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Source apportionment of $PM_{2.5}$ in top polluted cities in Hebei, China using the CMAQ model

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HIGHLIGHTS

- $PM_{2.5}$ source apportionment is pursued over the top three polluted cities in China (Shijiazhuang, Xingtai, and Handan).
- Regional contributions play a non-negligible role in the three cities.
- Spatial and sectoral contributions to the secondary species in $PM_{2.5}$ are quantified.

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ABSTRACT

Hebei has been recognized as one of the most polluted provinces in China, characterized by extremely high concentrations of fine particulate matter ($PM_{2.5}$) in many of its cities, especially those located in the southern area of the province and highly potentially northward transported to Beijing. Source apportionment of $PM_{2.5}$ is the basis and prerequisite of an effective control strategy. In this study, the Mesoscale Modeling System Generation 5 (MM5) and the Models-3/Community Multiscale Air Quality (CMAQ) modeling system are applied to East Asia and North China at 36- and 12-km horizontal grid resolutions, and the source apportionment of $PM_{2.5}$ in the three top polluted cities in Hebei, i.e., Shijiazhuang, Xingtai, and Handan, is performed using the Brute-Force method. It is concluded that the regional source contributions to $PM_{2.5}$ are 27.9% in Shijiazhuang, 46.6% in Xingtai, and 40.4% in Handan. The major local contributors are industrial, domestic and agricultural sources in all the three cities with the contributions of 39.8%, 15.8%, and 10.6% in Shijiazhuang, 30.5%, 13.6%, and 6.9% in Xingtai, 35.9%, 13.5%, and 6.2% in Handan, respectively. As to the secondary aerosols of sulfate (SO_4^{2-}), nitrate (NO_3^-), and ammonium (NH_4^+) in $PM_{2.5}$, which are important chemical species in $PM_{2.5}$ (about 30–40% in $PM_{2.5}$) and cannot be further apportioned by receptor models, the regional source contributions to the total concentrations of SO_4^{2-}, NO_3^-, and NH_4^+ are 40.9%, 62.0%, and 59.1% in Shijiazhuang, Xingtai, and Handan, respectively. The local industrial, domestic and agricultural contributions to those are 23.7%, 6.6%, and 29.8% in total in Shijiazhuang, 17.5%, 5.0%, and 17.7% in Xingtai, and 20.6%, 4.8%, and 17.8% in Handan, respectively. The regional joint controls of air pollution are more important in Xingtai and Handan than in Shijiazhuang, and the emission controls of agricultural sources need to be further considered in the future policy.

1. Introduction

Haze has been one of the most severe air pollution problems in China and the Beijing–Tianjin–Hebei area is the top polluted region, characterized by high frequencies of haze events and high concentrations of fine particulate matter ($PM_{2.5}$) (MEP, 2014, 2015). In 2012, the Ministry of Environmental Protection (MEP) in China enacted the new National Ambient Air Quality Standards (NAAQS), which sets the limits for $PM_{2.5}$ for the first time of 75 μg m^{-3} and 35 μg m^{-3} for the daily and annual average, respectively (MEP, 2012). Since 2013, the MEP began to report the top ten polluted cities in China every month. According to this report, in 2013, the top five polluted cities are Xingtai, Shijiazhuang, Handan, Tangshan,

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and Baoding (MEP, 2014). In 2014, they are Baoding, Xingtai, Shijiazhuang, Tangshan, and Handan (MEP, 2015). All of them are in Hebei Province and the three neighbor cities in the southern Hebei, Shijiazhuang (the capital of Hebei), Xingtai, and Handan are listed in the top polluted cities in China. The annual average $PM_{2.5}$ concentrations in 2014 are as high as 126 μg m^{-3} in Shijiazhuang, 131 μg m^{-3} in Xingtai, and 116 μg m^{-3} in Handan, according to the annual report by the Department of Environmental Protection in Hebei (Hebei EPD, 2015), which are over three times of the limit in the NAAQS. Shijiazhuang, Xingtai, and Handan have land areas of 1.4×10^4, 1.2×10^4, and 1.2×10^4 km^2, and populations of 9.5, 9.2, and 7.1 million, respectively (HBBS, 2014). Those cities are all industrialized. Their GDPs in 2013 are 486, 306, and 160 billion RMB, ranked 2nd, 3rd, and 7th in the eleven cities in Hebei, respectively (HBBS, 2014). Shijiazhuang city has a large amount of pharmaceutical, chemical, and textile industries. The major industries in Handan are iron and steel, coke, and ceramic production. As to Xingtai, the key industries are building material, coal, and metallurgical industry. Those industries result in a large amount of air pollutants emissions.

In 2013, the State Council of China required MEP to accelerate the $PM_{2.5}$ source apportionment in China to support the policy making for air pollution control. Hebei EPD also required all the 11 cities in Hebei to start $PM_{2.5}$ source apportionment studies in 2014, especially the top polluted cities including Shijiazhuang, Xingtai, and Handan. In the "Guidance for particulate matter source apportionment" published by MEP in 2013, two methods, i.e., source-oriented model and receptor model, are recommended, and the combination of those two methods are encouraged to avoid the shortages of each method (MEP, 2013). Although several receptor models, e.g., the Chemical Mass Balance (CMB) model, the Positive Matrix Factorization (PMF) model, and the Principal Component Analysis (PCA) model, have been reported to be applied in $PM_{2.5}$ source apportionment in Shijiazhuang and Handan (Wang, 2004; Wei et al., 2014a; Meng et al., in press), they all have the problem of not being able to recognize the sources of secondary aerosols in $PM_{2.5}$, which are non-negligible components in $PM_{2.5}$ in the cities of North China (Huang et al., 2014; Wang et al., 2014a; Zheng et al., 2015; Wei et al., 2014a, b). The source-oriented model can fill in this gap. Wang et al. (2014b) applied the Mesoscale Modeling System Generation 5 (MM5) and the Models-3/Community Multiscale Air Quality (CMAQ) modeling system to analyze haze characteristics in the three cities, Shijiazhuang, Xingtai, and Handan, and performed the $PM_{2.5}$ source apportionment using the Brute-Force method (Dunker et al., 1996) for the severely polluted period of January and February 2013. However, it has two shortages to answer the question of $PM_{2.5}$ source of origins. First, in this study those three cities are combined together in the Brute-Force simulations, i.e., the "local source contributions" are actually the total emission contributions of the three cities to the $PM_{2.5}$ concentration in each objective city, and the "local sectoral contributions" are actually the total contributions of each emission sector of the three cities. Those apportionments cannot be used by the city policymakers. Second, it only pursued the simulations of January and February 2013, which cannot be used to proximately estimate an annual average.

This study extends the work in Wang et al. (2014b). First, the source contributions of the real local (emissions within the city) and the regional (emissions outside each city) are evaluated using the MM5–CMAQ system, for the two months of January and July 2013, as well as the source contributions of the major emission sectors in each city (Shijiazhuang, Xingtai, and Handan). Second, the spatial and sectoral source contributions to each major chemical species in $PM_{2.5}$, especially the secondary aerosols of sulfate (SO_4^{2-}), nitrate (NO_3^-), and ammonium (NH_4^+), are analyzed to understand their source of origins and to provide necessary information to further apportion the "secondary aerosols" recognized by the receptor models, given the status that neither method can produce better source apportionment results due to their methodology limitations and uncertainties existing in the available input data (Tang et al., 2006; MEP, 2013; Zhang et al., 2015). Since January 2013 is reported as the most polluted month in the North China in the past 60 years (Lu et al., 2013), many studies focusing on this month have been reported in recent two years (Wang et al., 2014a, 2014c, 2014d; Quan et al., 2014; Huang et al., 2014; Che et al., 2014; Zheng et al., 2015). Those studies focus on the atmospheric observations during the episode (Quan et al., 2014; Che et al., 2014; Wang et al., 2014d; Huang et al., 2014), or shortage and improvement of present air quality models (Wang et al., 2014a; Zheng et al., 2015), or the long-range transport of air pollutants over the North China (Wang et al., 2014c). However, it still lacks the information on source apportionment for Hebei cities, which we believe is urgently required even some incompleteness exists in the present air quality models.

This paper is organized as follows: Section 2 describes the model configuration, evaluation protocols, and emission reduction scenarios. Section 3 presents the simulation results and evaluation of model performance. Section 4 examines the source contributions, including regional and local sectors, to $PM_{2.5}$ concentrations and its major chemical compositions in the three cities. Section 5 summarizes the major findings of this work.

2. Methodology

2.1. Model configurations and input

The modeling domains and configurations in this study are consistent with those in Wang et al. (2012, 2014b). The detailed descriptions can be found in Wang et al. (2012, 2014b). A brief description is provided as below.

As shown in Fig. 1, MM5 and CMAQ simulations are performed over two nested domains. Domain 1 covers East Asia with a grid resolution of 36 × 36 km and Domain 2 is over an area in northeastern China at a 12 × 12 km grid resolution that encompasses Beijing, Tianjin, and the four provinces including Hebei, Henan, Shandong, and Shanxi. January and July 2013 are simulated to represent the winter and summer of the year. A spin-up period of 5 days is used to minimize the influence of the initial conditions. MM5 version 3.7 and CMAQ version 4.7.1 are applied in this study. The vertical structure for MM5 is 23 sigma levels from surface to tropopause (~100 mb) and those for CMAQ are 14 levels of 1.000, 0.995, 0.988, 0.980, 0.970, 0.956, 0.938, 0.893, 0.839, 0.777, 0.702, 0.582, 0.400, 0.200, and 0.000. Two-way nesting is used for MM5 and one-way is used for CMAQ.

The major physics options in MM5 are: the Kain–Fritsch cumulus scheme (Kain and Fritsch, 1993), the high-resolution Blackadar PBL scheme (Zhang and Anthes, 1982), the mixed phase (Reisner 1) explicit moisture scheme (Reisner et al., 1998), the cloud atmospheric radiation scheme for both longwave and shortwave radiation (Dudhia, 1993), the force/restore (Blackadar) surface scheme (Blackadar, 1976; Deardorff, 1978), and the four-dimensional data assimilation (FDDA) using analysis nudging for wind, temperature, and the water vapor mixing ratio both in and above planetary boundary layer (PBL). The major options in CMAQ are: SAPRC-99 gas phase chemical mechanism (Carter, 1990, 2000) with aqueous and aerosol extensions, the AERO5 aerosol phase model (Binkowski and Shankar, 1995), the updated aqueous-phase chemical mechanism of the Regional Acid Deposition Model (RADM) model (Chang et al., 1987; Walcek and Taylor, 1986).

