



Local and regional contributions to fine particulate matter in Beijing during heavy haze episodes



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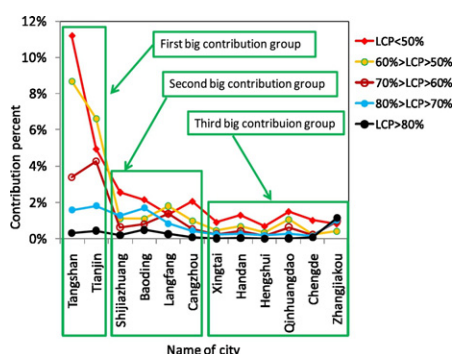
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HIGHLIGHTS

- Contributions to PM_{2.5} in Beijing from thirteen cities were simulated using PSAT.
- Extremely high PM_{2.5} concentration in Beijing is easily dominated by local emission.
- Transport contribution can easily lead to form moderate heavy haze episodes in Beijing.
- Tangshan, Tianjin, Shijiazhuang, Baoding, Langfang and Cangzhou are big contributors.

GRAPHICAL ABSTRACT



The average contribution percent of every other city in Jing-Jin-Ji region to the PM_{2.5} concentrations is larger than 75 $\mu\text{g m}^{-3}$ in Beijing urban for the pollution periods with different ranges of local (Beijing) contribution percent (LCP) during January 6–23, 2013.

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ABSTRACT

In order to alleviate extreme haze pollution, understanding the origin of fine particulate matter (PM_{2.5}) is crucial. In this study, we applied Particulate Matter Source Apportionment Technology (PSAT) in CAMx (Comprehensive Air Quality Model with Extensions) to quantify the impacts of emissions from different regions on PM_{2.5} concentrations in Beijing for haze episodes during January 6–23, 2013. Emission inventory was developed by Tsinghua University. Evolution of local and Regional contributions during local and non-local dominated haze episodes were discussed, separately. In the meanwhile, average contribution of other every city in Jing-Jin-Ji region to PM_{2.5} concentrations larger than 75 $\mu\text{g m}^{-3}$ in Beijing urban for each range of local contribution percent was analyzed. The results indicate that local emissions contributed 83.6% of PM_{2.5} at the urban center of Beijing, while regional transport from surrounding cities and parts of Shandong, Henan and Anhui provinces contributed 9.4%; long-range transport contributed the remaining 7.0% mainly from areas > 750 km away to the south of Beijing during this study period. Compared to non-local-dominated haze episodes, local-dominated heavy haze episodes in Beijing were easily resulted from unfavorable meteorological conditions with much lower PBL and wind velocity. Furthermore, local contribution is more easily to cause a sharp increase or sharp reduction of PM_{2.5} concentration in central Beijing, reflecting that Beijing local has much stronger potential to form

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extremely heavy haze episodes. The results indicated that controlling local emissions is a much more important measure to alleviate the extreme haze episodes in Beijing, like that on the night of Jan 12, 2013. Furthermore, emission control in Jing-Jin-Ji region, especially in Tangshan, Tianjin, Baoding, Langfang, Shijiazhuang and Cangzhou, as well as Henan and Shandong province, are important to reduce the $PM_{2.5}$ concentrations and the occurrence of haze episodes in Beijing.

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1. Introduction

With the rapid economic development and urbanization in China, air pollutant emissions have been increasing at an unprecedented rate over the recent two decades (Zhao et al., 2013c; Xing et al., 2011), and serious air pollution problems occur frequently in China, such as high concentrations of $PM_{2.5}$ (particulate matter with an aerodynamic diameter $<2.5 \mu m$) accompanying haze formation, attracting widespread attention (Fu et al., 2009; Lin et al., 2014; Wang et al., 2010; Wang and Hao, 2012; Wang et al., 2014d). Elevated levels of $PM_{2.5}$ have been linked to negative impacts on human health (Gurjar et al., 2010; Huang et al., 2012; Fann and Risley, 2011). In January 2013, an extremely severe haze occurred in eastern and central China with a record-breaking hourly $PM_{2.5}$ concentration of over $700 \mu g m^{-3}$ observed in Beijing, the capital city of China and the largest city in the Jing-Jin-Ji region which includes Beijing, Tianjin and Hebei, attracting worldwide attention. A number of papers have been published on the results of observation and modeling conducted during the severe haze episodes (Wang et al., 2014e; Cheng et al., 2014; Tao et al., 2014; Wang et al., 2014b; Wang et al., 2016). To tackle with heavy haze problems in China, especially in Beijing, in recent years, the government of China is trying to develop an effective control strategy. Accurately quantifying contribution of local region and other regions to the concentrations of $PM_{2.5}$ occurring during heavy haze episodes is crucial to establish effective control strategies to improve air quality.

The possibility of transport of particulates among the cities in Jing-Jin-Ji and its southern neighbors was suggested based on the analysis of weather phenomena and visibility observation data, SO_2 , NO_2 and PM_{10} concentrations, MODIS AOD, CARSNET AOD, and the CALIPOS0 extinction coefficient (Wang et al., 2014a). The meteorological and chemical characteristics during heavy haze events in Beijing were discussed based on a measured data (Zhang et al., 2015; Han et al., 2016). But

they did not analyze the regional contribution. Positive matrix factorization (PMF) as one of the receptor model is usually used to conduct source apportionment of $PM_{2.5}$, but receptor model can only obtain the contributions from some different emission categories instead of regional contributions (Yao et al., 2016). A potential source contribution function (PSCF) was used to analyze the potential region contribution to the ambient concentration, but it only depended on the meteorological data without any chemical affection (Yao et al., 2016).

Three-dimensional Eulerian chemical transport models (CTMs) such as CMAQ, CAMx and WRF-chem, which link emission to the ambient concentration, provide useful tool to quantify the source contributions of each region or city to the ambient concentrations of pollutants. Additionally, some approaches have been developed based on three-dimensional Eulerian CTMs, although developing a completely reliable source-receptor relationship remains a challenging task (Seinfeld and Pandis, 2006). The Brute Force method (BFM) as a very simple method was used in aMM5-CMAQ modeling system to quantify the source contributions of major source regions and sectors to $PM_{2.5}$ concentrations in the most polluted cities in southern Hebei province (Wang et al., 2014c), but the sum of all source contributions is not equal to the simulated concentration in the base case whenever the model response is nonlinear (Koo et al., 2009). The Decoupled Direct Method (DDM) developed by Dunker, (1980) and (1981)) is an efficient and accurate method for sensitivity analysis (Dunker et al., 2002), compared with the BFM. However, this approach is usually suitable for responses to small or moderate emission changes rather than contributions from different source regions or categories. Source Oriented External Mixture (SOEM) (Kleeman and Cass, 2001) as a method with tagged-species is potentially accurate, but it is computationally very demanding (Koo et al., 2009). As a more efficient and flexible approach, the PM Source Apportionment Technology (PSAT) (Wagstrom et al., 2008) can be used to apportion

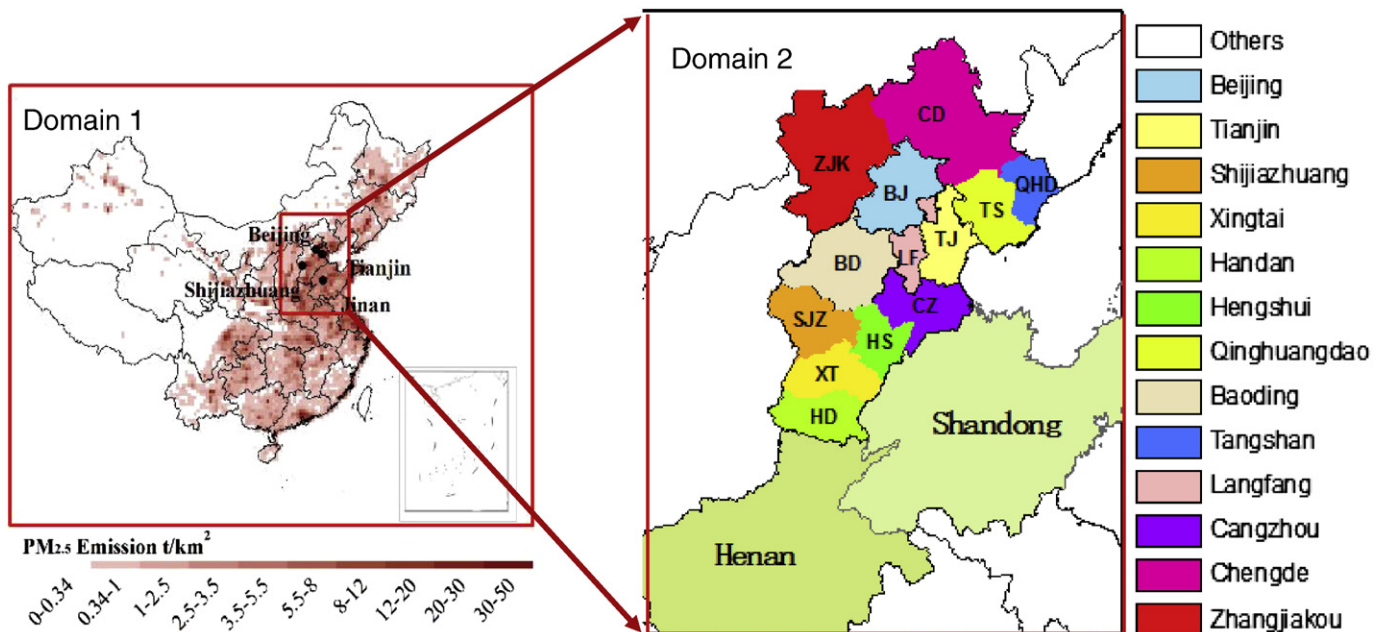


Fig. 1. CAMx modeling domains with horizontal grid cell resolutions of 36 km over China and 12 km over the Jing-Jin-Ji region. The colors on the left figure show the emission levels for $PM_{2.5}$. The second modeling domain (D2) covers the Jing-Jin-Ji region including thirteen cities, parts of Shandong and Henan provinces, and other regions colored white.

primary PM, secondary PM and gaseous precursors of secondary PM among different source categories and source regions (ENVIRON, 2011). Li et al. (2013) and Huang et al. (2012a) applied PSAT to investigate the contributions of SO_2 (sulfur dioxide) from emission sources in Tangshan and Beijing, Northern China, but, their studies did not identify source regions of $\text{PM}_{2.5}$. A research group has successfully conducted the source apportionment of secondary organic aerosol in Beijing to local and distant source contributions in summer (Lin et al., 2016). PSAT method has been a helpful approach to quantify the contributions from different regions to fine particulate matter.

