0.21	0.10	0.37	0.09	0.01	–	–	–	0.23
Gansu	0.21	0.09	0.37	0.08	0.01	–	–	–	0.25
Qinghai	0.22	0.07	0.35	0.06	0.01	–	–	–	0.29
Ningxia	0.24	0.08	0.36	0.07	0.01	–	–	–	0.24
Guizhou	0.20	0.11	0.36	0.09	0.02	–	–	–	0.22
Xinjiang	0.23	0.05	0.38	0.06	0.01	–	–	–	0.28
Taiwan	0.18	0.10	0.48	0.01	0.06	–	–	–	0.16

Table 3
Provincial average source contributions to the total $PM_{2.5}$ in 2013.

Province	Power	Residential	Industry	Transportation	Open burning	SOA	Sea salt	Wind-blown dust	Agriculture
Beijing	0.10	0.25	0.31	0.08	0.02	0.08	0.00	0.03	0.12
Tianjin	0.10	0.25	0.33	0.07	0.02	0.07	0.00	0.02	0.12
Shanghai	0.13	0.13	0.40	0.07	0.04	0.08	0.01	0.03	0.13
Chongqing	0.09	0.25	0.29	0.04	0.04	0.12	0.00	0.03	0.13
Hebei	0.11	0.22	0.34	0.07	0.02	0.08	0.00	0.03	0.13
Shanxi	0.13	0.19	0.34	0.05	0.02	0.09	0.00	0.06	0.12
Neimeng	0.12	0.17	0.24	0.03	0.03	0.09	0.00	0.22	0.10
Liaoning	0.12	0.22	0.34	0.06	0.01	0.07	0.00	0.03	0.14
Jilin	0.12	0.25	0.31	0.05	0.02	0.08	0.00	0.03	0.14
Heilongjiang	0.10	0.29	0.26	0.05	0.04	0.09	0.00	0.05	0.12
Jiangsu	0.12	0.20	0.32	0.07	0.03	0.09	0.01	0.02	0.13
Zhejiang	0.12	0.12	0.33	0.06	0.07	0.12	0.01	0.03	0.13
Anhui	0.11	0.24	0.28	0.07	0.04	0.11	0.00	0.02	0.12
Fujian	0.10	0.11	0.29	0.04	0.14	0.14	0.02	0.04	0.11
Jiangxi	0.09	0.16	0.27	0.05	0.14	0.14	0.01	0.03	0.11
Shandong	0.12	0.22	0.33	0.08	0.02	0.08	0.00	0.02	0.14
Henan	0.11	0.23	0.31	0.07	0.02	0.09	0.00	0.02	0.14
Hunan	0.09	0.20	0.28	0.05	0.11	0.12	0.00	0.02	0.12
Hubei	0.10	0.24	0.30	0.06	0.04	0.11	0.00	0.03	0.13
Guangdong	0.09	0.16	0.26	0.04	0.12	0.17	0.02	0.04	0.10
Guangxi	0.08	0.17	0.26	0.04	0.13	0.18	0.01	0.03	0.10
Hainan	0.07	0.17	0.23	0.03	0.10	0.25	0.03	0.04	0.08
Sichuan	0.08	0.27	0.25	0.04	0.06	0.14	0.00	0.06	0.11
Yunnan	0.05	0.12	0.16	0.02	0.29	0.26	0.00	0.04	0.07
Xizang	0.02	0.01	0.05	0.01	0.17	0.14	0.01	0.55	0.03
Shaanxi	0.11	0.24	0.27	0.05	0.02	0.12	0.00	0.06	0.12
Gansu	0.09	0.19	0.21	0.03	0.02	0.09	0.00	0.28	0.09
Qinghai	0.03	0.05	0.08	0.01	0.01	0.05	0.00	0.74	0.02
Ningxia	0.12	0.21	0.24	0.04	0.02	0.10	0.00	0.17	0.10
Guizhou	0.10	0.24	0.26	0.03	0.07	0.13	0.00	0.03	0.12
Xinjiang	0.07	0.05	0.15	0.02	0.01	0.05	0.00	0.59	0.06
Taiwan	0.08	0.10	0.21	0.02	0.16	0.21	0.06	0.06	0.09

Appendix A. Supplementary data

Supplementary data to this article can be found online at http://dx.doi.org/10.1016/j.scitotenv.2017.06.019.

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