Source Contributions of Urban PM_{2.5} in the Beijing–Tianjin–Hebei Region: Changes between 2006 and 2013 and Relative Impacts of Emissions and Meteorology

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31 **Abstract**

32 Anthropogenic emissions in China have been controlled for years to improve ambient air quality.
33 However, severe haze events caused by atmospheric aerosols with aerodynamic diameter less than
34 or equal to 2.5 μm ($\text{PM}_{2.5}$) have continued to occur, especially in the Beijing–Tianjin–Hebei (BTH)
35 region. The Chinese government has set an ambitious goal to reduce urban $\text{PM}_{2.5}$ concentrations
36 by 25% in BTH by 2017 relative to the 2012 levels. Source apportionment (SA) is necessary to the
37 development of the effective emission control strategies. In this work, the Comprehensive Air
38 Quality Model with extensions (CAMx) with the Particulate Source Apportionment Technology
39 (PSAT) is applied to the China domain for the years 2006 and 2013. Ambient surface
40 concentrations of $\text{PM}_{2.5}$ and its components are generally well reproduced. To quantify the
41 contributions of each emission category or region to $\text{PM}_{2.5}$ in BTH, the total emissions are divided
42 into 7 emission categories and 11 source regions. The source contributions determined in this
43 work are generally consistent with results from previous work. In 2013, the industrial (44%) and
44 residential (27%) sectors are the dominant contributors to urban $\text{PM}_{2.5}$ in BTH. The residential
45 sector is the largest contributor in winter; the industry sector dominates in other seasons. A slight
46 increasing trend (+3% for industry and +6% for residential) is found in 2013 relative to 2006,
47 necessitating more attention to these two sectors. Local emissions make the largest contribution
48 (40%–60%) for all receptors. Change of source contribution of $\text{PM}_{2.5}$ in Beijing and northern
49 Hebei are dominated by change of local emission. However, for Tianjin, and central and southern
50 Hebei, change of meteorology condition are as important as change of emission, because regional
51 inflow in these areas is more important than in Beijing and northern Hebei and can increase under
52 unfavorable weather conditions, indicating a strong need for regional joint emission control efforts.

The results in this study enhance the quantitative understanding of the source–receptor relationships and provide an important basis for policymaking to advance the control of PM_{2.5} pollution in China.

Key words: Source apportionment; Regional inflow; Meteorological influence; Joint emission control

1 Introduction

High concentrations of fine particulate matter (defined as particles with aerodynamic diameter less than or equal to 2.5 micrometers, or PM_{2.5}) is the most severe atmospheric pollution problem in many regions of China, with the Beijing–Tianjin–Hebei (BTH) region the most polluted area. During the haze events of January 2013, eight out of the 10 most polluted cities in the country were in BTH, including Xingtai, Shijiazhuang, Baoding, Handan, Langfang, Hengshui, Tangshan, and Beijing (L.-T. Wang et al., 2014). These cities were among the worst-ranked cities by seasonal or annual air quality index during 2013 and 2014. Effective emissions control requires the understanding of PM_{2.5} sources and relative contributions. However, PM_{2.5} consists of very complex chemical components, which may be generated via several paths. Primary particles are emitted directly from anthropogenic (e.g., industry, power plants, and vehicles) or natural (e.g., outdoor biomass burning and dust storms) sources. Secondary particles are produced in the atmosphere from precursor gases (e.g., sulfur dioxide (SO₂), nitrogen oxides (NO_x), ammonia (NH₃), and volatile organic components (VOCs)) emitted from sources similar to those for primary particles.

Quantitative assessment of sources of PM_{2.5} and its components and their contributions (also

known as source apportionment (SA)) is a prerequisite to the formulation of the effective emission control strategies. Source appointment of $PM_{2.5}$ in support of emission control strategies needs to meet two requirements. First, $PM_{2.5}$ must be fully apportioned to sources because total $PM_{2.5}$ is the target of control. Second, the locations of sources must be identified to quantify the role of pollution transport.

Various methods have been developed for SA; several of them are widely used. Chemical Mass Balance (CMB; Watson et al., 1984) is a receptor model that apportions observed pollution concentrations to sources with known source profiles. It assumes that the observed concentrations can be adequately explained by linear combination of a number of sources with fixed source profiles and variable source strengths in time. Other receptor-based approaches with the same assumption, e.g., Positive Matrix Factorization (PMF, Paatero, P. and Tapper, 1994) and UNMIX (Hopke, 2003), are used when source profiles are lacking. These approaches solve the long-term observations with statistical methods and require the user to identify source type from the result, using knowledge of source characteristics. Although receptor models have substantial efficiency in calculation and good performance in SA of primary species, they fail to apportion secondary particles or quantify the contribution of regional transport (Burr and Zhang, 2011a; Lin and Wen, 2015). Trajectory-based methods, e.g., the Potential Source Contribution Function (PSCF; Cheng et al., 1993; Zhang et al., 2013) and Effective Emission Intensity (EEI; Lu et al., 2012), combine gridded emission and back trajectories to estimate regional source contributions. Again, trajectory-based methods are not suitable for SA of secondary particles because they only treat simple chemistry.