Annual and monthly average (not about heavy haze episodes) source contributions to $\text{PM}_{2.5}$ of Beijing urban in 2006 and 2013 in the Jing-Jin-Ji (Beijing-Tianjin-Hebei) region have been conducted using PSAT Method (Li et al., 2015), but they did not quantify the contributions to Beijing from different cities in Jing-Jin-Ji region. Li and Han (2016) have conducted the source apportionment of $\text{PM}_{2.5}$ in Beijing during heavy haze episodes, but they did not quantify the contributions to Beijing from different cities in Jing-Jin-Ji region either. However, understanding the contribution of each city in Jing-Jin-Ji to Beijing's $\text{PM}_{2.5}$

concentration is very important. On the one hand, it will help Chinese Central Government to establish an effective regional joint framework of action system for Jing-Jin-Ji region. On the other hand, it will help individual cities in emission control in their own cities under a regional joint framework for Jing-Jin-Ji region. Furthermore, heavy haze episodes occur frequently in Beijing in recent years. How to reduce occurrence of heavy haze episodes has become a very important issue. How much is the fraction of $\text{PM}_{2.5}$ concentration in Beijing during heavy haze episodes contributed by local emission as well as by every other city in the Jing-Jin-Ji region? This is the key issue on setting an effective emission control strategy to reduce the $\text{PM}_{2.5}$ concentration and the occurrence of heavy haze episodes in Beijing.

In this study, CAMx (Comprehensive Air Quality Model with Extensions) with PSAT tool was applied to study the contributions of emissions from each city in the Jing-Jin-Ji region, and other regions to $\text{PM}_{2.5}$ concentrations in Beijing urban during heavy haze episodes on January 06–23, 2013. The Jing-Jin-Ji region comprises thirteen cities: Beijing, Tianjing, Shijiazhuang, Xingtai, Handan, Hengshui, Tangshan, Baoding, Qinhuangdao, Langfang, Cangzhou and Chengde. Their contributions

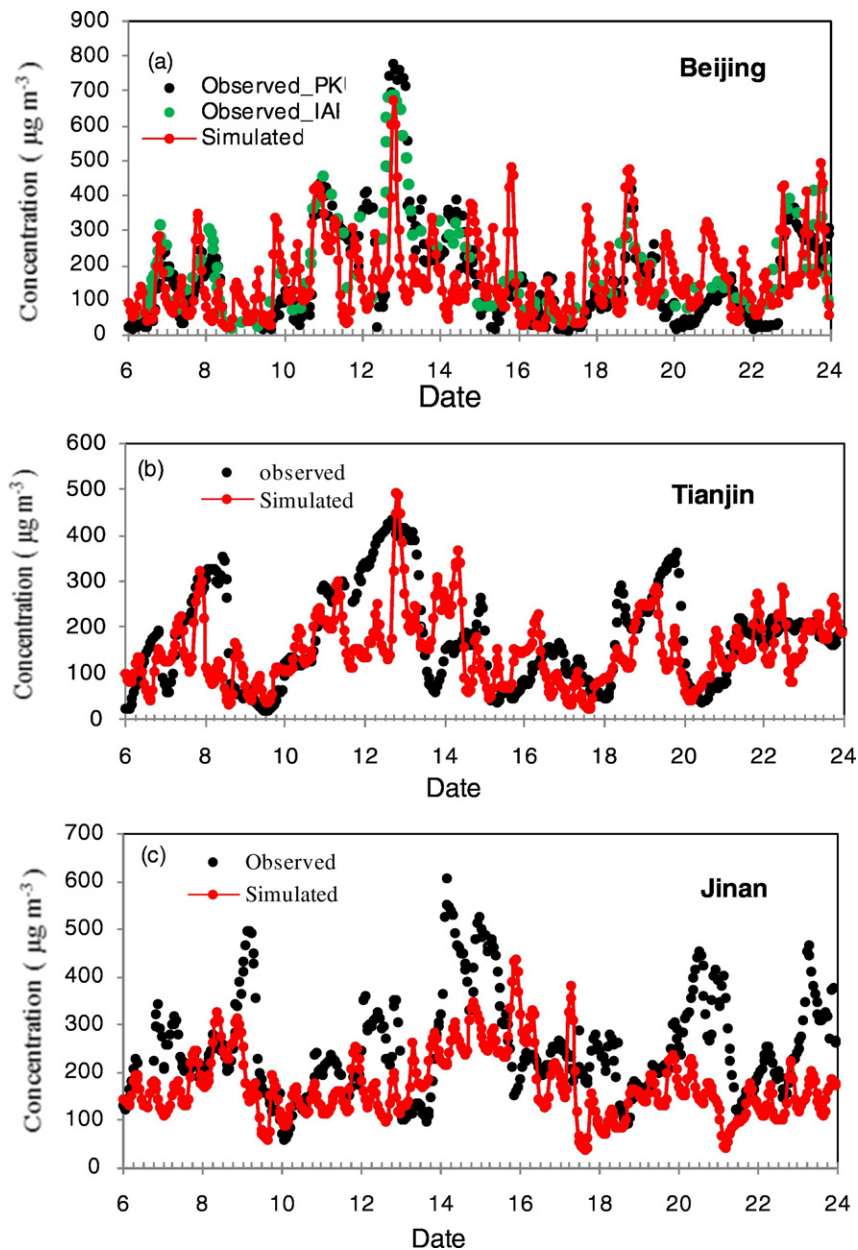


Fig. 2. Comparison of simulated and observed $\text{PM}_{2.5}$ concentrations during Jan. 6–23, 2013 (LT).

to Beijing's PM_{2.5} for both local dominated and non-local dominated episodes during heavy haze episodes were discussed. The results of this study can provide useful information for decision makers, helping them propose effective strategies to reduce the pollution level and the occurrence of heavy haze episodes in Beijing.

2. Model configuration and performance evaluation

2.1. Model configuration

In this study, two modeling domains are simulated, as shown in Fig. 1. The outermost (first) domain, with a horizontal grid spacing of 36 km, covers all of China, Japan, and Korea as well as parts of India and South-east Asia. The second domain, with a horizontal grid spacing of 12 km, covers the entire area of Beijing, Tianjin and Hebei province, parts of Shandong, Henan, Shanxi, Jiangsu, Anhui, Liaoning provinces and part of the Inner Mongolia Autonomous Region. The region covering Beijing, Tianjin and Hebei province is usually called as the Jing-Jin-Ji region, and the entire Hebei province which is composed of eleven administrative cities, namely Shijiazhuang, Xingtai, Handan, Hengshui, Tangshan, Baoding, Qinhuangdao, Langfang, Cangzhou, Chengde, and Zhangjiakou. Both domains have 14 layers extending vertically from the surface to an altitude of about 19 km above the ground, with the first layer about 40 m thick. The outputs from the first domain are used to provide the boundary condition for the second domain. The PSAT tool is only used for the second domain. The configurations of chemical initial conditions and emission inventory are consistent with our previous papers (Zhao et al., 2013b; Zhao et al., 2013a; Wang et al., 2014b). Biogenic emissions were generated using the Model of Emissions of Gases and Aerosols from Nature (MEGAN). Other anthropogenic emission inventories from point and area sources were established by Tsinghua University. These anthropogenic emission data were processed by separate programs and ArcGIS software instead of the Sparse Matrix Operator Kernel Emissions (SMOKE). All anthropogenic emission data are based on 2012, and the total anthropogenic PM_{2.5} missions are 61.5 Kt/a, 112.7 Kt/a, and 875.0 Kt/a for Beijing, Tianjin and Hebei, respectively; NO_x emission amounts of Beijing, Tianjin and Hebei province are 397.8 Kt/a, 392.5 Kt/a and 1619.7 Kt/a, respectively; And SO₂ emission amounts of Beijing, Tianjin and Hebei province are 183.0 Kt/a, 286.7 Kt/a, respectively.