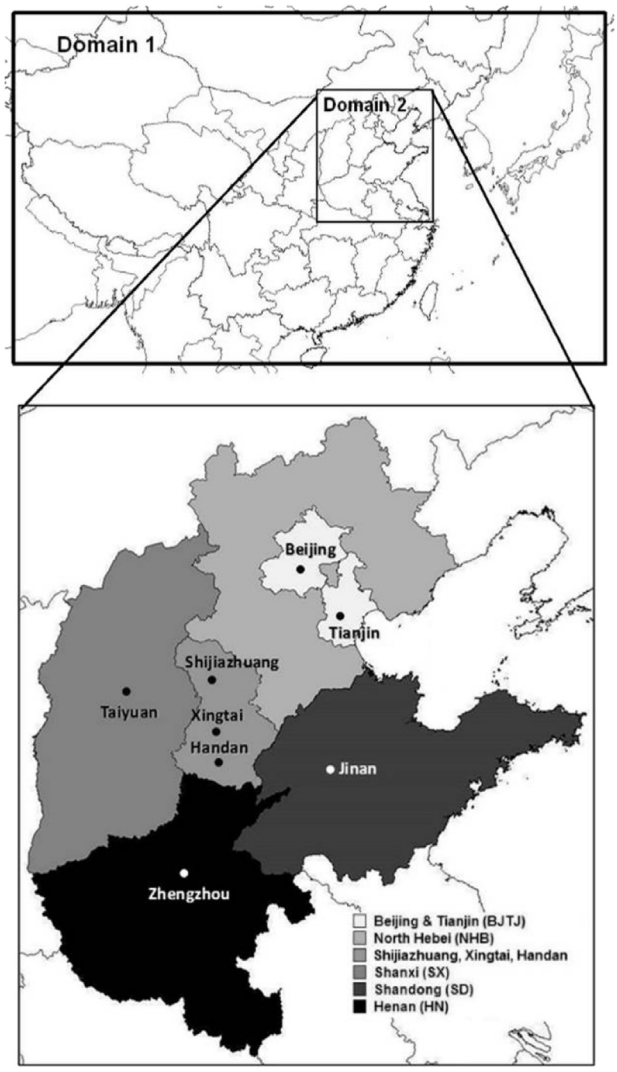


Fig. 1. CMAQ modeling domains at a horizontal grid resolution of 36-km over East Asia (Domain 1 with 164 × 97 cells) and 12-km over an area in northern China (Domain 2 with 93 × 111 cells).

The data sources for MM5 simulation are: the US Geological Survey terrain and land use data (ftp://ftp.ucar.edu/mesouser/MM5V3/TERRAIN_DATA/), the National Center for Environmental Prediction (NCEP) Final (FNL) Operational Global Analysis data sets for initial and boundary conditions, the NCEP Automated Data Processing (ADP) surface and upper air data for the FDDA. The input data for CMAQ simulations are: default clean profile for the initial (ICON) and boundary (BCON) chemical conditions for Domain 1, total ozone column data from the Ozone Measurement Instrument (OMI) (http://toms.gsfc.nasa.gov/ozone/ozone_v8.html) on the Aura satellite for the photolysis rates processor (JPROC), and the Multi-resolution Emission Inventory for China (MEIC) (He, 2012). The MEIC inventory provides all anthropogenic emissions of eight species, including sulfur dioxide (SO_2), nitrogen oxides (NO_x), carbon monoxide (CO), non-methane volatile organic compounds (NMVOCs), ammonia (NH_3), carbon dioxide (CO_2), coarse particulate matter (PM_{10}), and $PM_{2.5}$ in China for the years 1990–2010. The MEIC inventory has been demonstrated to provide reasonable estimations of the total emissions from cities, but uncertainties may exist in the spatial allocations of those emissions into fine grid resolutions (Wang et al., 2014b, 2015).

2.2. Evaluation database and protocols

The observational data used for model evaluation in this study are similar to those used in Wang et al. (2012, 2014b), which is summarized in Table 1. The National Climate Data Center (NCDC) integrated surface database are used to evaluate the meteorological predictions for the five major parameters: temperature at 2-m (T2), water vapor mixing ratio at 2-m (Q2), wind speed at 10-m (WS10), wind direction at 10-m (WD10), and daily precipitation. Data at every 1- or 3-h (most at 3-h) at a total of 371 sites within our domains in January and July 2013 are used in the evaluation.

As indicated in Wang et al. (2014b), two datasets of chemical concentrations are used. One is the real-time database from the China National Environmental Monitoring Center (CNEMC) of the real-time concentrations at 496 national monitoring stations in 74 major cities in China. The hourly $PM_{2.5}$ and PM_{10} concentrations are evaluated in this study. Note these data are unavailable for the period of January 1–13, the reasons for which are explained in Wang et al. (2014b). The second source is the observations at a site located in the Hebei University of Engineering (refer to as HEBEU) measured by the lead author's group since July 2012 (Wei et al., 2014a, b). The $PM_{2.5}$ chemical compositions measured at this site are applied in model evaluation. More detailed descriptions of those two datasets can be found in Wang et al. (2014b) and Wei et al. (2014a, b).

The meteorological evaluation is performed of those statistics: the Mean Bias (MB), the Root Mean Square Error (RMSE), the Normalized Mean Bias (NMB), and the Normalized Mean Error (NME), whose definitions can be found in Zhang et al. (2006). The criteria proposed by Emery et al. (2001) and Tesche et al. (2001), that MB $\leq \pm 0.5$ K for T2, MB $\leq \pm 1$ g kg^{-1} for Q2, MB $\leq \pm 0.5$ m s^{-1} for WS10, and MB $\leq \pm 10^{\circ}$ for WD10 for a satisfactory predictions, are used to judge meteorological performance.

The chemical species evaluation is performed in terms of the overall statistics of NMB, NME, the Mean Fractional Bias (MFB) and the Mean Fractional Error (MFE), following the guidance by U. S. Environmental Protection Agency (U. S. EPA, 2007). The model performance criteria are set on MFB $\leq \pm 60\%$ and MFE $\leq 75\%$ for particulate matter (PM) modeling followed the suggestions proposed by Boylan and Russell (2006). In addition, the concentrations of the major chemical species of $PM_{2.5}$, including SO_4^{2-}, NO_3^-, NH_4^+, elementary carbon (EC) and organic carbon (OC) observed at the HEBEU site are compared with the model predictions to evaluate the model capability of predicting the chemical compositions of $PM_{2.5}$.

2.3. Emission reduction scenarios and simulation design

Followed the study of Wang et al. (2014b), the Brute Force method (BFM) is applied to evaluate the source contributions of each region and emission sector (Dunker et al., 1996). The detailed explanation on why BFM method are chosen instead of other emission sensitivity methods such as the Decoupled Direct Method in 3 Dimensions (DDM-3D) (Yang et al., 1997; Dunker et al., 2002; Cohan et al., 2005; Napelenok et al., 2006) or source apportionment methods such as the Particulate Precursor Tagging Methodology (PPTM) (U. S. EPA, 2009) are presented in Wang et al. (2014b). In brief, the BFM method can simulate the non-linear relationship among PM and its precursors under large emission changes, and it has also been proved to be an effective way in many studies in recent years (Marmur et al., 2005; Streets et al., 2007; Chen et al., 2007; Wang et al., 2008; Fu et al., 2009; Zhou et al., 2010; Xing et al., 2011; Burr and Zhang, 2011; Zhang and Wu, 2013).

Nine source regions are selected in the spatial contribution analysis (See Fig. 1), which includes the three objective cities of

Table 1
Observational datasets for model evaluation used in this study.

Dataset[a]	Data type	Variable[b]	Data frequency	Site number	Time period	Data sources
NCDC	Meteorology	T2, Q2, WD10, WS10, Precipitation	Every 1 or 3-h Daily	371	Jan. and Jul., 2013	http://www.ncdc.noaa.gov/data-access/quick-links#ghcn
CNEMC	Air quality	$PM_{2.5}$	Hourly	496	Jan. 13–31 Jul., 2013	http://113.108.142.147:20035/emcpublish
HEBEU	Air quality	$PM_{2.5}$	Hourly	1	Jan. and Jul., 2013	Wei et al., 2014a,b

[a] NCDC – the National Climate Data Center integrated surface database. CNEMC – the real-time database from China National Environmental Monitoring Center. HEBEU – observations at the site of Hebei University of Engineering.
[b] T2 – temperature at 2-m; Q2 – water vapor mixing ratio at 2-m; WS10 – wind speed at 10-m; WD10 – and wind direction at 10-m.

Shijiazhuang, Xingtai, and Handan, five regions of northern Hebei (NHB, all the other cities in Hebei besides the three objective cities), Shanxi province (SX), Shandong province (SD), Henan province (HN), and a two-city cluster region covering Beijing and Tianjin (BJTJ), and all the other regions in Domain 1 excluding the above eight source regions (OTH). In the sectoral contribution analysis, five major emission sectors are considered including power plants (PO), industrial (IN), domestic (DO), transportation (TR), and agriculture (AG). Note in the NHB area, the most northern cities such as Zhangjiakou, Chengde, and Qinhuangdao, are far from the three objective cities and their contributions might be overwhelmed by the contributions from the cities in the middle Hebei and even in Beijing and Tianjin. Therefore, if considering the trans-boundary transport of air pollutants between Hebei cities, the contributions from NHB cannot be directly used to judge the contributions from those most northern cities in Hebei. More simulation and assessment will be needed for those cities.

Firstly, the source contributions of each source region to the three objective cities are calculated by zeroing out all anthropogenic emissions in each region (referred to as AL). Then the contributions of each emission sector in local, i.e., each of the three cities of Shijiazhuang, Xingtai, and Handan, are evaluated by zeroing out their emissions one by one. Therefore, a total of twenty-five 2-month simulations, including 24 emission zero-out scenarios (=9 source regions + 3 cities × 5 sectors) plus one baseline, are performed in this study.

3. Evaluation of model performance

3.1. Overall statistics

The evaluation of model performance for the month of January 2013 has been extensively discussed in Wang et al. (2014b). Here we focus on July 2013 but the results of January are presented as well for a comparison. Table 2 gives the overall statistics of meteorological predictions for Domain 1 at 36 km resolution and for Domain 2 at both 36 and 12 km resolutions. Note the MB, RMSE, NMB and NME for wind direction in January are revised, by using vector treatment, e.g., the difference of 1° and 359° is 2 instead to 358°, which had not been done in Wang et al. (2014b) (See Table 2).