The chemical transport model (CTM) has the potential to deal with regional SA and SA of

secondary particles, because both the transport and formation of secondary particles are considered. The simplest method is to turn off emission sources of interest one-by-one in each simulation and estimate their contributions from differences between results of these sensitivity simulations and those of the base one. Such a method, the so-called Brute Force Method (BFM) or zero-out method, has been widely used in source sensitivity studies of major pollutants worldwide (e.g., Burr et al., 2011a, b and Y.-J. Wang et al., 2014).

Source-oriented methods have been developed to obtain source contributions with a single simulation, by tagging species of interest with their sources through all processes in the host model. The source-oriented external mixture (SOEM; Ying and Kleeman, 2006) method is an example. Although SOEM can accurately perform SA, its calculation efficiency is a challenge because the burden of gas-phase chemistry substantially increases with additional source species. The Particulate Source Apportionment Technology (PSAT) does not simulate additional processes of transport, chemistry or deposition for these source species. Instead, changes of such species in these processes are computed in proportion to the change of the original species. PSAT has a much lower burden of gas-phase chemistry calculation than SOEM, and is, therefore, an efficient source tagging method. The Comprehensive Air Quality Model with extensions (CAMx)/PSAT (Yarwood et al., 2005b; Wagstrom et al., 2008) is maintained by ENVIRON and has been updated by both developers and CAMx users. The capability of CAMx/PSAT has been demonstrated by comparing its results with those of other methods (Burr and Zhang, 2011a, b; Bove et al., 2014) and through its wide application in different regions of the world (Bedogni et al., 2008; Wagstrom and Pandis, 2011; Wu et al., 2013; Skyllakou et al., 2014). Several studies have compared results of PSAT with other SA methods. Burr and Zhang (2011a, b) compared different category source

contributions between PSAT and BFM. They reported that the two approaches agreed well for primary species but produced significantly different results for secondary species owing to the consideration of nonlinear effects in the BFM but not in PSAT. Bove et al. (2014) compared SA results from PSAT and a receptor-based method, PMF, by reconstructing emission sectors in the PSAT result. Although results from the two methods agreed well for some categories, large uncertainties existed in this comparison.

Several studies have focused on the source contribution of $PM_{2.5}$ in China. Y.-J. Wang et al. (2014) used CAMx-PSAT in SA research in Shanghai, focusing on source contributions in five emissions categories and from local and surrounding areas. L.-T. Wang et al. (2014) applied the BFM to a Chinese domain to study source contributions in three cities in the Hebei Province, concluding that local emissions from southern Hebei contributed to > 60% of $PM_{2.5}$ in January 2013 for all three cities in the area.

While previous studies provided valuable knowledge of SA of PM, the source contributions will change with changing emissions and may also be affected by a changing climate. Zhang et al. (2014) found large differences of source contributions to $PM_{2.5}$ in the United States (U.S.) between 1999 and 2005, consistent with changes in emissions and/or their relative importance between the two years. Anthropogenic emissions in China have changed rapidly since the 1990s relative to those in developed countries (such as the U.S.) because of increasing population, urbanization, and emission controls. To provide timely information for emission control strategies, SA of $PM_{2.5}$ of China must be updated as frequently as the changes in emissions to quantify the up-to-date source–receptor relationships and should be performed periodically in conjunction with the major updates in emission inventories.

The goal of this work is to evaluate the change in SA of PM_{2.5} in the BTH region from 2006 to 2013 to support the adjustment of emission control policies for the region based on new features of source contributions. To achieve this goal, a regional CTM with the PSAT probing tool is applied over the China domain for two full years: 2006 and 2013. In Section 2, the methods used are described. The simulated results are evaluated in Section 3. SA results for 2006 and 2013 are presented and compared in Section 4. Finally, summarizes the main findings are summarized in Section 5.

2 Methodology

2.1 Model description and configuration

CAMx is an offline 3-D CTM (ENVIRON, 2013). CAMx v.6.0 is used in this work. In CAMx 6.0, the 2005 Carbon Bond (CB05; Yarwood et al., 2005a) mechanism is used for gas-phase chemistry. The aqueous-phase scheme in the Regional Acid Deposition Model (RADM-AQ) (Chang et al., 1987) scheme is used for aqueous-phase oxidation. Atmospheric aerosols, including sulfate (SO₄²⁻), nitrate (NO₃⁻), ammonium (NH₄⁺), elemental carbon (EC), primary organic carbon (POC), secondary organic carbon (SOC), other unknown particulate matter (other), and mineral dust are simulated using a static two-mode (fine and coarse modes) scheme in CAMx. In this scheme, the fine mode includes SO₄²⁻, NO₃⁻, NH₄⁺, EC, POC, and SOC, and both modes include “other” and dust. ISORROPIA (Nenes et al., 1999) is used to simulate inorganic aerosol thermodynamic equilibrium. Dry deposition of aerosols and gases are calculated following Zhang et al. (2001) and Zhang et al. (2003), respectively. Wet deposition of aerosols and gases are calculated following Seinfeld and Pandis (1998). A detailed description of CAMx can be found in the CAMx 6.0 User’s guide (ENVIRON, 2013).