Meteorological fields were modeled using WRF (the Weather Research and Forecasting Model) version 3.4, developed by the US National Center for Atmospheric Research. The WRF model configuration includes version 2 of the Kain-Fritsch cumulus cloud parameterization

(Kain, 2004), the Asymmetric Convective Model version 2 for the PBL (Pleim, 2006; Pleim, 2007), the RRTMG radiation mechanism and the Pleim-Xiu land-surface model (Pleim and Xiu, 1994; Xiu and Pleim, 2000) with indirect soil moisture and temperature nudging (Pleim and Gilliam, 2008; Pleim and Xiu, 2003). CAMx version 5.40 with PSAT tool was used to simulate air quality and apportion sources of fine particulate matter in Beijing. The Carbon Bond 05 chemical mechanism (CB05) (Yarwood et al., 2005) was used as the gas-phase chemical mechanism in the CAMx model and module ZHANG03 was chosen to simulate dry deposition. As to aerosol chemistry, inorganic aqueous chemistry (RADM-AQ), inorganic gas-aerosol partitioning (ISORROPIA), secondary organic aerosol formation/partitioning (SOAP) and the CF (two-mode coarse/fine) scheme were used in this study. All elevated point sources were processed using separated code, instead of the Plume-in-Grid (PiG) module, before CAMx modeling was begun.

In order to quantify the source apportionment of PM_{2.5} in Beijing with a high population density, one grid cell geographically covering Peking University (PKU) and the Institute of Atmospheric physics (IAP) air pollution observation stations was designated as the receptor which is located at the urban center of Beijing. The regions considered here as source regions are Beijing itself, Tianjin, eleven cities in Hebei province, part of Shandong province, part of Henan province, other regions within the second domain (D2) and those outside the second domain. The contribution from emissions in those regions outside the second domain can be regarded as the contribution from long-range transport. A spin-up period of 5 days is used to minimize the influence of the initial conditions. Back trajectories, following a parcel of air backward over several hours or days, were also used to analyze the formation of extremely high PM_{2.5} concentrations. These back trajectories were generated by HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) model, which is available on the website of the National Oceanic and Atmospheric Administration (NOAA) Air Resources Laboratory (<http://ready.arl.noaa.gov/HYSPLIT.php>).

2.2. Model performance evaluation

Fig. 2 shows temporal variations in modeled and observed PM_{2.5} concentrations in Beijing, Tianjin, and Jinan (the capital city of Shandong province). Hourly modeled data in Beijing are the average concentrations of PM_{2.5} in the grid cell where both the Peking University (PKU) and Institute of Atmospheric Physics (IAP) stations are located. Hence, the observations from the both these stations can be used for comparisons with the modeled data from that grid cell. The observed data at IAP station was obtained from one reference (Tian et al., 2014).

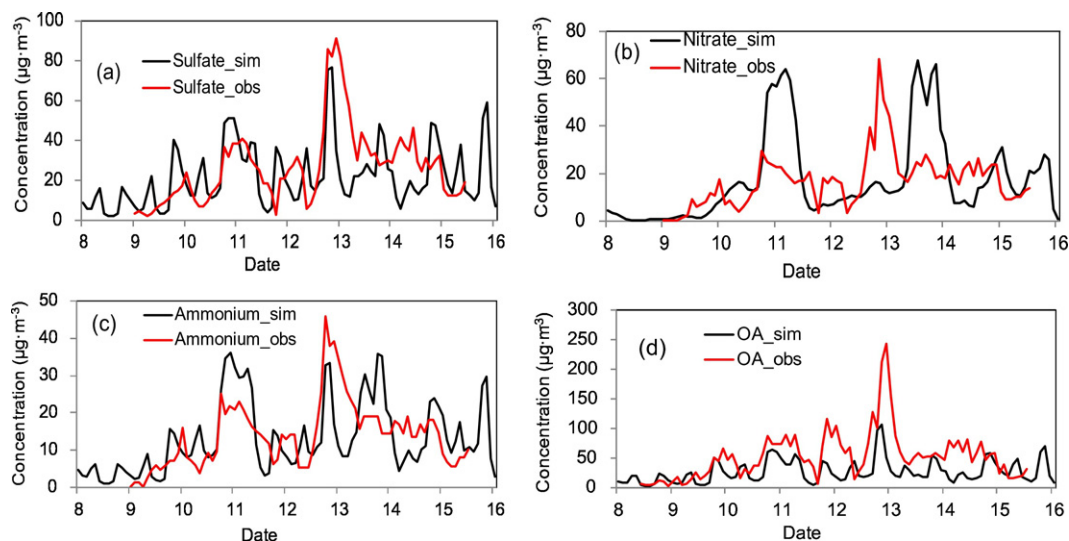


Fig. 3. Comparison of simulated and observed concentrations of sulfate, nitrate, ammonium and organic aerosol.

Table 1

Quantitative evaluation of predicted PM_{2.5} concentrations with hourly observations taken in Beijing, Tianjin and Jinan during January 6–23, 2013.

City	Predicted average ($\mu\text{g m}^{-3}$)	Measured average ($\mu\text{g m}^{-3}$)	Number of data pairs	BIAS ($\mu\text{g m}^{-3}$)	ERROR ($\mu\text{g m}^{-3}$)	RMSE ($\mu\text{g m}^{-3}$)	FBIAS	FERROR	IOA
Beijing	163.68	180.32	483	-16.64	109.92	149.19	3.71%	67.98%	0.647
Tianjin	150.56	176.05	427	-25.50	74.72	99.02	-7.07%	49.01%	0.678
Jinan	174.03	252.08	418	-78.05	103.46	131.56	-34.55%	47.15%	0.526

Very high concentrations of PM_{2.5} in Beijing were observed in January 2013, especially on Jan. 12 and Jan 13. The highest observed concentration reached an hourly average of more than 700 $\mu\text{g m}^{-3}$, a record-breaking high concentration of PM_{2.5}, on the night of Jan 12, Fig. 2 shows that most of the concentration peaks of observed PM_{2.5} are captured by the simulation. The highest modeled concentration of PM_{2.5} in Beijing is 675 $\mu\text{g m}^{-3}$, which is comparable to the observed average

values of 722 $\mu\text{g m}^{-3}$ from the PKU and IPA stations on the night of Jan 12. Both the overall trend and the peaks were captured very well by simulations for Jan. 6, 11, 19 and 23.

Since concentration data of observed chemical compositions in PM_{2.5} were not published by government, and there are only some data of concentrations of sulfate, nitrate, ammonium and organic aerosol (OA) in PM₁ instead of PM_{2.5} only for a short time period which can

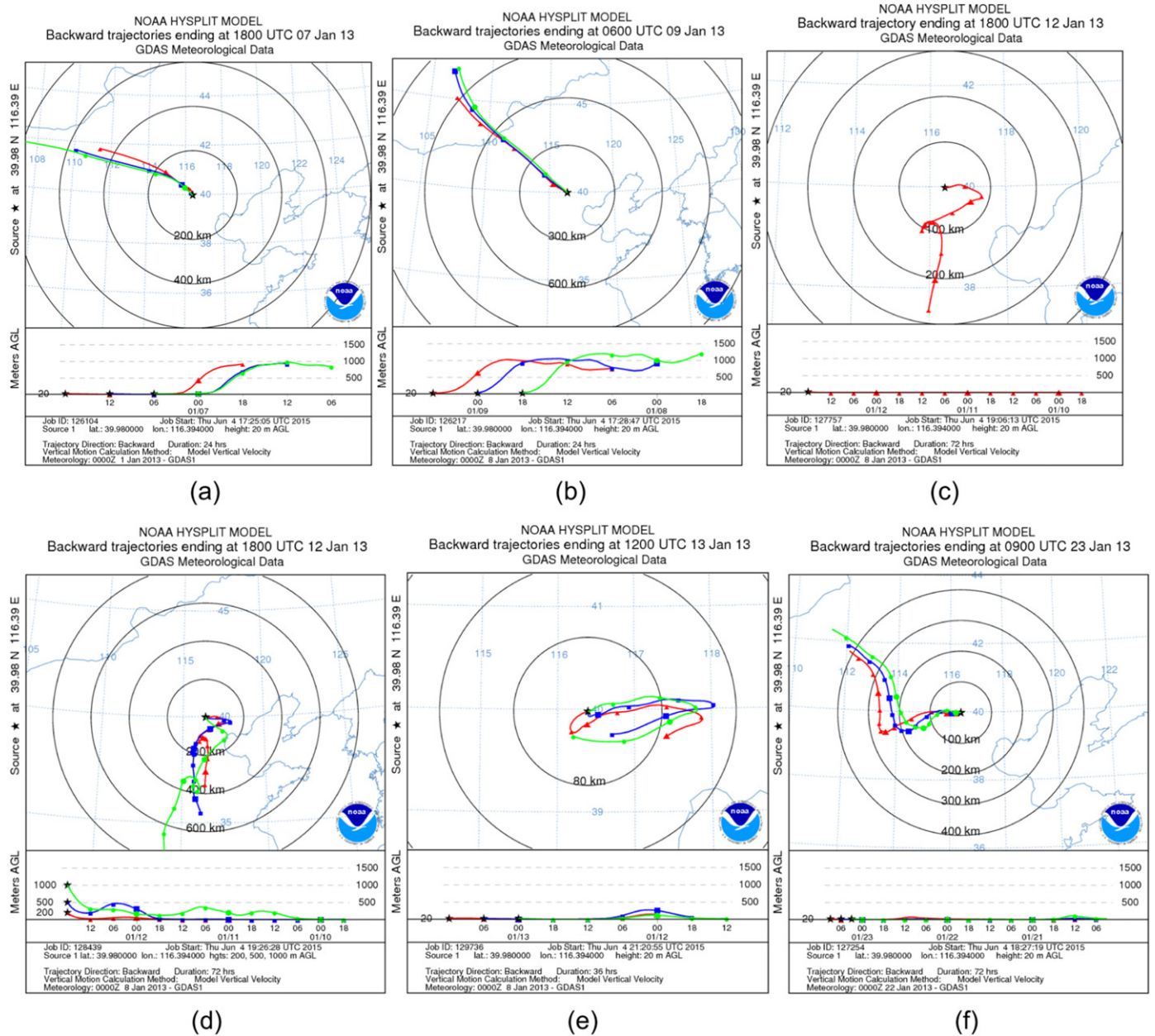


Fig. 4. Backward trajectories terminating at Beijing (116°22'E,39°58' N): (a) 24 h backward trajectories at 18:00, 12:00 and 06:00 on Jan 7 at 20 m; (b) 24 h backward trajectories at 06:00 and 00:00 on Jan 9 and at 18:00 on Jan 8, at 20 m; (c) 72 h backward trajectories at 18:00 on Jan 12 at 20 m; (d) 24 h backward trajectories at 18:00 on Jan 12 at 200 m, 500 m and 1000 m; (e) 36 h backward trajectories at 12:00, 06:00 and 00:00 on Jan 13 at 20 m; (f) 72 h backward trajectories at 09:00, 06:00 and 00:00 on Jan 23 at 20 m.