Comparing with in January, the model predictions for T2 in July agrees better with the observations, with the MBs of −0.4 °C over Domain 1 and 0.3 °C and 0.4 °C over Domain 2 at 36 and 12 km resolutions, respectively, which are all within the criteria of MB ≤ ±0.5 K by Emery et al. (2001) and Tesche et al. (2001). Q2 are overpredicted in July with the MBs of 0.0008 kg kg^{-1} for Domain 1 and 0.0016 kg kg^{-1} for Domain 2 at both resolutions, which are higher than those for January. It can be seen that the predictions in January for all the domains and in July for Domain 1 meet the criteria of MB ≤ ±1 g kg^{-1}, but the predictions over Domain 2 are over this criteria. However, the NMBs and NMEs in July are notably smaller than in January (e.g., 15.8% vs. 5.9% for NMBs for Domain 1; −12.6% vs. 10.3% for NMBs for Domain 2 at 12 km resolution).

The model overpredict the wind speed in both two months, with higher biases in July of the MBs of 0.7 m s^{-1} for Domain 1 and 0.7, 0.9 m s^{-1} for Domain 2 at 36-, 12-km resolutions, respectively. Those numbers are over the criteria of MBs ≤ ±0.5 m s^{-1} (Emery et al., 2001), therefore, it should be noted that the model may underpredict the pollutants concentrations due to the higher predicted wind. The predicted wind directions agree relatively well with the observations, with the MBs of −4.5° for Domain 1, and 4.4 and 3.0° for Domain 2 at 36 and 12 km resolutions, respectively, in July. They are all within the range of the criteria of MBs ≤ ±10° (Emery et al., 2001). The model underestimates the precipitation in both months, which is more obvious in July with the MBs of −1.7 mm for Domain 1 and −4.7 mm and −5.2 mm for Domain 2 at 36- and 12-km resolutions, respectively. The NMBs are −31.6% and −17.5% for January and July over Domain 1, and −44.2% and −38.0% over Domain 2 at 12 km resolution, respectively. The underestimation of precipitation may result in a higher PM predictions due to less amount of washing-out atmospheric particles.

In general, the model predictions of temperature and wind directions agree better with the observations in July than in January, but the water vapor mixing ratio and wind speed are on the contrary. Wind speed is overestimated in both months, which may result in an underprediction in pollutants concentrations. At the same time, the underprediction of precipitation may lead to an overestimation in pollutants concentrations.

Table 3 presents the performance statistics of $PM_{2.5}$ and PM_{10} concentrations over Domain 1 at 36-km grid resolution and Domain 2 at 36- and 12-km resolutions. The model overall underpredicts the $PM_{2.5}$ concentrations in Domain 1, with the NMBs of −11.0% and −7.9% for January and February, respectively. As for PM_{10}, the underprediction is more obvious with the NMBs of −32.0% and −37.6% for the two months, respectively. The reasons for this underprediction, as discussed in Wang et al. (2014b), may include the lack of dust emission in this simulation, the biases in meteorological predictions (e.g., overpredictions of wind speed), and the uncertainties in the emissions due to different base year of the emission inventory and the simulation. As for Domain 2, the model overpredicts the $PM_{2.5}$ and PM_{10} concentrations at 12-km grid resolution but underpredicts at 36-km resolution. In general, the performance on $PM_{2.5}$ at 36-km grid resolution (NMBs of −3.8% for January and −0.8% for July) is better than that of 12 km resolution (NMBs of 12.2% and 18.8%, respectively). This indicates the problem that the MEIC inventory may give a reasonable estimation of the total emissions of each city, but uncertainties exist in the distribution of the total emissions into the fine grid resolutions, as thoroughly discussed in Wang et al. (2014b). Different from $PM_{2.5}$, PM_{10} concentrations are overall underpredicted over Domain 2, no matter at 36 or 12 km grid resolution. The larger negative biases in PM_{10} predictions can be attributed to the lack of dust emissions, which contribute a large fractions of coarse particles. Due to the compromise of the uncertainties, the PM_{10} predictions over

Table 2
Overall statistics of meteorological predictions over Domain 1 at 36 km resolution and Domain 2 at 36 and 12 km resolution.

Variables	36-km (Domain 1)						36-km (Domain 2)						12-km (Domain 2)					
	Obs.	Sim.	MB	RMSE	NMB	NME	Obs.	Sim.	MB	RMSE	NMB	NME	Obs.	Sim.	MB	RMSE	NMB	NME
Jan.																		
T2 (°C)	−2.2	−3.6	−1.5	3.9	−69.4	136.0	−3.9	−5.1	−1.2	2.9	−30.1	57.3	−3.9	−5.0	−1.1	2.9	−27.8	57.3
Q2 (kg kg^{-1})	0.0026	0.0030	0.0004	0.0009	15.8	23.8	0.0020	0.0023	0.0003	0.0005	−12.6	19.3	0.0020	0.0023	0.0003	0.0006	−12.6	27.9
WSPD10 (m s^{-1})	2.3	2.9	0.6	2.1	27.4	69.8	2.3	2.8	0.5	0.8	23.2	60.4	2.3	2.9	0.6	1.9	26.7	62.0
WDIR10 (degree)	196.6	186.6	5.6	74.2	2.9	28.7	198.7	205.9	4.4	65.3	2.2	24.0	198.7	198.8	4.1	63.6	2.0	23.1
Precipitation (mm)	1.3	0.9	−0.4	2.4	−31.6	88.8	2.2	1.3	−1.0	2.9	−43.9	62.7	2.2	1.2	−1.0	3.0	−44.2	62.7
Jul.																		
T2 (°C)	24.3	23.9	−0.4	3.1	−1.5	9.3	26.4	26.7	0.3	2.2	1.0	6.3	26.4	26.9	0.4	2.3	1.6	6.5
Q2 (kg kg^{-1})	0.0131	0.0138	0.0008	0.0026	5.9	14.7	0.0160	0.0176	0.0016	0.0028	10.0	13.5	0.0160	0.0177	0.0016	0.0028	10.3	13.5
WSPD10 (m s^{-1})	2.4	3.2	0.7	2.2	30.2	70.3	2.4	3.1	0.7	1.9	31.5	60.9	2.4	3.2	0.9	1.9	36.0	63.6
WDIR10 (degree)	186.1	181.6	−4.5	71.7	−2.4	28.9	187.2	181.2	4.4	63.9	2.3	24.6	187.2	179.3	3.0	63.0	1.6	24.2
Precipitation (mm)	9.6	7.9	−1.7	16.5	−17.5	93.0	13.6	8.9	−4.7	20.1	−34.7	85.9	13.6	8.4	−5.2	21.6	−38.0	91.0

Domain 2 are relatively better at 12-km grid resolution (NMBs of −13.6% for January and −18.5% for July) than those at 36-km resolution (NMBs of −28.9% and −33.7%, respectively). Nevertheless, all $PM_{2.5}$ predictions meet the criteria of MFB ≤ ±60% and MFE ≤ 75% proposed by Boylan and Russell (2006). As for PM_{10}, only the MFE over Domain 1 in July (82.8%) is slightly higher than the criteria (75%), as all other statistics indicate an acceptable model performance.

3.2. $PM_{2.5}$ chemical composition at HEBEU site

Fig. 2 gives a comparison of predicted (from Domain 2 at a 12 km grid resolution) and observed $PM_{2.5}$ chemical compositions at the HEBEU site. In January, the predicted and simulated average $PM_{2.5}$ concentrations are 213.0 and 224.8 μg m^{-3}, respectively, with the NMB of 5.5%. The model generally reproduces the magnitude and temporal variation of $PM_{2.5}$ concentrations, although it underpredicts the extremely severe polluted episode during January 10–13, as discussed in Wang et al. (2014b). One of the major causes, despite the lack of dust emissions in this study, is the model's weakness in the treatment of the heterogeneous chemistry of gaseous precursors on the surface of particles, e.g., the heterogeneous transformation of SO_2 to sulfate particles, which has been extensively discussed in Wang et al. (2014a) and Zheng et al. (2015). The $PM_{2.5}$ concentrations during January 21–29 are overpredicted by the model, which compromise the above underpredictions to some extent. The predicted average concentrations of SO_4^{2-}, NO_3^-, NH_4^+, OC, and EC are 33.5, 31.1, 21.4, 46.6, and 22.6 μg m^{-3}, respectively, as the observed concentrations are 36.7, 23.4, 24.1, 34.1, and 15.1 μg m^{-3}, respectively. The biases of NO_3^-, OC, and EC are relatively larger with MBs of 7.7, 12.6 and 7.5 μg m^{-3}, respectively.

In July, the model generally underpredicts the $PM_{2.5}$ concentrations (86.1 vs. 72.3 μg m^{-3}), which may attribute to several reasons including the lack of dust emissions, uncertainties in the secondary aerosol chemistry, and the biases in meteorological predictions. The model generally captures the temporal variation of the concentrations of $PM_{2.5}$ and its major components. The average observed and predicted concentrations of SO_4^{2-}, NO_3^-, NH_4^+, OC, and EC are 23.5 vs. 15.9, 11.7 vs. 11.4, 8.6 vs. 9.3, 6.0 vs. 7.8, and 2.6 vs. 5.1, respectively. SO_4^{2-} concentrations are underpredicted by 7.6 μg m^{-3}, as discussed above, that may attribute to the model's weakness in reproduce the atmospheric chemical processes of its precursors. EC and OC are overpredicted by 1.8 and 2.5 μg m^{-3}, respectively. In general, the model produces acceptable predictions on the $PM_{2.5}$ concentrations and its major chemical compositions over Northern China and can be used in scenario analysis.

4. Results and discussions

4.1. Regional source contributions

In this paper, the regional source contributions refer to the contributions from the emissions outside the objective city, and the local source contributions refer to the contributions from the anthropogenic emissions source within the objective city. Given the multiple emission reduction scenarios conducted in this study, we can use two methods to estimate the regional source contributions to each city. One is by zeroing out the all the anthropogenic emissions in the objective city, then the predicted concentrations can be seen as the regional source contributions, as expressed in the equation (1). The other is using the base case predictions minus the local source contributions. The local source contributions are calculated by adding up the contributions of each local emission sector, which is estimated by zeroing out the emissions from each

Table 3
Overall statistics of $PM_{2.5}$ and PM_{10} predictions over Domain 1 at 36 km resolution and Domain 2 at 36 and 12 km resolution.