As a species tagging method, PSAT labels emission categories and source regions for particles of interest designed by the user. It then tracks these source species in model integration. After each chemical or physical process (e.g., chemical reaction, deposition, and transport), these species are updated by apportioning the change of corresponding species in the host model (CAMx) to each source. As a result, source information of each selected species on each grid at each time step are delivered and evolved, during which the simulation of the host model is unaffected.

The horizontal resolution is 36-km. The model domain stretches from 68-152°E and 10-54°N, covering the whole mainland of China, and the vertical resolution is 23 layers from the surface up to 100 hPa with 10 layers below 3-km. The meteorological fields simulated by the Weather Research and Forecast (WRF) model version 3.5.1 are used to drive CAMx. WRF simulations use the same domain projection, and horizontal and vertical resolutions as those used for CAMx. The domain size of the WRF simulation is designed to be three grid cells larger than that of CAMx on each boundary to ensure a reasonable meteorological boundary condition. The configuration of the WRF simulations is described in Zheng et al. (2014). Initial and boundary conditions (ICBC) of chemical species are interpolated from the output of GEOS-Chem model (Bey et al., 2001, Geng et al., 2014). One-week spin-up is used to minimize the influence of initial conditions on model results. The anthropogenic emissions are based on the Multi-resolution Emission Inventory for China (MEIC, <http://www.meicmodel.org>). The major source categories for these emissions are further described in Section 2.2. Outdoor biomass burning emissions includes forest and grassland fires and crop burning in fields (Huang et al., 2012). The Model of Emissions of Gases and Aerosols from Nature version 2.1 (MEGAN2.1, Guenther, et al., 2012) is used to provide

biogenic emissions. The dust emissions are generated offline using the dust emission module of Zender et al. (2003) modified by Wang et al. (2012) as implemented in an online-coupled meteorology-chemistry model, WRF with chemistry (WRF/Chem) with physics packages from the Community Atmospheric Model version 5 (WRF-CAM5) (Zhang et al., 2015).

2.2 Simulation design

The selected six emission categories include five anthropogenic sectors of the MEIC; industry, power plant, residential, transportation, and agriculture, and natural sources. Natural sources include outdoor biomass burning, biogenic emissions, and mineral dust. To investigate the role of regional transport, the entire domain is separated into 11 source regions (Fig. 1); Beijing, Tianjin, Northern Hebei (N. Hebei), Central Hebei (C. Hebei), Southern Hebei (S. Hebei), Liaoning, Inner Mongolia, Shanxi, Henan, Shandong, and other areas. Including initial and boundary conditions, there are 68 emission source groups considered in the model ($[6 \text{ sources} \times 11 \text{ source regions}] + 2 = 68$). Thirteen cities in the BTH region are defined as receptors, including Beijing (BJ), Tianjin (TJ), Tangshan (TS), Qinhuangdao (QHD), Chengde (CD), Zhangjiakou (ZJK), Shijiazhuang (SJZ), Baoding (BD), Langfang (LF), Cangzhou (CZ), Hengshui (HS), Xingtai (XT) and Handan (HD). In this work, we use the grid cell at the center of each city to represent the receptor (There are alternative ways to define receptors, such as receptor area. Impact of receptor definition is discussed in Sect. 4 of supplemental materials). Regional SA of urban $\text{PM}_{2.5}$ is defined as the average SA of $\text{PM}_{2.5}$ at the centers of all cities in each source region. For instance, SA of urban $\text{PM}_{2.5}$ for the S. Hebei region refers to the average SA in the cities of Hengshui, Xingtai, and Handan.

CAMx modeling simulations are conducted for 2006 and 2013 with corresponding emissions

and meteorological conditions (referred to as E06M06 and E13M13, respectively, see Table 1). The difference between E13M13 and E06M06 reflects the combined impact of changes in emissions and meteorological conditions. The third simulations used 2006 emissions and 2013 meteorology (E06M13). Then the differences between E06M13 and E06M06 represent the effect of changes in meteorological conditions between 2006 and 2013, while the differences between E13M13 and E06M13 (E13M13 minus E06M13) represent the effect of emission changes between two years. All three simulations are conducted for 1 month in each season (January, April, July, and October) to examine the seasonality of model predictions and the resulting SA.

3 Model evaluation

Several observational datasets are used to evaluate meteorological and chemical prediction in this study. Datasets and protocol used in this evaluation are described in supporting materials. Temperature and relative humidity are generally well reproduced, except that July temperature is underpredicted for both years, which may be affected by predictions of cloud fraction. The wind speed is slightly overpredicted, and, for each season, the U and V components of the wind speed from the prevailing wind direction perform much better than those from the opposite directions. Precipitation is generally underpredicted with Normalized Mean Bias (NMB) from -7.8% to -43.5% and Normalized Mean Error (NME) from 97.4% to 114.1%. Detail performance of meteorological prediction is discussed in supporting materials.