be obtained from previous study (Sun et al., 2014). Apparently, concentrations of these compositions in PM_{10} are lower than those in $PM_{2.5}$, since PM_{10} is only a fraction of $PM_{2.5}$. However, they usually have similar variation trends with time, and some information about model's ability also can be acquired if observed concentration of compositions in PM_{10} is used to compare with simulated concentration of compositions in $PM_{2.5}$. In Fig. 3, simulated concentrations of sulfate, nitrate, ammonium and organic aerosol in $PM_{2.5}$ were compared with observed concentrations of those species in PM_{10} at Beijing's urban center. The variation trends of simulated data and observed data matched very well during a severe haze period from the afternoon on 10th Jan. to the afternoon on 11th Jan. Their variation trends of sulfate, ammonium, and organic aerosol also matched well during the heaviest haze episode around midnight of 12 Jan. although model failed to reproduce enough high concentration of nitrate. The discrepancy of simulated and observed nitrate concentrations at night of 12 Jan and in the early morning of 13 Jan. is more likely the result from a failure to accurately capture the significant formation process of nitrate by heterogeneous reaction in nighttime during the extremely heavy haze episode. For organic aerosol, simulated concentrations were generally lower than those observed values, suggesting that the model failed to reproduce enough secondary organic aerosol due to deficiency of chemistry mechanism in models. However, there are still similar variation trends of simulated and observed OA concentrations with time. Generally, variation trends of simulated concentration for these compositions of $PM_{2.5}$ can capture the pattern of observed data.

Quantitative evaluation of predicted $PM_{2.5}$ concentrations with hourly observations taken in Beijing, Tianjin and Jinan is shown in Table 1. The evaluation is based on RMSE (Root-Mean-Square Error), Gross ERROR, BIAS, FBIAS (Fraction of BIAS), FERROR, IOA (Index of Agreement) as the statistical parameters (Emery et al., 2001). These statistical metrics have not been calculated for sulfate, nitrate, ammonium and OA in $PM_{2.5}$ due to their observed concentration of species in PM_{10} instead of $PM_{2.5}$. Overall, the model's performance in reproducing concentrations of $PM_{2.5}$ in Beijing, Tianjin and Jinan varied from average (FBIAS $\leq \pm 60\%$ and FERROR $\leq 75\%$) to good (FBIAS $\leq \pm 30\%$ and FERROR $\leq 50\%$) (USEPA, 2007; Morris and Koo, 2005). Consequently, the model performance is acceptable to analyze the source apportionment of $PM_{2.5}$ at Beijing's urban center during this modeling period.

3. Results and discussion

3.1. Trajectory analysis

All backward trajectories shown in Fig. 4 terminated at the Institute of Atmospheric Physics located at the urban center of Beijing. The five trajectories shown in Fig. 4(a) represent the paths of air masses whose end time varied from 14:00 Jan 07 to 02:00 Jan 08 (LT) and which moved from northwestern China over an 18-hour period at a very low height. High concentrations of $PM_{2.5}$ (more than $300 \mu\text{g m}^{-3}$) were observed during this period because of weak vertical dispersion of air pollutants in these masses. Fig. 4(b), on the other hand, shows a backward trajectory with relatively strong dispersion of pollutants, from the upper air near or about 1000 m; correspondingly its $PM_{2.5}$ concentration, on the morning of Jan 9, was relatively low. However, the parcel of air at 02:00 on Jan 13 (LT) had a complicated backward trajectory over 72 h, as displayed in Fig. 4(c). This trajectory exhibits three features: 1) The air mass moved very slowly over 60 h within a small horizontal range with a diameter of 100 km and speed less than about 1.0 ms^{-1} . For more than 12 h its speed was very low, less than about 0.5 ms^{-1} on average. 2) The air mass remains data very low height for over 72 h, indicating a weak vertical dispersion of air pollutants. 3) When the air mass was in the northern part of Baoding city, about 100 km from Beijing, it remained stagnant for a long time. Its backward trajectory shows this parcel of air circulating around Baoding city and stagnant for about 30 h, subsequently entering Langfang and Tianjin

cities, and finally approaching Beijing. The end time of three trajectories in Fig. 4(d) is the same as in Fig. 4(c), but their end heights were 200m, 500 m and 1000 m. All three air parcels came from the ground surface, as shown in Fig. 4(d), suggesting that air pollutants within these air masses are easily concentrated from surface emissions and the airflow in the upper air are quite difficult to disperse. Correspondingly, an extremely high concentration of $PM_{2.5}$ was observed in central Beijing at 02:00 on Jan 13 (LT). These paths are closely related to geographical and meteorological conditions. At that time, air circulation was remarkably zonal at 500 h Pa over North China: an inversion layer, caused by warm air advection, was present; a low pressure field was dominant; a wind convergence zone was established along the plain-mountain transition area; and a weak south wind prevailed over the Piedmont plain in the Jing-Jin-Ji region (Wang et al., 2013). The pollutants in the southern region were carried onto the Piedmont plain by the wind and accumulated, resulting in extremely high concentrations of $PM_{2.5}$ (about $700 \mu\text{g m}^{-3}$). The backward trajectories in Fig. 4(e) showed strong air circulation in the region covering Beijing, Tangshan, Tianjin, Langfang, Baoding and Cangzhou, suggesting there was relatively strong transportation of pollutants between these cities. The three backward trajectories displayed in Fig. 4(f) were at $<100 \text{ m}$ in height over 72 h, and their velocities were very low before entering Beijing, resulting in very weak dispersion of air pollutants in these air masses, both vertically and horizontally. The pollutants were concentrated at the surface layer due to the intense, thick inversion layer and low mixed-layer height (Wang et al., 2013). Thus, a heavy haze episode occurred in Beijing on January 23, which was characterized by high concentrations of $PM_{2.5}$, as shown in Fig. 2.

3.2. Average modeled $PM_{2.5}$ concentrations

The average modeled ground-level concentrations of $PM_{2.5}$ in the second domain for January 6–23, 2013 is depicted in Fig. 5. Very high concentrations of $PM_{2.5}$ were seen over some cities, especially over the urban areas of Beijing, Tianjin, Tangshan and Shijiazhuang, which are densely populated with high energy consumption accompanied by emissions of $PM_{2.5}$ about 4–10 times greater than in cities to the

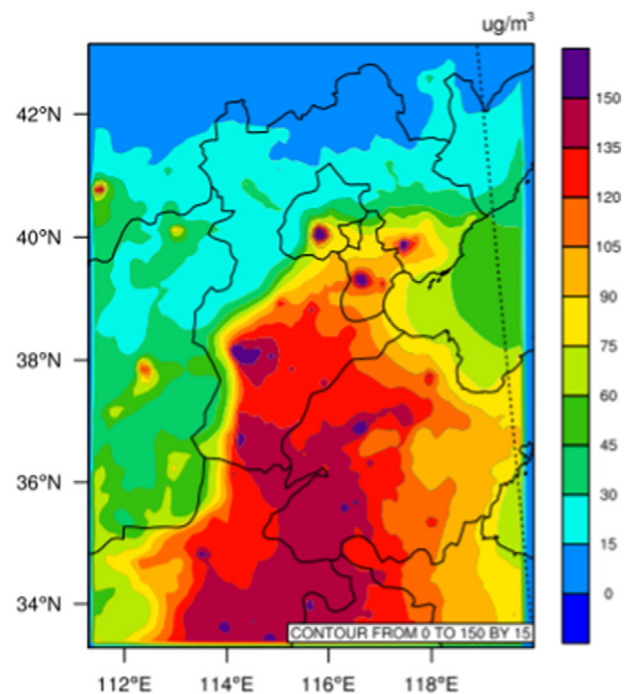


Fig. 5. Average modeled ground-level concentrations of $PM_{2.5}$ in the second domain for January 6–23, 2013 (LT).