		Jan.			Jul.		
		36-km (Domain 1)	36-km (Domain 2)	12-km (Domain 2)	36-km (Domain 1)	36-km (Domain 2)	12-km (Domain 2)
$PM_{2.5}$	Obs. (μg m^{-3})	132.5	156.8	156.8	35.2	50.5	50.5
	Sim. (μg m^{-3})	117.9	150.8	175.9	32.4	50.1	60.0
	MB (μg m^{-3})	−14.6	−5.9	19.1	−2.8	−0.4	9.5
	NMB (%)	−11.0	−3.8	12.2	−7.9	−0.8	18.8
	NME (%)	51.3	45.2	54.2	66.0	62.9	79.9
	MFB (%)	−19.5	−4.3	8.7	−28.4	−2.7	3.0
	MFE (%)	58.3	46.8	51.1	69.4	61.4	66.6
PM_{10}	Obs. (μg m^{-3})	189.9	232.5	232.5	59.7	86.7	86.7
	Sim. (μg m^{-3})	129.0	165.4	200.8	37.3	57.5	70.7
	MB (μg m^{-3})	−60.8	−67.1	−31.6	−22.4	−29.2	−16.0
	NMB (%)	−32.0	−28.9	−13.6	−37.6	−33.7	−18.5
	NME (%)	51.0	45.7	49.5	64.1	57.6	67.7
	MFB (%)	−43.3	−28.3	−16.0	−59.0	−39.0	−31.2
	MFE (%)	65.2	53.3	52.7	82.8	69.3	73.0

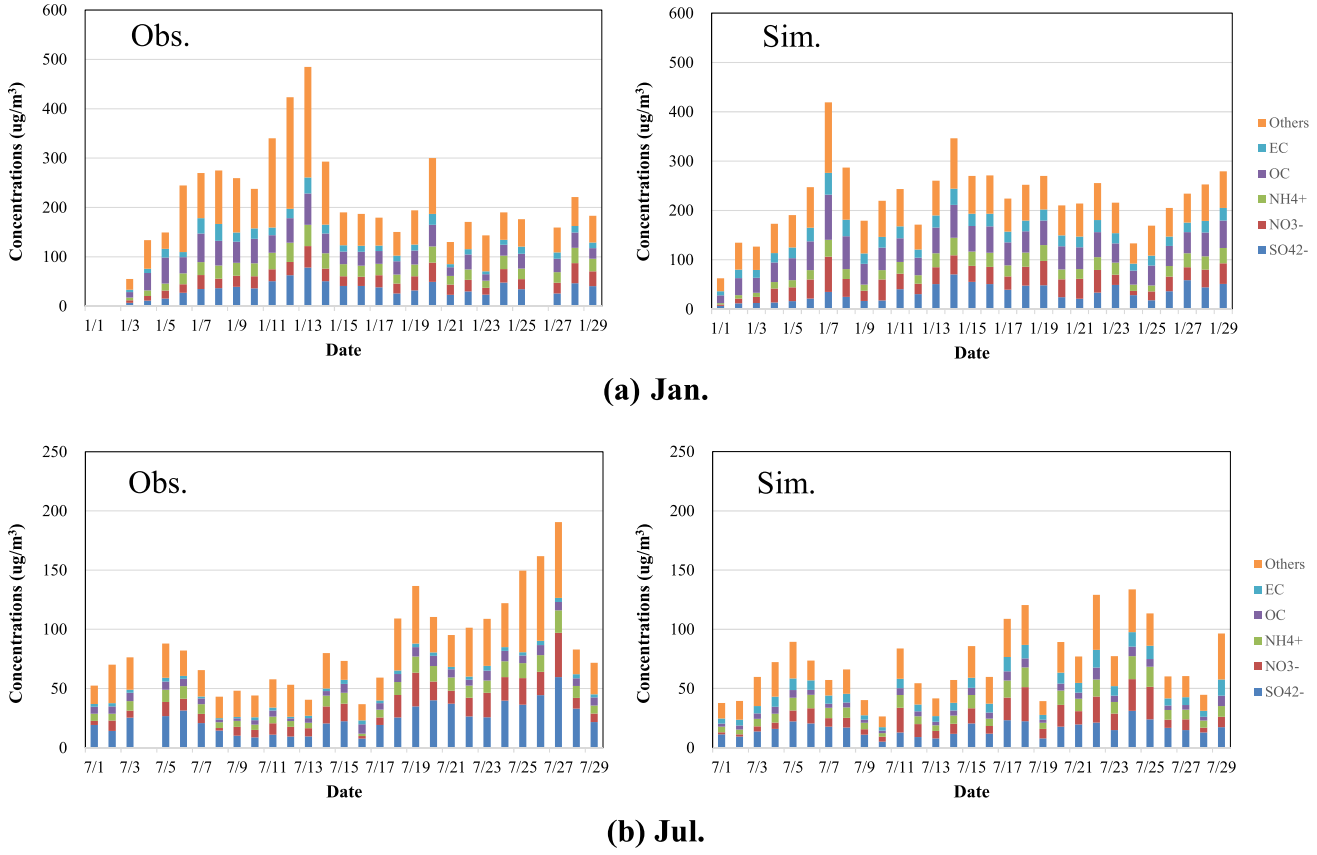


Fig. 2. Comparison of observed and simulated $PM_{2.5}$ chemical compositions at HEBEU site.

sector and then calculating their differences to the base case predictions, as shown in equation (2).

$$C_{R,i} = C_{Base} - C_{i-0} \quad (1)$$

$$C_{R,i} = C_{Base} - \sum_{k=1}^{n}(C_{Base} - C_{i,k-0}) \quad (2)$$

where $C_{R,i}$ is the regional source contributions to city i; C_{Base} is the base case predictions; C_{i-0} is the predictions by the scenario of zeroing out all the anthropogenic emissions in the city i; $C_{i,k-0}$ is the predictions by the scenario of zeroing out the emissions from the sector k in city i; n is the number of the sectors, which is 5 in this study (PO, IN, DO, TR, and AG). All C can be either concentrations or the percentage to the base case concentrations).

Table 4 presents the regional source contributions to the urban average $PM_{2.5}$ concentrations in Shijiazhuang, Xingtai and Handan city, calculated by the above two equations. It can be seen that the two methods give quite similar estimations, e.g., the regional contributions in January in Shijiazhuang are both 33.0% by the equations (1) and (2), and in summer, the difference between the two methods is 2.4% (23.9% vs. 21.5%) for Shijiazhuang, which may partially attribute to the more active atmospheric chemistry

Table 4
The average regional source contributions (%) to $PM_{2.5}$ concentrations in Shijiazhuang, Xingtai, and Handan.

		Jan.	Jul.	Average
Shijiazhuang	Equation (1)	33.0	23.9	28.4
	Equation (2)	33.0	21.8	27.4
	Average	33.0	22.9	27.9
Xingtai	Equation (1)	51.1	42.9	47.0
	Equation (2)	51.2	41.4	46.3
	Average	51.1	42.1	46.6
Handan	Equation (1)	48.1	33.9	41.0
	Equation (2)	47.6	32.0	39.8
	Average	47.9	32.9	40.4

between the aerosol and its gaseous precursors that induces more obvious non-linear relationship between the emissions and atmospheric concentrations. Nevertheless, the average regional contributions in January and July are 33.0% and 22.9% in Shijiazhuang, 51.1% and 42.1% in Xingtai, and 47.9% and 32.9% in Handan, respectively. The annual average, as an approximate estimation by averaging the two months, is 27.9% in Shijiazhuang, 46.6% in Xingtai, and 40.4% in Handan. The regional contribution in Shijiazhuang is the smallest out of the three cities, which may attribute to its more stable atmosphere and lower PBL, according to Wang et al. (2014b). This result is consistent with the source apportionment results published by the Hebei EPD (23–30% of regional contributions, http://www.hebhb.gov.cn/hjzw/hbhbzxd/dq/201409/t20140901_43629.html). The regional source contribution of Xingtai is as high as 46.6%, which can be attributed to its location (between the two cities with large emission density, e.g., Shijiazhuang and Handan rank 2nd and 3rd in GDP in Hebei, and Xingtai ranks 7th (HBBS, 2014)), smaller land area, and different meteorological conditions comparing to Shijiazhuang, i.e., the atmosphere in Xingtai are not as stable as in Shijiazhuang (Wang et al., 2014b), and the prevailing southeast or northwest wind may blow in the pollutants from either Handan or Shijiazhuang. Averagely, the regional contribution to $PM_{2.5}$ in Handan is 40.4%, which is also a significant number that should be considered in future air pollution control in Handan. One important point that should be noted is that overestimation may exist in the above regional contribution analysis, due to the lack of dust emissions, which contribute more from the local than from the regional transport. According to the source apportionment results published by Hebei EPD (http://www.pm25.com/news/496.html) by receptor models, the contributions of the local dust emissions to $PM_{2.5}$ in Shijiazhuang and Handan, out of the total local sources, are 22.5% and 18.4%, respectively (HEBEU, 2015; http://www.pm25.com/news/496.html). Therefore, we can roughly estimate the underestimation of the regional contributions due to the lack of local dust emissions, which are 4.8% and 4.8% for Shijiazhuang and Handan, respectively (e.g., for Shijiazhuang, the revised regional contribution is $27.9\%/((1-27.9\%)/(1-22.5\%)+27.9\%)=23.1\%$). As for Xingtai, the underestimation might be in the range of 5.0–6.2%, if we consider its larger regional contributions (46.6%) and suppose its local dust contribution in the range of those for Shijiazhuang and Handan.