Table 2 shows the performance of simulated $\text{PM}_{2.5}$ for both years. Monthly mean surface concentrations of $\text{PM}_{2.5}$ in 2006 are comparable to observations, with NMB from -13.5% to 20.1% and NME from 40.0% to 51.1%. However, the surface concentrations of most $\text{PM}_{2.5}$ components are not reproduced as well as total $\text{PM}_{2.5}$, except for SO_4^{2-} and NO_3^- (Table S3). The overall good

performances of PM_{2.5} in 2006 likely come from the combination of overprediction of anthropogenic PM_{2.5} and underprediction of mineral dust. Simulated PM_{2.5} also shows generally good performance for 2013, except that it is overpredicted by 71.3% in July. Actually, all short-lived species show larger positive biases in July than in other three months. This overprediction is likely due to uncertainties in MEIC and may be associated with the underprediction of 2-m temperature by WRF. It is noted that the PM_{2.5} overprediction do not affect much SA results in the BTH region in this work, as the overprediction mainly occurs in southern China. Detailed performance of PM_{2.5} components in 2006 and non-PM_{2.5} species in 2013 are discussed in supporting materials.

4 Results and Discussions

4.1 SA of PM_{2.5} in 2013

4.1.1 Sectoral SA in 2013

Figure 2a displays the spatial distribution of annual mean PM_{2.5} concentrations on the North China Plain (NCP) during 2013. The simulated concentrations of PM_{2.5} in most areas exceed the 35 µg m⁻³ standard for annual mean concentrations set by the Ministry of Environment Protection of China. PM_{2.5} concentrations exceed 100 µg m⁻³ in a large area of BTH, in Henan and Shandong, and tend to peak in cities. The PM_{2.5} concentrations are apportioned into six source sectors in Figs. 2 (b–g). The ranking of the domain average source contributions of PM_{2.5} sources from the largest to the smallest is industry, residential, power plant, agriculture, transportation, and natural sources (which is a combination of outdoor biomass burning and mineral dust). In addition to the dominance in magnitude, industry-induced PM_{2.5} also best explains hotspots in the distribution of

total PM_{2.5}. Compared to industry-induced PM_{2.5}, residential-induced PM_{2.5} exhibits a more uniform spatial distribution, because residential emissions are more spatially uniform than industrial emissions. Compared with the former two contributors, PM_{2.5} attributed to power plants, transportation, and agriculture shows much more uniform distributions, although the emissions of power plants and transportation have obvious hotspots in cities. This probably results from the combined effect of several factors, including high emission altitudes of power plant sources, uniform distribution of agricultural sources, long lifetime of secondary inorganic particles, and different major emitted species for each sector (e.g., SO₂ and NO_x for power plants, NO_x for transportation, and NH₃ for agriculture). Unlike the spatial distributions of SA from anthropogenic sources, for natural PM_{2.5}, the impact of dust decreases from west to east, and hotspots are caused by outdoor biomass burning.

Average SA of PM_{2.5} in 13 BTH cities is calculated to represent the SA of urban PM_{2.5} in the region in 2013 (Figs. 3a–e). The average annual urban PM_{2.5} concentration is 117.6 µg m⁻³ in BTH. Its contributions are dominated by industry (44%) and residential (30%), followed by power plants (8%), agriculture (8%), and transportation (7%). The remaining 3% are caused by natural sources and boundary transport. Although this ranking is similar to that of the domain average, the relative magnitudes of individual source contributions vary, reflecting the diversity in PM_{2.5} spatial distribution for different source categories. The concentration of urban PM_{2.5} in BTH peaks in January (221.9 µg m⁻³) with a dominant source contribution from residential (48%), rather than industry (33%), because of substantial residential emissions during winter for heating. The level of urban PM_{2.5} during other months is lower and the residential sector contributions (~10%) are similar to those from the power plant, agriculture and transportation sectors. The source

contributions of natural sources show some seasonality. Outdoor biomass burning and mineral dust tend to peak in summer and spring, respectively. This tendency is clearer in the SA of PM_{10} (Fig. S1) because natural sources are more important than anthropogenic sources for coarse particles.

4.1.2 Regional SA

The above ranking of SA categories provides quantitative information on the sources of total $PM_{2.5}$. However, further understanding of $PM_{2.5}$ control, especially at sub regional scale, requires knowledge of $PM_{2.5}$ source locations. Figure 4 shows regional and categorical SA of urban $PM_{2.5}$ for five selected receptors. For each receptor, SA labeled as sources within corresponding sub regions is considered as local contributions, and sources with contributions > 2% as non-negligible. As expected, local emissions are the most important contributor for each receptor, accounting for 40%–60% of $PM_{2.5}$ (However, when focusing on entire city, e.g. great Beijing, regional contribution could be even higher, as discussed in section 4 of supplementary materials). For most categories, these contributions maximize locally and attenuate with distance. Urban $PM_{2.5}$ at all receptors is dominated by either local industry (e.g., in N. Hebei and Tianjin) or local residential contributions (e.g., in Beijing), with important contributions from local agriculture and local transportation.