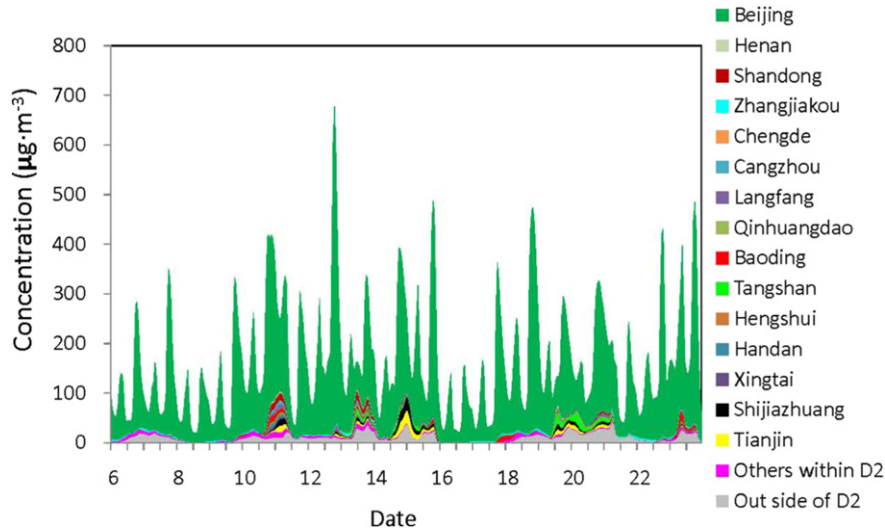


Fig. 6. Temporal variation of contributions of emissions from different regions to PM_{2.5} concentrations at the urban center of Beijing during January 6–23, 2013 (LT).

north. This suggests that the emission of pollutants from local region contributes greatly to ambient PM_{2.5} concentration in each of these cities. The locations of these cities are shown in Fig. 1. Compared with the high PM_{2.5} concentrations in Beijing, there are very low PM_{2.5} concentrations over areas to north and northwest of Beijing, because of geographic features. Beijing is in the northwest of the North China Plain, near the meeting point of the Taihang and Yanshan mountain ranges. The municipality’s outlying districts and counties extend into the mountains that surround this city from the southwest to the northeast, but the city itself lies on flat land that opens to the east and south. PM_{2.5} concentrations are usually low over these mountainous areas, to the north and northwest of Beijing, because of low anthropogenic emissions. In contrast, PM_{2.5} concentrations are high in Beijing and the areas to its east and south. High concentrations of PM_{2.5} are also found on the southern boundary of the simulation domain, as shown in Fig. 5, suggesting notable transportation of pollutants from areas outside this domain during the simulation period when the wind came from south.

3.3. Temporal variation of source apportionment results

The temporal contribution of emissions from different regions to PM_{2.5} concentrations in central Beijing during January 6–23, 2013 is shown in Fig. 6. In the legend, D2 refers to the second domain of CAMx modeling in this study as shown in Fig. 1). PM_{2.5} concentrations in central Beijing are apportioned into nineteen categories, seventeen

of which vary with time, and the concentrations from the initial condition and the ocean region can be ignored due to their extremely low values. The contribution from boundary conditions of the second domain, which is the finest domain in this work, can be roughly taken as the contribution from long-range transport. From Fig. 6, it can be easily seen that the contribution from Beijing (local emissions) accounted for a big fraction of PM_{2.5} concentrations in central Beijing for most of the simulation period. For example, the total PM_{2.5} concentration rose to more than 700 µg m⁻³ at 19:00 on January 12 (LT), while the contribution from areas outside Beijing only accounted for about 30 µg m⁻³ of the total. Fig. 7 displays the hourly distributions of velocity field and planetary boundary layer (PBL) for 17:00–20:00 January 12 (LT). At 17:00, the PBL in Beijing ranged from 150 m to 800 m, while the PBL was much higher and the wind blew from south to north to the south of the simulation domain. But, 1 hour later the PBL was <100 m over the entire Beijing city, while the air over central Beijing was nearly stationary and there was air circulation around Beijing. After 18:00, the PBL in Beijing, even in the whole Jing-Jin-Ji region, continued to decrease at first and was then stationary for a few hours. The stagnant meteorological conditions were unfavorable for pollutant dispersion and occurred simultaneously, while large amounts of pollutants were continuously emitted from surface sources into the air within this very low layer, resulting in the accumulation of pollutants at surprising levels and the abnormally high PM_{2.5} concentrations observed on the evening of January 12.

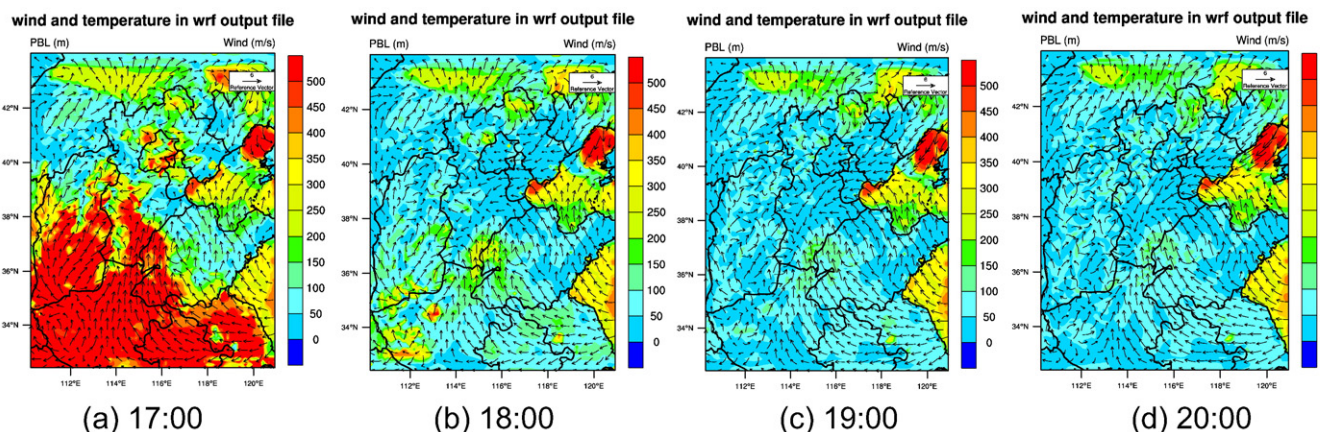


Fig. 7. Hourly distribution of velocity field and PBL on January 12, 2013 (LT).

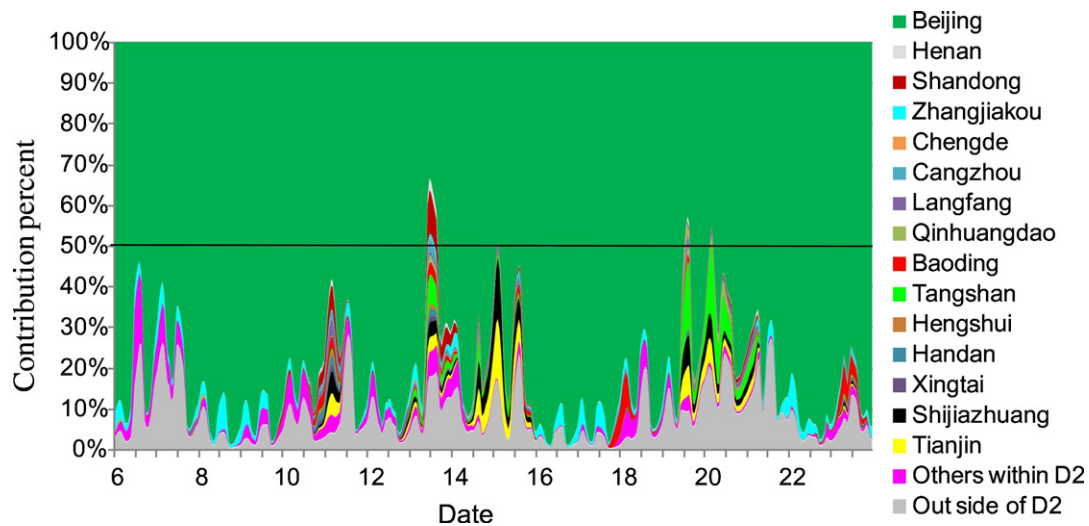


Fig. 8. Temporal variation of contribution percentages of emission from different regions to $PM_{2.5}$ concentrations in central Beijing during Jan. 6–23, 2013 (LT).

The temporal variation of contribution percentages of emissions from different regions to $PM_{2.5}$ at the urban center of Beijing during January 6–23, 2013 (LT) are shown in Fig. 8. Very high $PM_{2.5}$ concentrations were generally dominated by local contributions during this simulation period: local Beijing emissions contributed 93.0% of the high $PM_{2.5}$ levels of more than $700 \mu\text{g m}^{-3}$ on the night of January 12. Helped by the perturbation of the weather system, this concentration decreased sharply after January 12. Even so, the $PM_{2.5}$ level in the afternoon and evening of January 13 was still high. As shown in Fig. 4(e), a distinct circulation

of air masses occurred and carried pollutants into Beijing from Tianjin, Boding, Shijiazhuang, Tangshan, leading to a significant contribution from areas outside Beijing on the afternoon of January 13, which accounted for >50.0% of the $PM_{2.5}$ concentrations in central Beijing, although total $PM_{2.5}$ concentrations were very high, compared with abnormally high concentrations in the evening of January 12. Additionally, emissions from Shijiazhuang, Tianjin and Tangshan accounted for >25.0% of total $PM_{2.5}$ concentrations on the afternoon of January 19. Among the cities in Jing-Jin-Ji region, there were only Shijiazhuang