4.2. Local sector source contributions

The local sector contributions in each city are calculated by the zeroing out the emissions from each sector in each season. The results are summarized in Table 5, including the average contributions and the percentage of each sector contribution within the local source contributions. It can be seen that for all the three cities, the DO contributions are much higher in January than in July, due to the winter heating in those cities. Other sectors' contributions are on the contrary. Generally, the IN contributions in each city are significantly higher in July than in January (51.3% vs. 28.3% for Shijiazhuang, 39.6% vs. 21.3% for Xingtai, and 24.2% vs. 47.6% for Handan), as well as the contributions from PO (1.8% vs. 0.7% for Shijiazhuang, 1.1% vs. 0.4% for Xingtai, and 1.2 vs. 0.6% for Handan), although their absolute values are quite low. The contributions from TR are not significant, as on average 5.1% in Shijiazhuang, 2.0% for Xingtai, and 3.8% for Handan. It should be noted that the CMAQ model tends to underestimate the contributions from mobile sources (Wang et al., 2014b). In the official report on $PM_{2.5}$ source apportionment in Shijiazhuang (http://www.hebhb.gov.cn/hjzw/hbhbzxd/dq/201409/t20140901_43629.html), the contribution of mobile sources in Shijiazhuang is 15.0% (vs. 7.0% in this study) within the local source contributions. The AG contributions in Shijiazhuang are 8.9% and 12.4% in January and July, respectively, revealing that the NH_3 emissions from agricultural activities play a non-negligible role in $PM_{2.5}$ formation and should be considered in future emission control plan. In Xingtai and Handan, the agricultural source contributions are 4.8% and 4.6% for January, and 9.0% and 7.8% for July, respectively. In generally, within the local sources, the major contributors are IN, DO and AG for all the three cities. Their contributions are 39.8%, 15.8%, and 10.6% in Shijiazhuang, 30.5%, 13.6%, and 6.9% in Xingtai, 35.9%, 13.5%, and 6.2% for Handan, respectively. Of the total local source contributions, those fractions are 54.9%, 21.8%, and 14.7% in Shijiazhuang, 56.8%, 25.3%, and 12.9% for Xingtai, and 59.6%, 22.3%, and 10.3% for Handan, respectively. It should be noted that the total local source contributions might be underestimated because of the lack of the dust emissions, which contribute more in local than in regional, as mentioned above. Another point is, according to the report on the state of the environment in China, in 2014 the $PM_{2.5}$ concentration over Beijing–Tianjin–Hebei area is reduced by 12.3% on average comparing to in 2013 (MEP, 2015). The $PM_{2.5}$ concentrations in Shijiazhuang and Handan are reduced by 19.5% and 16.6% respectively (MEP, 2015; HEBEU, 2015). It may imply that the local source contributions in those two cities might slightly decrease in 2014 due to the smaller decrease on regional average. It also indicates that the control of industrial emissions and reduction in coal consumption are effective measurements in those cities in the recent two years.

Given the high frequencies of the occurrence of extremely severe pollution episodes and high concentrations of $PM_{2.5}$, it may be important to understand the possible variations of sector contributions under different pollution level, besides their average contributions. Fig. 3 presents the scatterplots of the baseline hourly $PM_{2.5}$ concentrations vs. the local sector source contributions for the three cities of Shijiazhuang, Xingtai, and Handan. If we focus on the three major contribution sectors, IN, DO and AG, it is interesting that the IN and DO contributions in January are relatively lower when the $PM_{2.5}$ concentrations are in the middle range, i.e., 200–400 $\mu g\ m^{-3}$, as the average percentages are 34.2%, 25.6%,

Table 5
The average source contributions (%) to $PM_{2.5}$ concentrations in Shijiazhuang, Xingtai, and Handan by each local emission sector.

		PO	IN	DO	TR	AG	Sum
Shijiazhuang	Jan.	0.7	28.3	24.8	4.3	8.9	67.0
	Jul.	1.8	51.3	6.9	5.9	12.4	78.2
	Average	1.2	39.8	15.8	5.1	10.6	72.6
	% in local[a]	1.7	54.9	21.8	7.0	14.7	100.0
Xingtai	Jan.	0.4	21.3	20.8	1.4	4.8	48.7
	Jul.	1.1	39.6	6.4	2.5	9.0	58.6
	Average	0.8	30.5	13.6	2.0	6.9	53.7
	% in local[a]	1.4	56.8	25.3	3.6	12.9	100.0
Handan	Jan.	0.6	24.2	20.2	2.8	4.6	52.4
	Jul.	1.2	47.6	6.7	4.7	7.8	68.0
	Average	0.9	35.9	13.5	3.8	6.2	60.2
	% in local[a]	1.5	59.6	22.3	6.2	10.3	100.0

[a] Percent of the sector source contributions in the total local source contributions.

34.4% for IN when the $PM_{2.5}$ concentrations are in the range of $\leq$200, 200–400, and >400 μg m^{-3}, respectively, and for DO those are 26.9%. 23.6% and 27.7%, respectively. The contributions of AG are on the contrary that the percentages are 5.8%, 9.8%, and 7.5% for the ranges of $\leq$200, 200–400, and >400 μg m^{-3}, respectively. It indicates that when extremely high $PM_{2.5}$ concentrations appear (e.g., higher than 400 μg m^{-3} on hourly average), NH_3 might be not as so important in the process of secondary $PM_{2.5}$ formation as when $PM_{2.5}$ concentrations are in the middle range. This is consistent with several recent studies on this haze episode over North China in January 2013 (Wang et al., 2014a; Zheng et al., 2015), which reveals that during extremely heavy pollution episode, the percentages of sulfate significantly increased instead of nitrate and ammonium. In July's case, the contributions of DO greatly decrease, because of the large decrease in energy consumptions due to the shutdown of winter heating system. The contributions of IN have the tendency of, different from the January's case, slight increase when $PM_{2.5}$ concentrations are lower than 300 μg m^{-3} and then decrease, with the percentages of 50.5%, 53.9%, and 51.0% for the concentration ranges of $\leq$200, 200–300, and >300 μg m^{-3}, respectively. The contributions of AG keep a slight increase as $PM_{2.5}$ concentrations increase, with the contributions of 11.4%, 12.4%, and 15.6%, for $PM_{2.5}$ concentrations of $\leq$200, 200–300, and >300 μg m^{-3}, respectively. It reveals that in summertime, NH_3 plays an important role in secondary $PM_{2.5}$ formation and reducing NH_3 emissions might effectively decrease the peak $PM_{2.5}$ concentrations in Shijiazhuang.

In Xingtai and Handan, the contributions of IN and DO in January have the similar trends as in Shijiazhuang, e.g., the contributions are relatively lower as $PM_{2.5}$ in the middle range. Averagely the IN contributions are 21.7%, 19.9%, and 26.5% for $PM_{2.5}$ concentrations of $\leq$200, 200–400, and >400 μg m^{-3} in Xingtai, and 27.3%, 20.8%, and 27.9% in Handan, respectively, in January. And the DO contributions are 21.5%, 20.6%, and 22.2% for Xingtai, and 19.5%, 18.7%, and 22.1% for Handan, for $PM_{2.5}$ concentrations of $\leq$200, 200–400, and >400 μg m^{-3}, respectively. The contributions of AG in Xingtai and Handan are also similar to those of Shijiazhuang, with lower contributions when $PM_{2.5}$ concentrations are low or high, and higher contributions when $PM_{2.5}$ concentrations are in the middle level.

The July's case for Xingtai and Handan is quite different from Shijiazhuang. The source contributions in Xingtai are 42.8%, 36.0%, and 43.1% for IN at $PM_{2.5}$ concentrations of $\leq$200, 200–300, and >300 μg m^{-3}, i.e., they keep a decrease–increase trend. And those numbers for AG are 10.1%, 8.1%, and 10.9% respectively. But the IN and AG contributions in Handan are keeping increase when $PM_{2.5}$ concentrations increase, e.g., the contributions of IN are 42.3%, 49.0%, and 64.8% for $PM_{2.5}$ concentrations of $\leq$200, 200–300, and >300 μg m^{-3}, respectively.

In general, the analysis of the relationship between the contribution and concentration indicates that in wintertime, the emission control of IN and DO are more effective during extremely heavily polluted periods in the three cities, but in summer time, the controls of IN are relatively more effective in Xingtai and Handan, and control of AG emissions are more effective in Shijiazhuang and Xingtai.

4.3. Sources of secondary $PM_{2.5}$

Recent studies reveal that a large fraction of $PM_{2.5}$ over North China is secondary aerosol (Huang et al., 2014; Wang et al., 2014a; Zheng et al., 2015; Wei et al., 2014a, b), e.g., secondary inorganic aerosol (SIA) including SO_4^{2-}, NO_3^-, and NH_4^+, and secondary organic aerosol (SOA). It is necessary to understand and quantify the sources of those secondary species in $PM_{2.5}$. However, the receptor models, e.g., CMB or PMF, can recognize the fractions of secondary $PM_{2.5}$, but cannot distinguish their spatial and sectoral origins and quantify their contributions (Tang et al., 2006; Zhang et al., 2015), which is very important in the policy making on a city level. The policy makers need to know the secondary aerosols come from regional or local sources, and if from local, what sectors are the major contributors. Only source models can answer those questions. Therefore, in this section, the spatial and sectoral source contributions to the secondary species of $PM_{2.5}$, as well as other major primary species, in the three cities are presented and discussed. It should be noted that because the SOA formation is poorly understood at present and the CMAQ model has the tendency of underpredict the SOA concentrations due to the missing of precursors and mechanisms (Beak et al., 2011), the SOA concentrations predicted in our simulations are quite low and not applied the source contribution analysis due to its high uncertainties.

As shown in Fig. 4(a), SO_4^{2-} accounts for 16.2% in the total $PM_{2.5}$ in Shijiazhuang in January, of which 33.1% comes from the source outside the city of Shijiazhuang, thus the regional contributed SO_4^{2-} is of 5.4% in total $PM_{2.5}$ (16.2% × 33.1%). In those regional sources, NHB is the largest contributor with 47% in total SO_4^{2-}, and the second one is the OTH, i.e., the other regions in Domain 1 outside the four provinces (HB, HN, SX, and SD) and BJTJ, with 14%. The third one is SX with the contributions of 10%. In the local sectors, IN, DO and AG are important source contributors with the percentages of 25.3%, 22.2%, and 12.8%. As for NO_3^- (7.6% in total $PM_{2.5}$), the regional sources are important that contribute 71.6% (5.4% in total $PM_{2.5}$), of which a large fraction comes from NHB (38%), SX (20%) and OTH (9%). In the local sources, the AG is the most important one (with 33.4%) due to importance of reactions between NO_x and NH_3 to generating secondary aerosol of NH_4NO_3. As for NH_4^+ (7.9% in total $PM_{2.5}$), the regional contribution is 42.3% (3.3% in total $PM_{2.5}$), of which the three top contributors are NHB (45%), SX (13%), and