All five receptors are also influenced by emissions from surrounding areas, across most categories. Regional industry and regional residential sources dominate the emissions inventory of every sub region and hence the inflow contributions. Although local power plant emissions have minor contributions to each receptor, the effect of regional power plants does not attenuate with distance as much as with other sectors and the total contributions of power plants (local plus

regional) are as important as those of transportation and agriculture sectors. Anthropogenic source regions typically have a substantial influence on nearby receptors only. One exception is Shandong Province whose influence covers the entire BTH region, because of the large amount of emissions (e.g., industry) in Shandong and the highly efficient transport of $PM_{2.5}$ by coastal winds. For natural sources, outdoor biomass burning-induced $PM_{2.5}$ are mainly produced locally, whereas fine dust particles originate from the Inner Mongolia and other arid areas.

4.1.3 Comparison with previous results

As shown in Table 3, despite some differences for several sectors, the categorical SA from this work is generally comparable with other work. Those differences are mainly due to different methods employed in different studies. Compared to the results of L. -T. Wang et al. (2014) using BFM that accounts for the nonlinear relationship of inorganic aerosols, predicted source contributions to $PM_{2.5}$ are larger for power plant and transportation but smaller for agriculture at all three receptors in January 2013. Taking NH_3 as an example, while PSAT accounts for the change in NH_4^+ concentrations resulting from changes in NH_3 emissions, the BFM also accounts for the related change in SO_4^{2-} and NO_3^- , in addition to that for NH_4^+ . What is actually reflected in L. -T. Wang et al. (2014) is that inorganic $PM_{2.5}$ in winter BTH is more sensitive to the variation of agricultural NH_3 than that of SO_2 or NO_x emitted mainly from power plants and vehicles.

Mismatching in categorical partitioning is a challenge when comparing SA from receptor-based methods. Therefore, the reconstruction of categories is necessary. The total biomass burning includes residential combined with outdoor biomass burning but neglects residential coal combustion. This is why the predicted biomass burning contributions to EC and POC are generally greater than those reported by Cheng et al. (2013). More complex steps are

required to compare with the SA result of Zhang et al. (2013). First, SO_4^{2-} , NO_3^- , and NH_4^+ from all sources are combined to obtain a factor of secondary inorganic aerosol (SIA). The remaining contribution of transportation is primary particles from vehicles. Following Cheng et al. (2013), 50% of total EC and POC are then considered to represent biomass burning contributions. Mineral dust is renamed as soil dust. Finally, all other contributions are combined into a mixed category, coal combustion plus industrial processes. Comparability between these reconstructed categories and those of Zhang et al. (2013) remains poor. For example, the PMF factor SIA does not actually include all inorganic mass, whereas this is positive in this work, giving greater SIA contribution but smaller contribution of the mixed category. Meanwhile, owing to a lack of fugitive dust, the simulated soil dust contribution (1%–2%) by this work is much smaller.

Our regional SA results agree well with conclusions from previous studies summarized in Table S5. The predicted city contribution of Shijiazhuang is larger than those of Xingtai and Handan in January 2013, likely due to a higher emission rate and a larger area of Shijiazhuang. These results are consistent with L. -T. Wang et al. (2014). The seasonality of local anthropogenic PM_{10} in North China shown by Li et al. (2014) with the Nested Air Quality Prediction Model System (NAQPMS) is also well reproduced by the PSAT in this study. Both methods predict extremely small local contributions of PM_{10} in the dust season. Among all $\text{PM}_{2.5}$ species, SO_4^{2-} and NO_3^- tend to have smaller local contributions. SO_4^{2-} and NO_3^- have the smallest local contribution in the warm and cold seasons, respectively, as shown by Ying et al. (2014) and in the present work.

4.2 Changes between 2006 and 2013

4.2.1 Changes in emissions between years

Anthropogenic emissions in China over the past decade have increased because of increased industrialization and urbanization or decreased because of the adoption of emission controls. Control of primary particles and SO₂, mainly from power plants, has been enforced since 2005. NO_x was not controlled until 2010. Figure 5 displays the percentage emission changes (from 2006 to 2013) of relevant species in BTH and sub-regions. Focusing on the total amount of emissions in BTH (see black bars in left most panels in Fig. 5), the emissions of SO₂ decrease by 30%, reflecting the effects of emission control on power plants. The emissions of NO_x increase by 23%, reflecting increased emissions from industry. The emissions of NH₃ decrease by 22%, reflecting decreased emissions from agriculture. Note that there are large uncertainties for this sector. The emissions of EC increase by 10%, reflecting increased emissions from industry partially offset by decreased emissions from transportation. The emissions of POC increase by 7%, largely from an increase of emissions from industry. The emissions of other primary PM_{2.5} decrease by 8%, reflecting emission control on power plants.