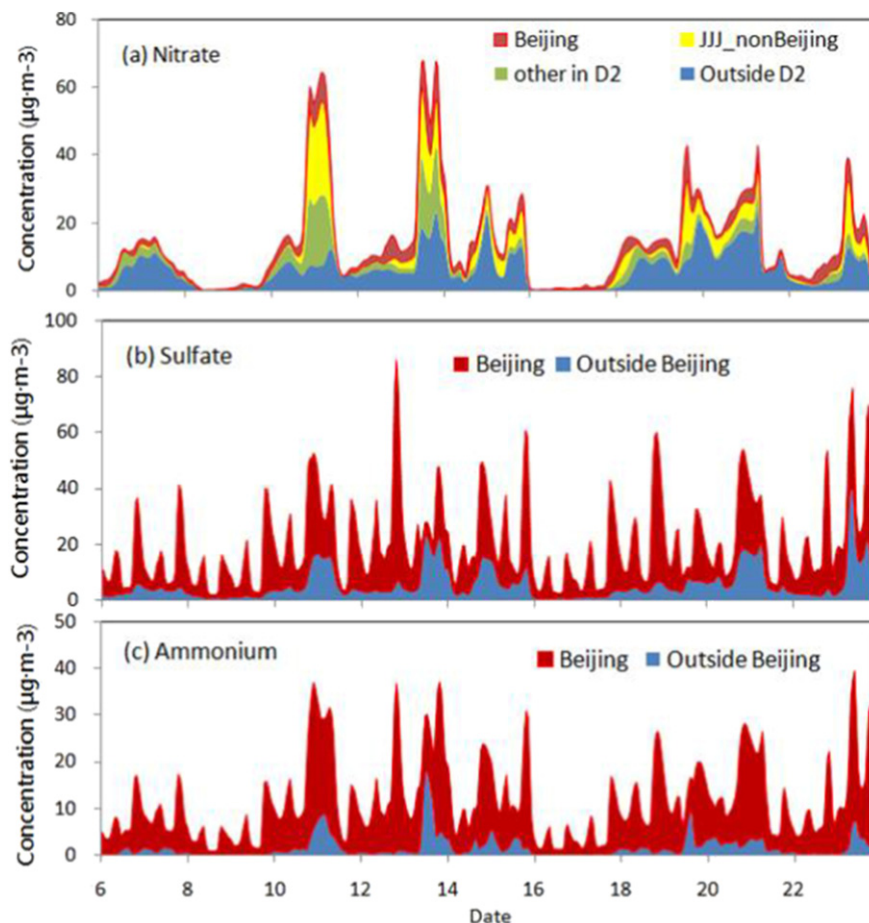


Fig. 9. Temporal variation of contributions from different regions to nitrate, sulfate and ammonium at the urban center of Beijing during Jan. 6–23, 2013 (LT).

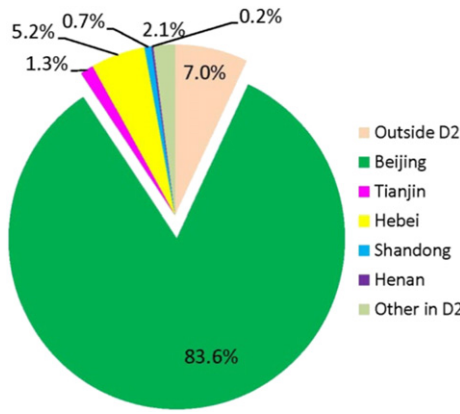


Fig. 10. Average contribution percentages of different regions to PM_{2.5} concentrations in central Beijing during January 6–23, 2013.

and Tianjin which contributed obviously and sum of them accounted for about 30.2% of total PM_{2.5} concentrations during the haze pollution at the night of January 14. Figs. 6 and 8 show that PM_{2.5} concentrations in central Beijing were usually in the range of 100 μg m⁻³– 300 μg m⁻³ when dominated by sources from areas outside Beijing. In contrast, high PM_{2.5} concentrations of over 300 μg m⁻³ in central Beijing were usually dominated by local, Beijing, emissions.

Nitrate, sulfate, and ammonium are the important secondary components in PM_{2.5}. The total mass concentration of nitrate, sulfate and ammonium accounted for 29.7% of PM_{2.5} in central Beijing during Jan. 6–23, 2013 (LT). In order to better understand the formation of these inorganic species during heavy haze episodes, contributions from different region to nitrate, sulfate and ammonium in PM_{2.5} at the urban center of Beijing during Jan. 6–23, 2013 (LT) were simulated using the PSAT, and the results is presented in Fig. 9. Nitrate concentrations have apportioned into contributions from Beijing, JJJ_nonBeijing, other in D2 and areas outside D2. Contribution from each region outside Beijing is larger than Beijing local contribution to nitrate. Region outside D2 generally contributed a big fraction of nitrate concentrations at the urban center of Beijing during this modeling period, although Jing-Jin-Ji region excluding Beijing, Shandong and Henan provinces contributed greatly during severe haze episodes. By comparison, Beijing contributed only a small fraction of nitrate concentrations during severe haze episodes. On the contrary, the local contribution dominated averagely both sulfate and ammonium concentrations at the urban center of Beijing during Jan. 6–23, 2013. As secondary compositions, nitrate, sulfate and ammonium accounted for significant fractions in PM_{2.5} during this modeling period.

3.4. Average contributions from local and other regions

Fig. 10 shows average contribution percentages from different regions to PM_{2.5} concentrations in central Beijing during January 6–23,

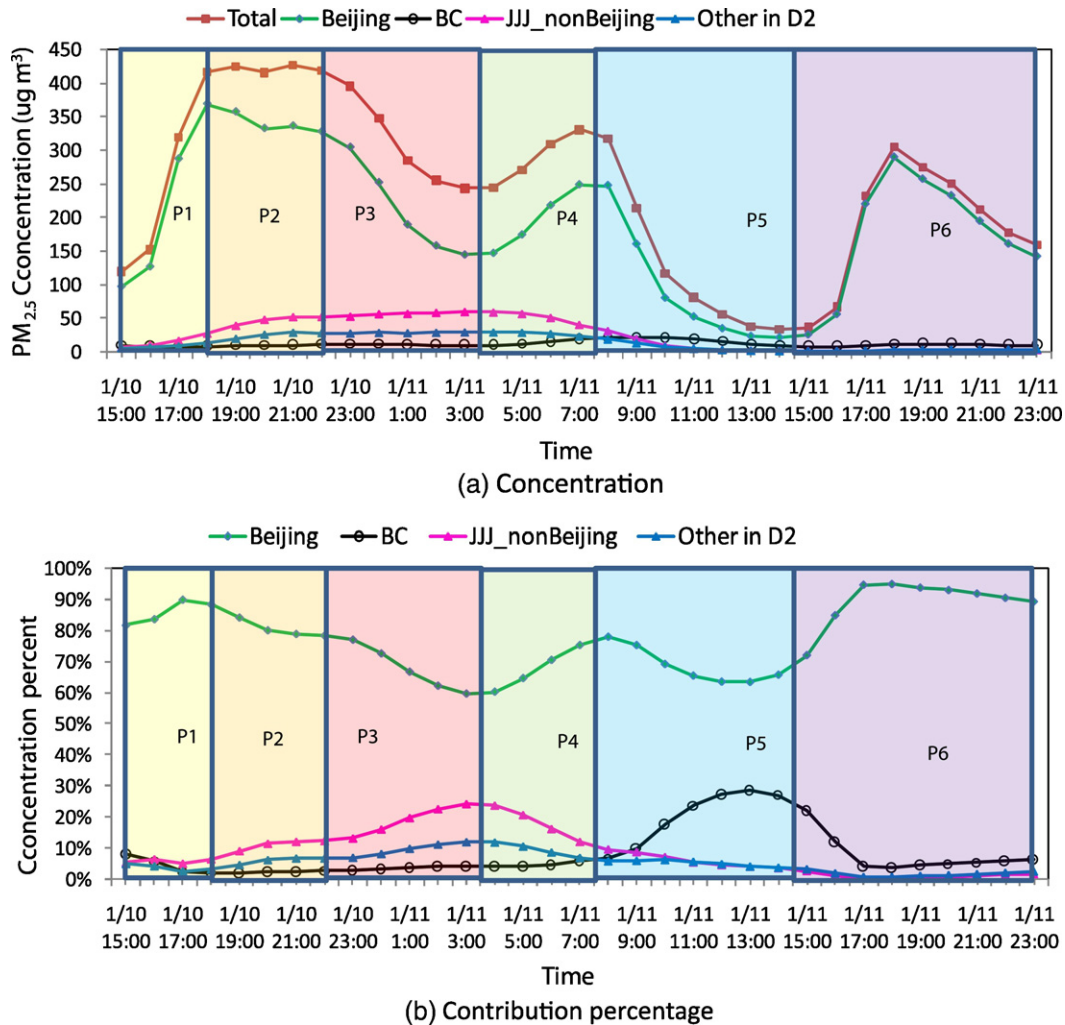


Fig. 11. Evolution of contributions and their percentages of different regions to PM_{2.5} concentrations in central Beijing in a haze case during 15:00 January 10 to 23:00 January 11, 2013.

2013. The average local (Beijing) contribution reaches 83.6%, indicating that $PM_{2.5}$ concentration in central Beijing was dominated by local contributions on average. Regional transport contributes 9.4%: 1.3% from Tianjin city, 5.2% from cities in Hebei Province, 0.7% from Shandong, 0.2% from Henan and 2.0% from other cities in the simulation domain (D2). Regions outside D2, representing long-range transport, contribute 7.0%. These results show that the most effective measure to reduce average $PM_{2.5}$ concentrations in central Beijing during the simulation period, would be controlling emissions in Beijing.