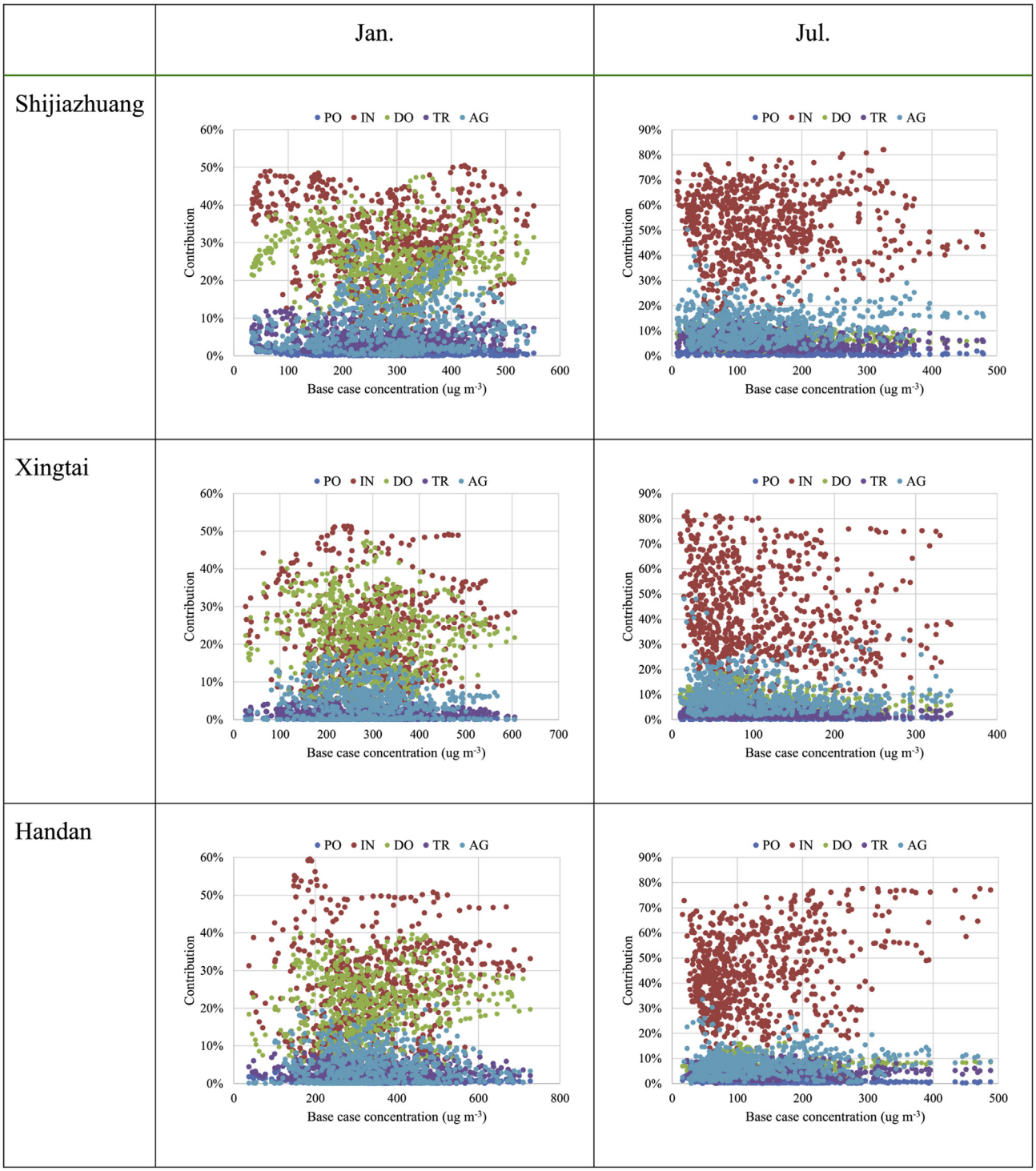
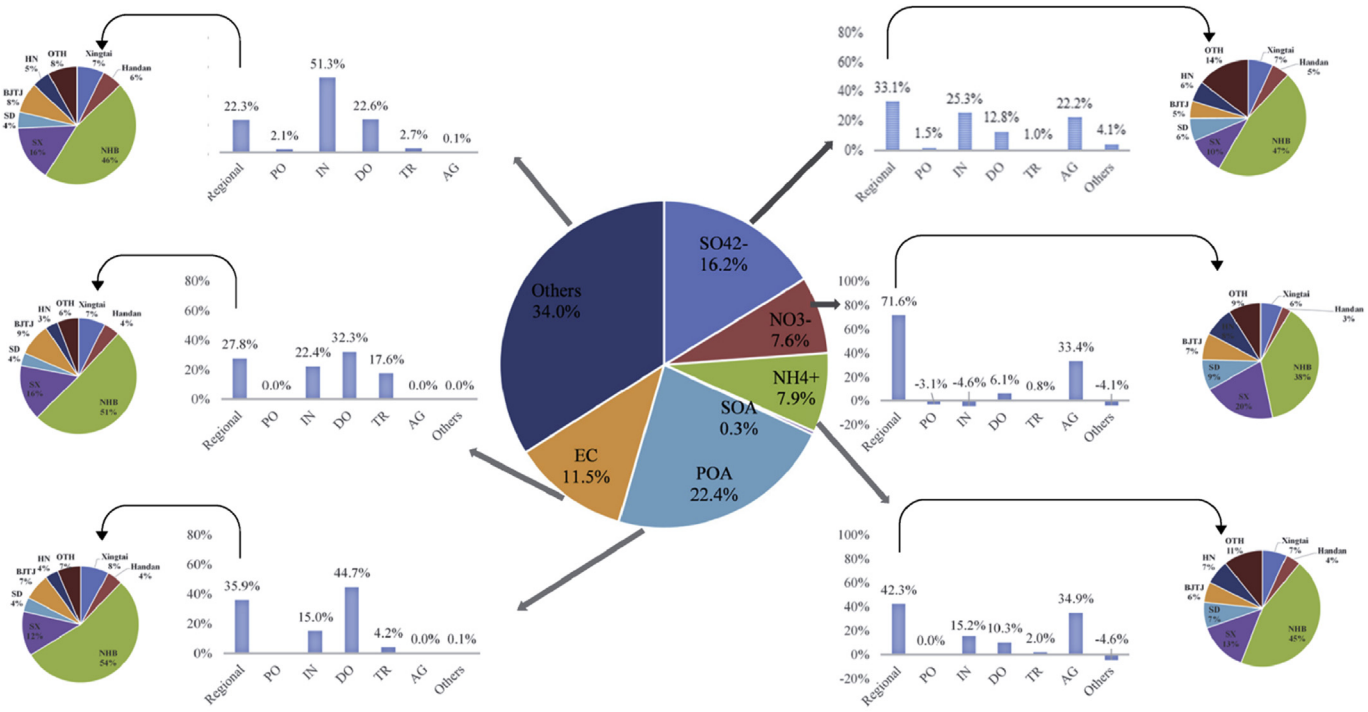


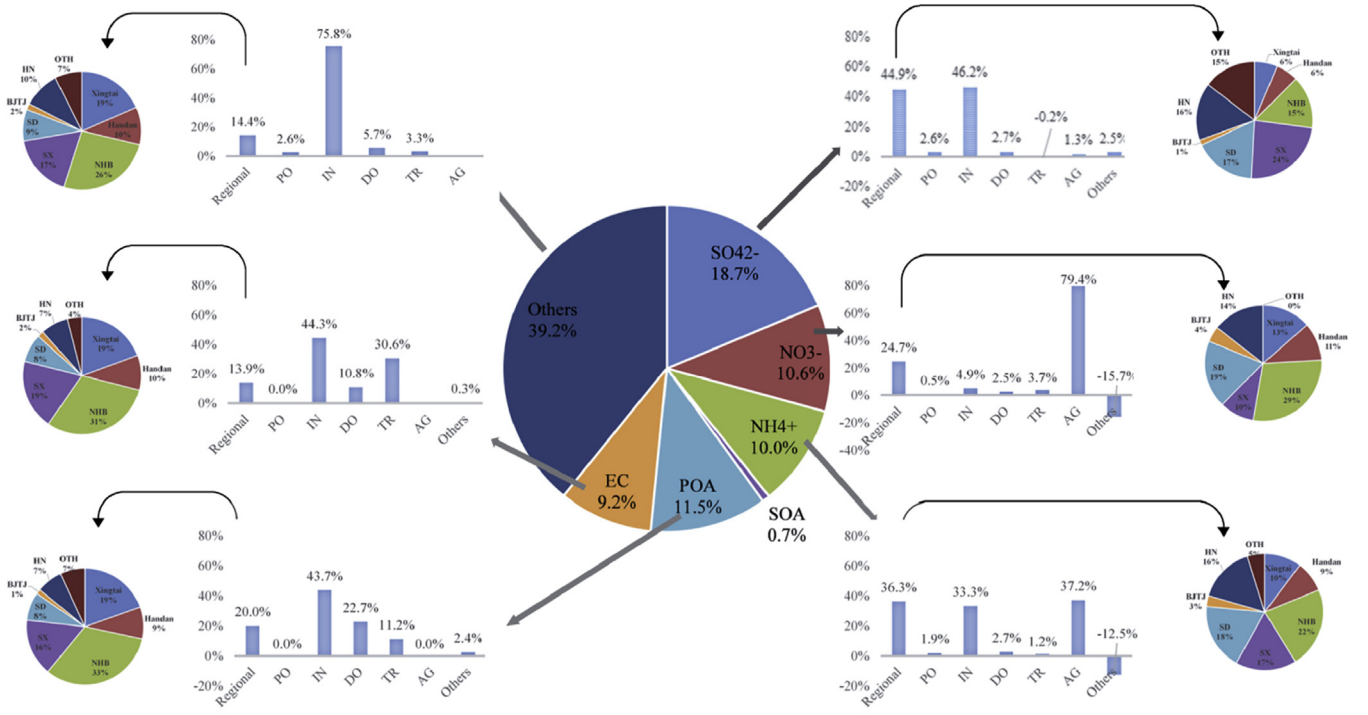
Fig. 3. The scatterplot of the base case $PM_{2.5}$ concentrations and the local sector contributions (%) in the Shijiazhuang, Xingtai and Handan.

OTH (11%). The local three key sectors are AG (34.9%), IN (15.2%), and DO (10.3%). The secondary $PM_{2.5}$, as counted as the sum of the SO_4^{2-}, NO_3^-, and NH_4^+ (given SOA is extremely low and of large uncertainties), is 31.7% in total $PM_{2.5}$ concentrations, and its regional contribution is 14.2% (5.4% + 5.4% + 3.3%) in total $PM_{2.5}$ and 44.5% in total secondary $PM_{2.5}$ (14.2%/31.7%). The local sector contributions to the total secondary $PM_{2.5}$, calculated by the same method, are 0.03% for PO, 15.6% for IN, 10.5% for DO, 1.2% for TR, and 28.0% for AG. Therefore, this plot can be used to estimate the relative regional and local source contributions to secondary $PM_{2.5}$ species, as well as the local sector source contributions. The results for primary organic aerosol (POA), EC, and other species in $PM_{2.5}$ are presented in Fig. 4(a) as well.