Although emission changes of all six species in the three sub regions of Hebei are similar to those of the total for BTH, the emission changes in Beijing and Tianjin exhibit very different behaviors. Owing to stricter control measures, the emissions of all species shown in Fig. 5, including NO_x, EC, and POC, decrease in Beijing. The decline of NO_x emissions is attributable to decreased emissions from transportation and power plants. Decreases in the emissions of EC and POC are attributable to decreased emission from industry, residential and transportation. Decreased emissions of SO₂ and other primary PM_{2.5} in Beijing are of larger magnitudes than those for other sub regions. As shown from the EC and POC panels in Fig. 5, the change in emissions of carbon aerosols in Tianjin is distinct from that in other areas and is driven by changes

in the residential sector. The increase of POC in Tianjin is an order of magnitude greater than that of the total in BTH.

4.2.2 Changes of SA from 2006 to 2013

The emission changes described above combined with different meteorological conditions lead to changes in SA in the BTH region. Figure S2 and 2 shows the $PM_{2.5}$ SA in 2006 and 2013, respectively, and illustrates the dominant role of industry and residential sectors, their increased contributions in most of BTH, and the domain-wide decrease of power-plant related $PM_{2.5}$. Unfortunately, the decrease in power-plant related $PM_{2.5}$ is offset by an increase in $PM_{2.5}$ from the other sectors, resulting in an overall increase of $PM_{2.5}$ over most of the North China Plain. Figures S3a and 3a quantify the changes of contributions to urban $PM_{2.5}$ in BTH from industry (from 41% to 44%), residential (24% to 30%), and power plants (13% to 8%). The remaining panels in Fig. S3 and Fig. 3 show that the seasonal SA changes from 2006 to 2013 generally follow the trends in the annual changes.

Annual $PM_{2.5}$ concentrations in Beijing decreases from $135.3 \mu g m^{-3}$ in 2006 to $109.6 \mu g m^{-3}$ in 2013 (Figs. 6a, b), generally consistent with the trend of $-3.2 \mu g m^{-3} yr^{-1}$ ($-4.3\% yr^{-1}$) during 2005 to 2013 reported by Zhang et al. (2015). This decrease is a combination of slight decreases in the absolute contributions of all sectors (note that, changes in percentage contribution may not be always consistent with the absolute changes, e.g. residential, the sector with the smallest absolute decrease, shows an increase in its percentage contribution from 31% to 37%). Annual $PM_{2.5}$ concentrations in Tianjin, however, increases from 108.7 to $123.2 \mu g m^{-3}$. There is a clear decrease from power plants (from 13% in 2006 to 8% in 2013) and increase from residential (20% in 2006 to 31% in 2013), consistent with the absolute contribution from each anthropogenic source (Fig.

6f). In Hebei, the simulated annual $\text{PM}_{2.5}$ increases from 107.2 to 118.4 $\mu\text{g m}^{-3}$. The power plant contribution decreases from 13% to 8% while the contributions from industry (40% to 44%) and residential (23% to 29%) increase.

The right panels in Figure 6 show the $\text{PM}_{2.5}$ contributions by sector. As discussed above, the simulated industry contribution to $\text{PM}_{2.5}$ declines in Beijing and rises in Tianjin and Hebei. The decrease in Beijing is attributable to a decrease of other primary $\text{PM}_{2.5}$ and SO_4^{2-} . The increase in Hebei is dominated by POC and SO_4^{2-} . Decreased power plant contributions at all three receptors are always dominated by SO_4^{2-} (see that for Tianjin in Figure 6). Increased residential contribution in both Tianjin and Hebei is dominated by POM (Primary Organic Matter, POC multiplied by 1.8) and EC. Decrease of the transportation contribution to $\text{PM}_{2.5}$ in Beijing is dominated by decreased EC and NO_3^- . The decline for agriculture in Beijing is caused by decrease of NH_4^+ (figure not shown), NH_4^+ is the only species considered in this sector. Generally, the SA changes discussed above are consistent with emission variations shown in Section 4.2.1, except that there is no evidence for the increase of residential emissions in Hebei, especially POC. This turns out to be an impact of meteorology, which will be discussed subsequently.

To further investigate the changes in major contributors for each receptor, all category and regional contributors for year 2013 are combined into 7 local categories and 10 regions (a total of 17 source groups). The top 10 of the 17 are displayed in Fig. 7 (left) with their changes due to meteorology and emissions (Fig. 7 right). These top contributors together explain around 90% of total $\text{PM}_{2.5}$ concentrations for all receptors. Local industry and local residential are always the top two contributors, followed by regional inflow from surrounding area. Changes of these contributors together explain around 90% of total changes of $\text{PM}_{2.5}$ concentration between 2006

and 2013.