Local contribution of $PM_{2.5}$ in January 2013 in Beijing was conducted using RAQMS (a regional air quality model system) and BFM (brute force method) in Ref. (Li and Han, 2016). Their results show that nearly 44% of $PM_{2.5}$ in Beijing in January 1–31, 2013 was contributed by local emission. Differences between this study and their work may be mainly due to different models and methods employed. It is not very easy to compare RAQMS with CAMx because many parameters involved might be different, although they are both three-dimensional Eulerian models. The comparison of BFM with PSAT has been discussed above. In addition, the study periods are not totally the same and the Emission inventory of 2010 was used in their study. In recent years, rapid change of economic and industrial activities in China leads to an obvious change of emission from 2010 to the study year. Source apportionment of $PM_{2.5}$ in Beijing was conducted using CAMx with PSAT method (Li et al., 2015). In their study, annual average local contribution percent of $PM_{2.5}$ in Beijing in 2013 was >50%, but corresponding results for heavy haze episodes in 2013 were not reported. Their study revealed that change of source contribution to $PM_{2.5}$ in Beijing was dominated by change of local emission. Our results are generally comparable with previous work. Although the overall performance of the model in simulating $PM_{2.5}$ is acceptable, uncertainties of meteorology, emission inventory and parameterizations in model exist, subsequently, affecting the accuracy of source apportionment results.

3.5. Contribution evolution during local-dominated haze episodes

To better understand the contributions from different regions to the total $PM_{2.5}$ concentration in central Beijing, we chose a haze episode that occurred during January 10–11, 2013 to analyze the evolution of the contributions from local (Beijing), JJJ_nonBeijing (Jing-Jin-Ji region excluding Beijing), other areas within D2, and areas outside D2 representing the Boundary condition or long-range transport. Fig. 11 shows the evolution of contributions and their percentages from these regions in the local dominated haze case where non-local contribution percentage is enough obvious to analysis. As shown in Fig. 11, the total $PM_{2.5}$ concentrations in central Beijing increased sharply from $120 \mu g m^{-3}$ to about $430 \mu g m^{-3}$ with the Beijing contribution increased sharply, and both contributions from JJJ_nonBeijing and others in D2 increased slowly during P1 period. During P2 period, Beijing contribution decreased, while both contributions from JJJ_nonBeijing and others in D2 increased, but the total $PM_{2.5}$ concentrations in central Beijing remained almost the same elevated concentration, suggesting that the increase of contribution from JJJ_nonBeijing and other in D2 can offset the reduction of Beijing contribution. The $PM_{2.5}$ concentration contributed from BC which representing long range transport impact maintained a low level during this case as shown in Fig. 9(a), although its contribution percentage is as high as 28.6% at 13:00 on January 11, as shown in Fig. 9(b), because of the very low total concentration of $PM_{2.5}$ in central Beijing. Both contributions from JJJ_nonBeijing and other in D2 are relative high during P2, P3 and P4 periods, compared with those during P1, P5 and P6 periods. The variation of total $PM_{2.5}$ concentrations in central Beijing were controlled by the variation of Beijing contribution in this case except during P2 period. In sum, based on the source apportionment results of this case we can conclude that the sharp increase and sharp reduction are both controlled by the local (Beijing) contribution, consequently Beijing should take the main responsibility for the high $PM_{2.5}$ concentration in this kind of case.

3.6. Contribution analysis in a non-local dominated case

As shown in Figs. 6 and 8, there were still a few haze episodes during which $PM_{2.5}$ concentrations in central Beijing were dominated by areas outside Beijing during Jan. 6–23, 2013. In order to better understand the impact of areas outside Beijing on the haze formation in central Beijing, a case on Jan. 13 was chosen to analyze the contribution from areas outside Beijing, especially from each city in Jing-Jin-Ji region. Fig. 12 shows temporal variation of contributions and their percentages from Beijing, JJJ_nonBeijing (i.e. areas including Hebei province and Tianjin city), others within D2 and BC (areas outside D2) at 7:00 to 18:00 on January 13, 2013 (LT). The total $PM_{2.5}$ concentration in central Beijing decreased from $217 \mu g m^{-3}$ to $147 \mu g m^{-3}$ with time varying from 7:00 to 9:00, because the contribution from Beijing decreased greatly although the other regions' contributions increased gradually. With continuous reduction of Beijing contribution and continuous increase of areas outside Beijing contribution, the contribution percentage of areas outside Beijing contribution reached 62.2% at 10:00, showing the dominating role in the high total $PM_{2.5}$ concentration in central Beijing. During 10:00–15:00 period the total $PM_{2.5}$ concentration ($129.8 \mu g m^{-3}$ – $162.6 \mu g m^{-3}$) in central Beijing dominated by areas outside Beijing by 58.4%–66.6%, and contribution from BC was quite similar to contribution from the D2 excluded Jing-Jin-Ji region, while the contribution from JJJ_nonBeijing (Jing-Jin-Ji region excluded Beijing) was larger than those from BC and other in D2. After 15:00 Beijing contribution became dominated again because the Beijing contribution increased sharply while the others contribution decreased gradually, leading to the total $PM_{2.5}$ concentration in central Beijing going up sharply.

The contribution percentages from Shandong, Henan, each city in Jing-Jin-Ji region and the other within D2 during 10:00–15:00 Jan. 13, 2013 (LT) are displayed respectively in Fig. 13. According to the magnitude of contribution percentage of different regions, there are three groups which can be divided. The biggest contribution group is composed of Shandong, Tangshan and other in D2 which covers a large area including partial of inner Mongolia, Shanxi, Anhui, Jiangsu and Liaoning in D2 as shown in Fig. 1; the second big contribution group is composed of Tianjin, Shijiazhuang, Cangzhou, Baoding and Henan;

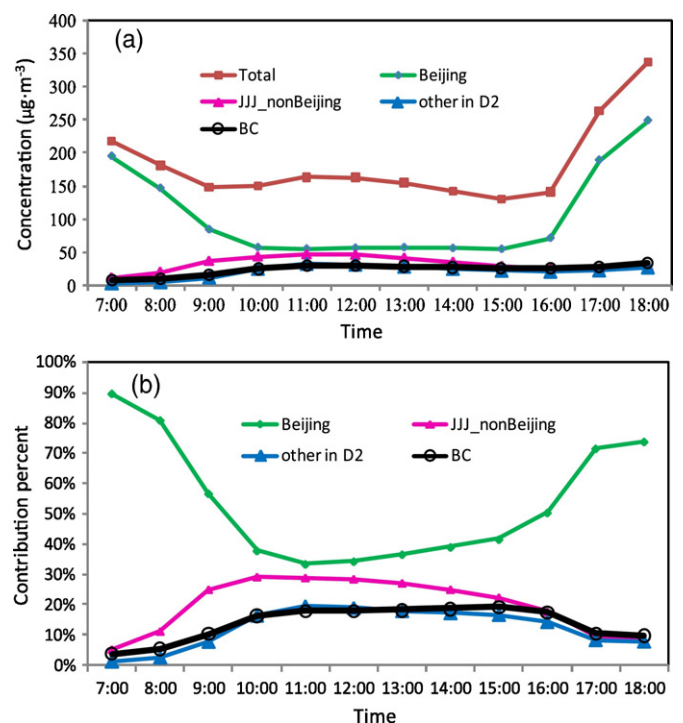


Fig. 12. Temporal variation of contributions and their percentages of Beijing, JJJ_nonBeijing, others within D2 and BC during 7:00–18:00 January 13, 2013 (LT).

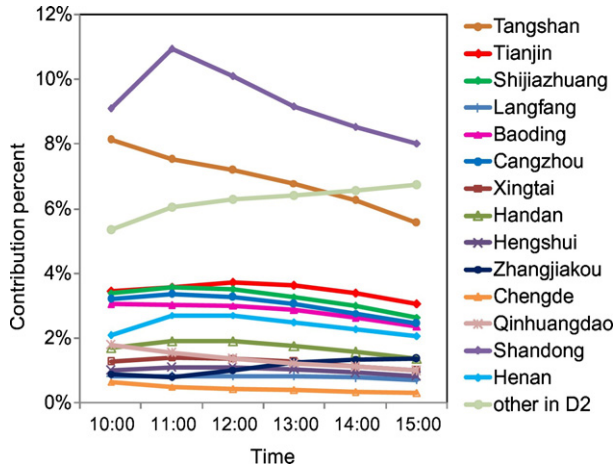


Fig. 13. Contribution percentages from Shandong, Henan, each city in Jing-Jin-Ji region, and other within D2 during 10:00–15:00 on January 13, 2013 (LT).

The small contribution group is composed of the rest cities of Jing-Jin-Ji region. The cities in first group contributed large while the cities in third group contributed small. As displayed in Fig. 4(e), there was a strong circulation over Beijing, Tangshan, Tianjin, Langfan, Baoding and Cangzhou, resulting relatively big contributions of transport among these cities.