In July's case for Shijiazhuang (Fig. 4(b)), the fractions of the secondary species in $PM_{2.5}$ (39.3% in total) are larger than in January, due to the more rapid chemical reaction speed in the formation of secondary aerosols. The regional contributions to SO_4^{2-}, NO_3^-, and NH_4^+ are 44.9%, 24.7%, and 36.3%, respectively,

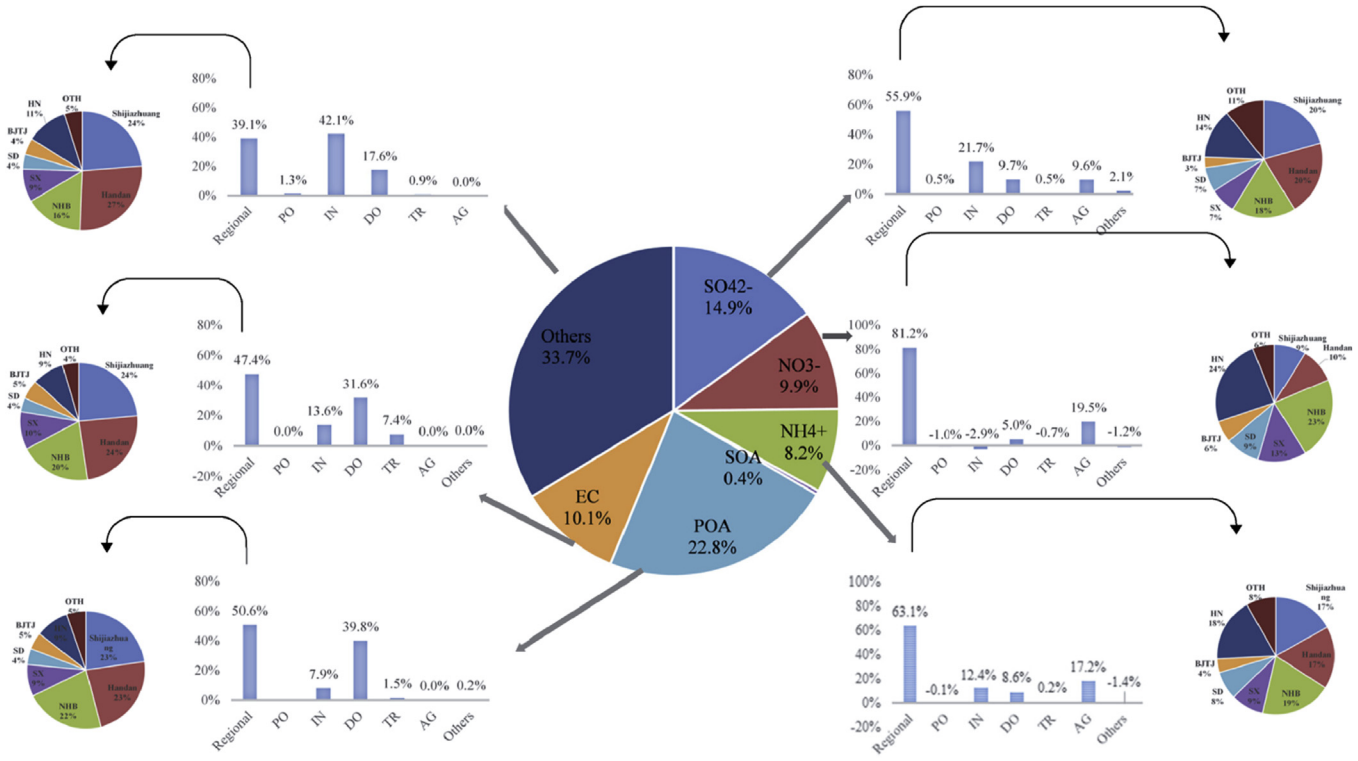


(a) Shijiazhuang, Jan.

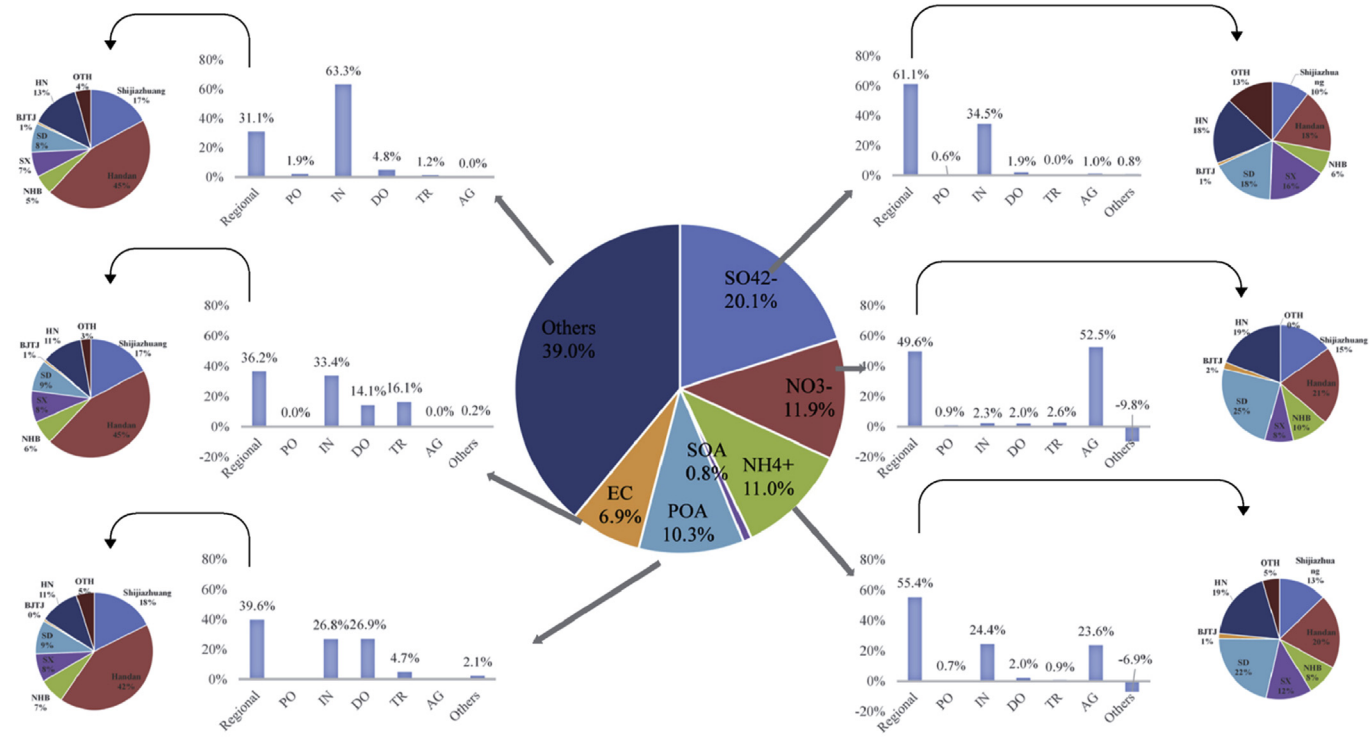


(b) Shijiazhuang, Jul.

Fig. 4. Regional and local sectoral source contributions to the major species in $PM_{2.5}$ in Shijiazhuang, Xingtai, and Handan. The pie charts in the middle of each plot are the chemical compositions of $PM_{2.5}$ in the city ("Others" represents the other species in $PM_{2.5}$). The bar charts are the contributions of the regional total and each local sector to each species in $PM_{2.5}$ ("Others" are the other sources besides the sources shown in the left and include the uncertainties resulted by non-linear response of $PM_{2.5}$ to its precursors). The pie charts along each bar chart depict the contributions of each region in the total regional contributions ("OTH" are the regions in Domain 1 besides the four provinces of HB, HN, SX, SD, and the two cities of BJ and TJ).



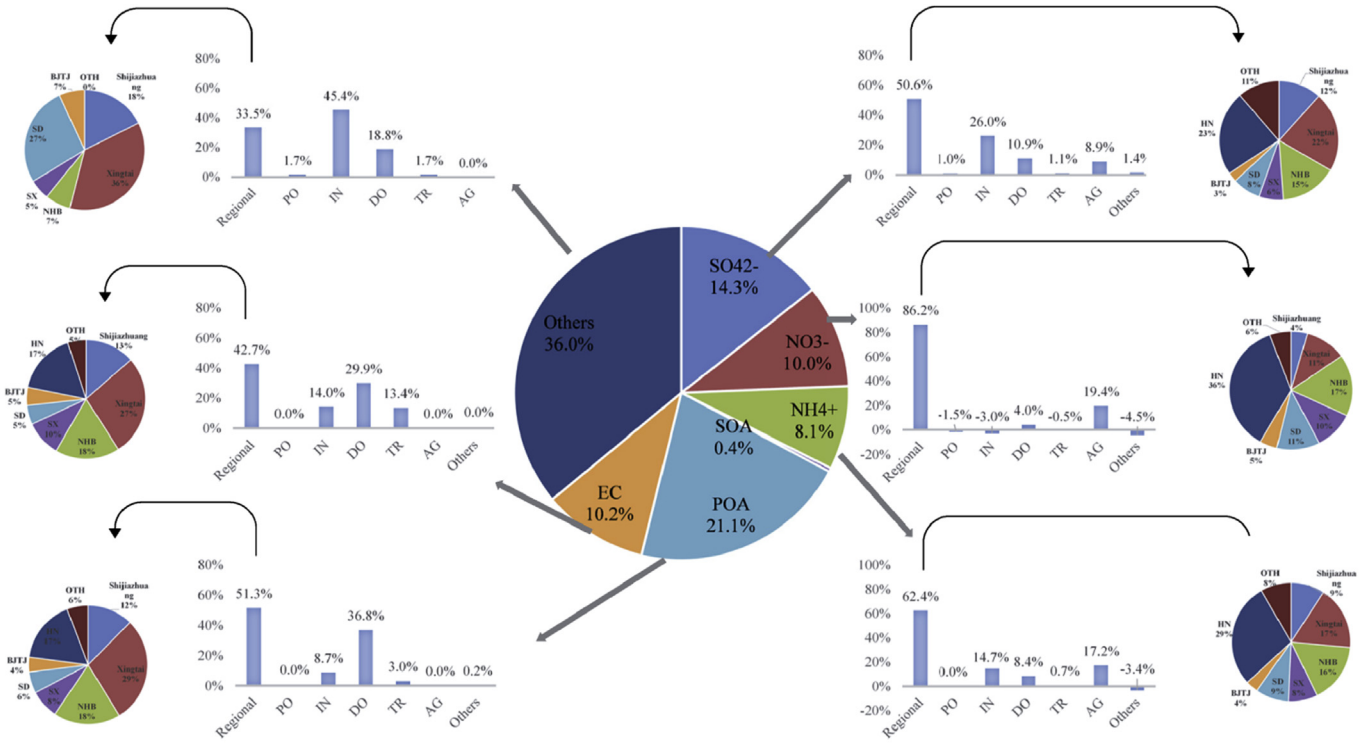
(c) Xingtai, Jan.