There are only two notable changes of $PM_{2.5}$ in N. Hebei; local industry and residential. The increase in local industry contribution to $PM_{2.5}$ is caused by a change in emissions, while the increase in local residential is caused by a change in meteorology. Variations of all regional inflows are negligible (less than 2% of $PM_{2.5}$ concentration in E06M06). Similar to N. Hebei, the local residential contribution in Beijing is enhanced by meteorology in 2013. Fortunately, this increase is offset by strict emission controls of sectors in Beijing; the source contributions of local industry, local residential, and local transportation decrease significantly. Again, the role of regional inflow is negligible. As a result, both $PM_{2.5}$ concentration and local contribution percentages decline in Beijing from 2006 to 2013.

In contrast to the situation in N. Hebei and Beijing, Tianjin, C. Hebei, and S. Hebei are generally more vulnerable to the impacts of regional inflows. In Tianjin, the source contributions of local residential are enhanced by both emission change and meteorology with the former being more important. In C. Hebei, the local industry contribution is enhanced because of an emission increase, and contributions of both local industry and local residential are enhanced by meteorology. $PM_{2.5}$ in C. Hebei also benefits from decreasing emission in Beijing, although this benefit is offset by an increase in other contributors. In S. Hebei, significant changes are increased local residential source contribution and inflows from N. Hebei and Shandong due to meteorological changes. The meteorology in 2013 favors the enhanced contribution of local surface emissions, which is also obvious in comparison of local and non-local contributors (Figure S8). In all three receptors of Hebei, the contribution of residential emissions to $PM_{2.5}$ increases, reflecting changes in meteorological, rather than in emissions (see Fig. 5).

5. Conclusions

CAMx/PSATv6.0 has been applied to a Chinese domain to estimate SA of $PM_{2.5}$ in the BTH region. Emission categories and source regions are derived in a single simulation. Driven by meteorological fields from the WRF, simulated results capture temporal and spatial characteristics of $PM_{2.5}$ and its components across the domain reasonably well. The $PM_{2.5}$ concentrations in July 2013 are overpredicted by 70%, which are likely due to uncertainties in MEIC and may be associated with underprediction of 2-m temperature by WRF. This overprediction generally locates in south China and does not notably affect the SA result for BTH. The SA results are generally consistent with conclusions from previous studies.

The industry and residential sectors are found to continue to dominate urban $PM_{2.5}$ in BTH from 2006 to 2013; together they contribute to around 70% of urban $PM_{2.5}$. In winter, residential source contributes the most (~50%), followed by industry (~30%). In other seasons, the industry contribution (~50%) is dominant. Contributions to $PM_{2.5}$ from industry and residential increase slightly from 2006 to 2013, by 3% and 6%, respectively, reflecting increased emissions of NO_x and carbon aerosol from the two sectors. When formulating future control policy in BTH, more attention should be given to the residential sector in winter and to industry year round.

Local emission is the most important contributor at all five receptors (40%–60%), although the role of regional inflow is not negligible. Two of the five receptors (Beijing and N. Hebei) have local contribution always > 50% for both years. $PM_{2.5}$ at these two receptors are generally independent of changes of regional inflow, suggesting that local controls should have a higher priority in improving air quality in Beijing and Northern Hebei than in other receptor areas. $PM_{2.5}$ at the other three receptors is more susceptible to regional inflow. Unfavorable meteorological

conditions in 2013 seem to enhance the regional transport of $PM_{2.5}$ over the North China Plain, which is demonstrated to be the same important as emission increases from industry and/or the residential sectors. Regional joint actions are clearly an important element of a control strategy.

There are several limitations in this study. In PSAT, a fundamental assumption is that PM is only apportioned to the primary precursor for each PM type, implying that nonlinear relationships among all precursors and secondary PM species are not considered. Therefore, although PSAT gives a true SA result by definition, caution should be executed in using PSAT results to support emission control policy, especially for precursors of secondary inorganic particles. Additionally, source partitioning is currently sector-based, which restricts comparability with results from receptor-based methods and does not provide information on policy of inner-sector emission control. As for all chemical transport modeling work, the uncertainties in meteorology, emission inventories, and formulations and parameterizations used in the 3-D model affect the accuracy of SA results. Although the overall performance of the model in simulating total $PM_{2.5}$ is acceptable, the biases in its components may also affect the SA of total $PM_{2.5}$ to some extent, because the SA of $PM_{2.5}$ components may differ from each other. The contribution of regional inflow is sensitive to the predictions of meteorological variables, e.g., wind speed and boundary layer height. Despite these limitations, the SA results presented in this work provide valuable information regarding the sources and their relative contributions for urban $PM_{2.5}$ in the Beijing-Tianjin-Hebei region in 2006 and 2013.