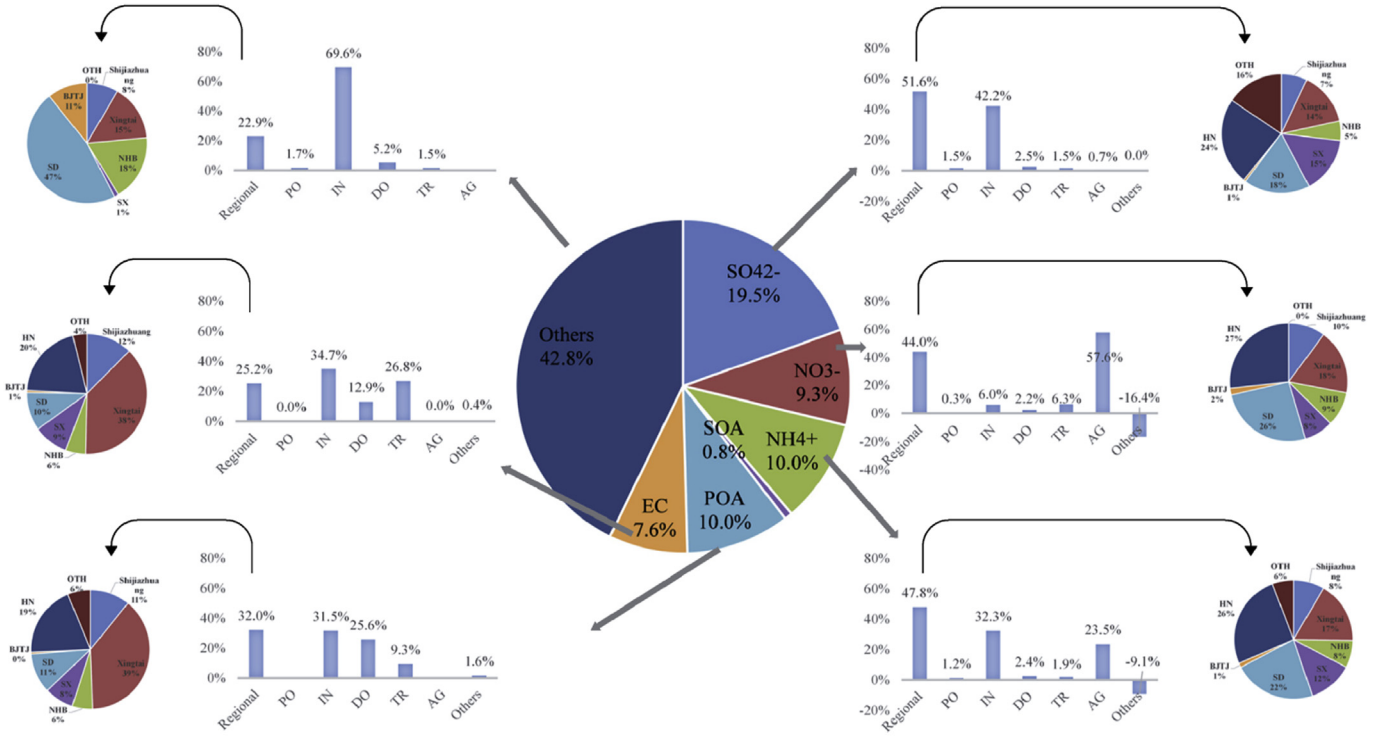
(d) Xingtai, Jul.

Fig. 4. (*continued*).

accounting for 37.3% in total secondary $PM_{2.5}$. The contributions of local sectors of PO, IN, DO, TR, and AG are 1.9%, 31.8%, 2.6%, 1.2%, and 31.5%, respectively, in total secondary $PM_{2.5}$. The other difference between July and January is the increase of the contribution of local IN sector and decrease of the DO, because of the shutdown of the domestic heating system in summer. In the regional contributions to secondary $PM_{2.5}$, the decrease of NHB contributions and increase of SX or SD contributions are quite obvious, which may mainly



(e) Handan, Jan.



(f) Handan, Jul.

Fig. 4. (*continued*).

attribute to the change in meteorological patterns in different seasons. Nevertheless, the two plots depict the spatial and sectoral origins of the secondary species in $PM_{2.5}$ in Shijiazhuang, and can be used to distinguish the regional and local contributions to secondary $PM_{2.5}$ resolved by receptor models, as well as the local sector contributions.

The plots for Xingtai (Fig. 4(c), (d)) and Handan (Fig. 4(e), (f)) are similar to those for Shijiazhuang. Comparing to Shijiazhuang, the regional contributions to the inorganic secondary $PM_{2.5}$ in Xingtai (accounting for 33.0% and 42.9% in $PM_{2.5}$ concentrations in January

and July) are obviously higher, with the fraction of 65.3% (vs. 44.5% for Shijiazhuang) in January. In July this number is 58.8% (vs. 37.3% for Shijiazhuang). The local sector contributions are −0.1%, 12.0%, 8.0%, 0.1%, and 14.5% for PO, IN, DO, TR, and AG in January, and 0.7%, 23.0%, 2.0%, 0.9%, and 21.0% in July, respectively. As for Handan (Fig. 4(e), (f)), the regional source contributions to total inorganic secondary $PM_{2.5}$ (accounting for 32.4% and 38.8% in $PM_{2.5}$ concentrations in January and July) are 69.4% and 48.8% in January and July, respectively. The local sector contributions are −0.4%, 10.3%, 7.2%, 0.3%, and 15.4% for PO, IN, DO, TR, and AG in January, and 1.1%, 31.0%, 2.4%, 2.8%, and 20.2% in July, respectively.

In summary, the regional contributions to the inorganic secondary $PM_{2.5}$, (i.e., SO_4^{2-}, NO_3^-, and NH_4^+), are 40.9%, 62.0%, and 59.1% for Shijiazhuang, Xingtai, and Handan, respectively, as an estimation of annual average. Within the local source, IN, DO, and AG are three key contributors to the secondary $PM_{2.5}$, with their contributions of 23.7%, 6.6%, and 29.8% in Shijiazhuang, 17.5%, 5.0%, and 17.7% in Xingtai, and 20.6%, 4.8%, and 17.8% in Handan, respectively. Regional joint air pollution controls and local controls of IN and AG are the keys to decrease the SIA concentrations in $PM_{2.5}$ in those three cities.

5. Conclusions

In this paper, the MM5–CMAQ air quality modeling system is applied over the East Asia and North China at 36- and 12-km horizontal grid resolutions for January and July, 2013. The model performance are evaluated against the NCDC meteorological surface observations and the air quality monitoring database in China by CNEMC, as well as the $PM_{2.5}$ chemical compositions observed at HEBEU site. Then the contributions of regional and local emissions, including major local sectors, to the $PM_{2.5}$ concentrations in the three top polluted cities, Shijiazhuang, Xingtai, and Handan, are assessed using the Brute-Force method with the simulations of 24 emission zero-out scenarios. In addition, the regional and local sectoral contributions to each chemical species in $PM_{2.5}$, especially the secondary species, are quantified and discussed to provide the understanding and the information to distinguish the spatial and sectoral origins of the secondary aerosols in those three cities, which takes a large fraction in $PM_{2.5}$ and cannot be apportioned by receptor models.

In general, the model performances are acceptable in meteorological predictions, although it tends to overpredict the wind speed and underpredict the precipitation in both months. $PM_{2.5}$ and PM_{10} predictions agree well with the observations with the trends, with all the $PM_{2.5}$ predictions meet the criteria of MFB $\leq \pm 60\%$ and MFE $\leq 75\%$. The chemical compositions of $PM_{2.5}$ at HEBEU site are reproduced although the model obviously underpredict the SO_4^{2-} during the extremely severe polluted episode in January, which may be mainly attributed to its weakness in the treatment of the heterogeneous chemistry.

The regional source contributions to the $PM_{2.5}$ concentrations in the three cities are estimated as 27.9% in Shijiazhuang, 46.6% in Xingtai, and 40.4% in Handan. The regional joint air pollution controls are more important in Xingtai and Handan than in Shijiazhuang, especially considering the trans-boundary transport and interactions of air pollutants between those three cities. On a provincial level, an integrated control policy should involve the considerations on pollutant transport between all the cities in Hebei, as well as the joint activities with adjacent provinces. Within the local emission sources, the major contributors are industrial, domestic and agriculture for all the three cities. Their contributions are 39.8%, 15.8%, and 10.6% in Shijiazhuang, 30.5%, 13.6%, and 6.9% in Xingtai, 35.9%, 13.5%, and 6.2% for Handan. The regional source contributions to the inorganic secondary $PM_{2.5}$ are 40.9%, 62.0%, and 59.1% in Shijiazhuang, Xingtai, and Handan, respectively, as an estimation of annual average. Industrial, domestic, and agricultural sources are three key local contributors to secondary $PM_{2.5}$ with the percentages of 23.7%, 6.6%, and 29.8% in Shijiazhuang, 17.5%, 5.0%, and 17.7% in Xingtai, and 20.6%, 4.8%, and 17.8% in Handan, respectively. The regional joint emission controls and the controls of local IN and AG, are important to reduce the SIA concentrations in those three cities.

Several limitations exist in the study. First, dust emissions are not included in this CMAQ version, which may lead to overestimation of regional total contributions and local sectoral contributions. Second, only the $PM_{2.5}$ chemical composition data at HEBEU site is used in model performance evaluation due to unavailability of data at other site. More observations and data sharing are needed to support the regional joint air pollution studies. Third, the sources of secondary organic aerosol are not explored because of the model's weakness in reproducing the formation of SOA in the atmosphere. Nevertheless, it still provides valuable information on source apportionment of $PM_{2.5}$ in those top polluted cities in China, which may be the first question that needs to be answered when designing future pollution control strategies by the city policymakers.

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