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Table 1. Simulation design

Simulation Index	Meteorology	Emission	PSAT
E06M06	2006	2006	on
E13M13	2013	2013	on
E06M13	2013	2006	on

598

Table 2. Performance evaluation of simulated PM_{2.5}^a

	Month	Samples	Corr	MB	ME	RSMB	NMB	NME	MFB	MFE
2006	1	17 ^b	0.76	17.2	38.1	54.3	20.1%	44.6%	-0.6%	48.9%
	4	17	0.64	-9.0	26.6	33.7	-13.5%	40.0%	-20.2%	50.1%
	7	18	0.77	7.7	21.2	26.7	14.8%	40.8%	-3.3%	46.7%
	10	17	0.66	11.6	35.6	47.7	16.7%	51.1%	-14.4%	57.7%
2013	1	61280	0.48	24.8	76.9	120.0	18.1%	56.3%	8.5%	50.4%
	4	91057	0.46	7	30.1	47.7	12.1%	52.3%	4.8%	49.3%
	7	96504	0.43	27.7	40.2	65.6	71.3%	101.9%	34.7%	67.4%
	10	103734	0.5	-3.1	37.8	63.1	-3.9%	48.7%	-3.2%	49.0%

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^a The units of PM_{2.5} are µg m⁻³.

600

^b Sample number for 2006 is actually site number.

Table 3. Comparison of category contributions with past studies

Species	Receptor	Source	Period	Previous ¹	This Work ²	Reference & Method
PM _{2.5}	Shijiazhuang	Industry	January 2013	36.2	37.0	L. -T. Wang et al. 2014 CMAQ-BFM
		Power Plant		0.4	6.0	
		Residential		38.0	45.0	
		Transportation		4.2	5.0	
		Agriculture		14.5	6.0	
	Xingtai	Industry		34.2	30.0	
		Power Plant		0.3	6.0	
		Residential		40.5	52.0	
		Transportation		1.8	5.0	
		Agriculture		16.8	6.0	
	Handan	Industry		35.8	30.0	
		Power Plant		0.3	6.0	
		Residential		38.1	51.0	
		Transportation		2.9	6.0	
		Agriculture		16.8	6.0	
OC	Beijing	Biomass Burning	Annual 2011	about 50%	78(77)	Cheng et al., 2013 PMF
EC				about 50%	63(58)	
PM _{2.5}	Beijing	Soil dust	Apr2009-Jan2010	15.0	1(2)	Zhang et al., 2013 PMF
		Coal Combustion + industry process		43.0	37(38)	
		Biomass Burning		12.0	23(20)	
		Primary Particles from Traffic		4.0	4(5)	
		SIA		26.0	36(35)	

¹All contributions are in %.

²For this study, contributions in 2013 are always listed and, when necessary, corresponding contributions in 2006 are listed in parentheses.

Figure 1. Map of source regions (numbers in white: 1, Beijing; 2, Tianjin; 3, Northern Hebei; 4, Middle Hebei; 5, Southern Hebei; 6, Liaoning; 7, Inner Mongolia; 8, Shanxi; 9, Henan; 10, Shandong; 11, other areas) and receptors (black circles: BJ-Beijing, TJ-Tianjin; TS-Tangshan; QHD-Qinhuangdao; CD-Chengde; ZJK-Zhangjiakou; BD-Baoding; LF-Langfang; CZ-Cangzhou; SJZ-Shijiazhuang; HS-Hengshui; XT-Xingtai; HD-Handan).

Figure 2. Simulated spatial distributions of total annual mean PM_{2.5} concentrations over the North China Plain in 2013 (a) and contributions from: industry (b), power plants (c), residential (d), transport (e), agriculture (f), and natural sources (g).

Figure 3. Source apportionment of urban PM_{2.5} of BTH for 2013: annual (a) and seasonal (b to e).

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619 Figure 4. Annual regional and categorical SA of urban PM_{2.5} in Beijing, Tian, N. Hebei, C. Hebei,
 620 and S. Hebei for 2013. Colored cells represent percentage contribution of each source region (row)
 621 and each source category (column). White lines represent regional contributions (i.e. sum of each
 622 row), which is marked by top X-axis.

623

624 Figure 5. Change from 2006 to 2013 in BTH and its sub-regions of total emissions and emissions
 625 from industry, power plants, residential, and transportation of (clockwise from top left) SO₂, NO_x,
 626 NH₃, other PM_{2.5}, POC, and EC.

Figure 6. SA of urban $PM_{2.5}$ in Beijing, Tianjin, and Hebei during 2006 (a, d, g) and 2013 (b, e, h). The graphics on the right show comparison of $PM_{2.5}$ attributed to each anthropogenic source category at each receptor (c, f, i). Pie charts to the right of bars show apportionment of the change of $PM_{2.5}$ by chemical category of PM component (gray slice is balance).

Figure 7. (Left) Top 10 contributors of ambient $PM_{2.5}$ in 2013 among local categories and regions, and (right) the changes of these contributors from 2006 to 2013 for each receptor. The numbers represent the percentage contribution to either total $PM_{2.5}$ concentration (left) or total changes (right) explained by the top 10 contributors together for each receptor. Changes are split into impact of meteorology (yellow) and emission (green) change. Changes that exceed $\pm 2\%$ of total $PM_{2.5}$ in 2006 (dashed lines) are identified.