The PM_{2.5} concentration in the simulation domain for 10:00–15:00 on January 13, 2013 (LT) was apportioned into four parts: the contributions from Beijing, JJJ_nonBeijing, others within D2 and the Boundary condition, as shown in Fig. 14. The average velocity field and PBL distribution in the simulation domain during this period are shown in Fig. 15. Although the PBL in central Beijing was as high as about 850 m, the wind in Beijing blew slowly from west and from east in the same time as displayed in Fig. 15, thus air gathered to the central Beijing and flowed slowly out to the northeast and southwest, resulting a obvious contribution to the southern region and a big local contribution to PM_{2.5} concentrations in central Beijing as shown in Fig. 14(a). Furthermore, the contribution from Jing-Jin-Ji region excluding Beijing to central Beijing is nearly equivalent to that from local emissions. Fig. 14(c) shows an obvious contribution from the areas within D2 and outside Jing-Jin-Ji regions to average PM_{2.5} concentrations in central Beijing during 10:00–15:00 on January 13, 2013 (LT), due to a great amount of emission, especially over Shandong and Henan, two populous provinces with high energy consumption. From Fig. 14(d) shows a large contribution from areas outside the simulation domain (Boundary condition)

wind and temperature in wrf output file

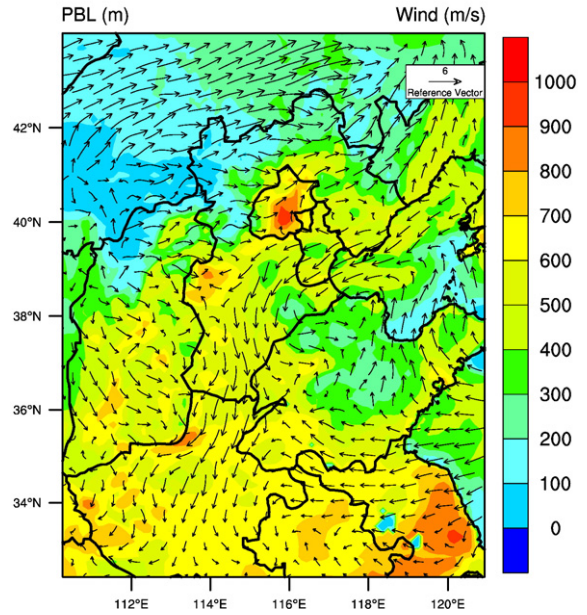


Fig. 15. Average wind velocity field and PBL distribution in the simulation domain during 10:00–15:00 on January 13, 2013.

mainly came from the south boundary which was >750 km away to the south of Beijing.

Average contribution percentage of different emission region to PM_{2.5} concentration in central Beijing for 10:00–15:00 on January 13, 2013 is depicted in Fig. 16. As mentioned before, local emissions were not the dominant contribution to PM_{2.5} concentrations at Beijing's urban center during this period. Here, the average local emission in Beijing only accounts for 37.1% of the total, while Hebei province contributed as much as 23.5%, followed by long-range transport (18.0%), Shandong (9.3%), others in D2 (6.2%), Tianjin (3.5%) and Henan (2.4%). Fig. 16(b) shows the average contribution percentage of each city in the Jing-Jin-Ji region except Beijing to PM_{2.5} concentrations in central Beijing during this period. Among the twelve cities in the Jing-Jin-Ji region excluding Beijing, Tangshan is the biggest contributor, accounting for about 6.9% of the PM_{2.5} concentration in central Beijing during this period, followed by Tianjin (3.5%), Shijiazhuang (3.2%), Cangzhou (3.0%) and Baoding (2.8%). The contribution from each of the other cities is <2.0%. Obviously, the Jing-Jin-Ji region excluding Beijing

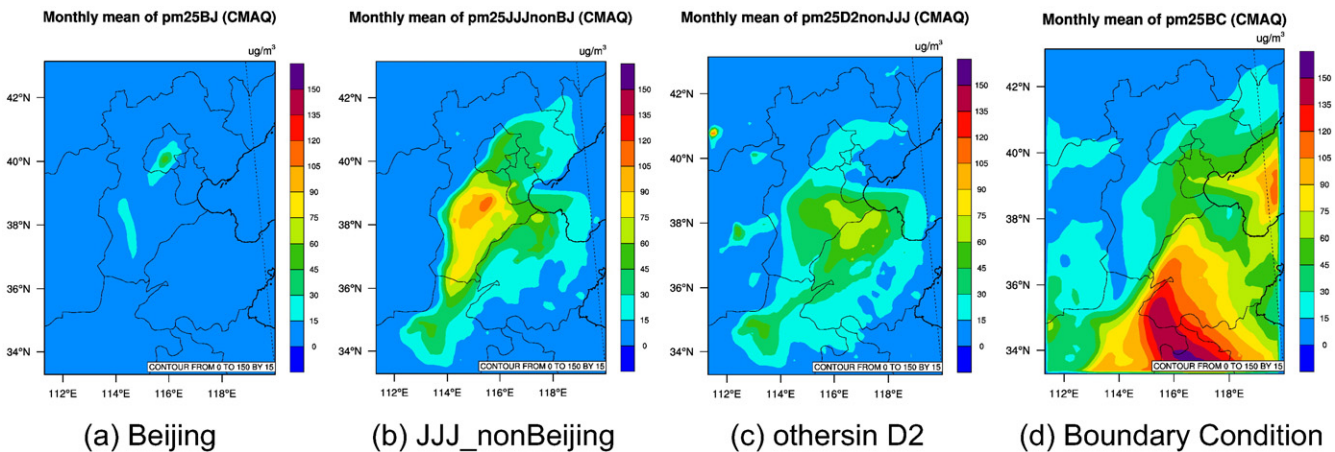


Fig. 14. Average distribution of contribution from different regions during 10:00–15:00 on January 13, 2013 (LT). Here (c) others in D2 means the region within the simulation domain and outside the Jing-Jin-Ji region.

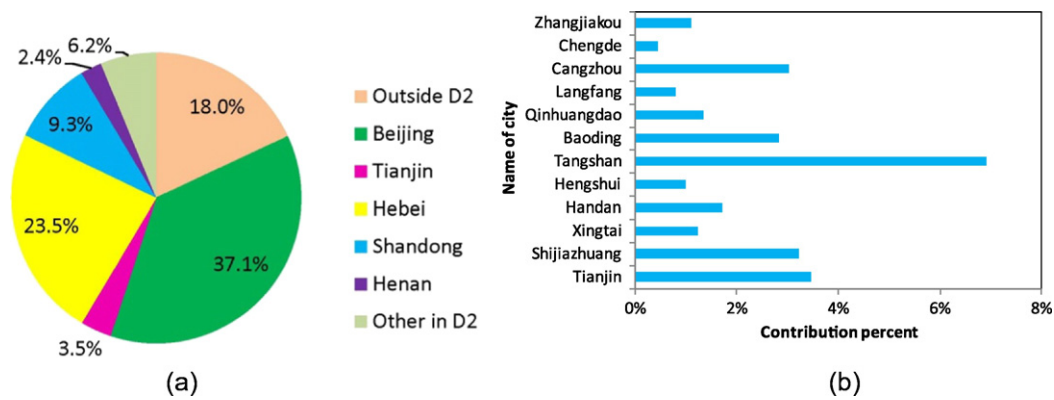


Fig. 16. Average contribution percentages of different regions for 10:00–15:00 on January 13, 2013. (a) regions include Beijing, Tianjin, Hebei, Shandong, Henan, other areas within D2 and BC (areas outside D2); (b) each city except Beijing in Jing-Jin-Ji region.

contributed significantly (27.0%) to average $PM_{2.5}$ concentrations in central Beijing during this period. To alleviate non-local-dominated haze episodes in Beijing, control of air pollution in Tangshan, Tianjin, Shijiazhuang, Cangzhou and Baoding is necessary, as well as in Shandong and Hunan provinces.

Based on simulated results in this study, when a moderate PBL height and moderate wind velocity occurred together in Beijing, pollutants from other regions can be easily transported into and easily accumulated over Beijing resulting in the formation of a non-local-dominated haze in central Beijing during January 6–23, 2013. During this non-local dominated haze episode, the $PM_{2.5}$ concentration in central Beijing was in the range of moderate high, which was much lower than the extremely high concentration of $700\mu g m^{-3}$ that occurred in Beijing on January 12, 2013, but is much higher than the limit of $PM_{2.5}$ concentration set in national ambient air quality standards. There was very few $PM_{2.5}$ concentrations which is extremely high in non-local-dominated haze episodes at Beijing's urban center compared to the local dominated haze episodes during January 6–23, 2013. In other words, during January 6–23, 2013, the extremely high $PM_{2.5}$ concentrations at Beijing's urban center was local-dominated, especially so for such abnormally high $PM_{2.5}$ concentrations as $700\mu g m^{-3}$. Compared to non-local contribution, local contribution was more easily to cause a sharp increase or sharp reduction of $PM_{2.5}$ concentration in central Beijing, and Beijing local contribution had much stronger potential to form extremely heavy haze episodes.

3.7. Contributions of cities in Jing-Jin-Ji region for different $PM_{2.5}$ polluted periods

Beijing urban suffered $PM_{2.5}$ pollution with different local contribution percent (LCP) during January 6–23, 2013, and we classify all $PM_{2.5}$ polluted periods into 5 categories, which are $LCP < 50\%$, $60\% > LCP > 50\%$, $70\% > LCP > 60\%$, $80\% > LCP > 70\%$, $LCP > 80\%$. Here, we assume that hourly $PM_{2.5}$ concentrations larger than $75\mu g m^{-3}$ is $PM_{2.5}$ polluted level, considering that $75\mu g m^{-3}$ is the limit of daily average concentration for attainment in the National Ambient Air Quality Standards of China. Fig. 17 shows average contribution percent from other every city in Jing-Jin-Ji region to $PM_{2.5}$ concentrations in Beijing urban for $PM_{2.5}$ polluted periods with different LCP categories during this modeling period. For non-local dominated haze episodes ($LCP < 50\%$), Tangshan contributes (11.2%) the biggest, followed by Tianjin (4.9%), and the other every city contributes $< 3.0\%$. For $60\% > LCP > 50\%$, Tangshan and Tianjin contribute 8.7% and 6.6%, respectively, while the other every city contributes $< 1.8\%$. For $70\% > LCP > 60\%$, Tangshan and Tianjin contribute 3.4% and 4.3%, respectively, while the other every city contributes $< 1.4\%$. For $LCP > 70\%$, the contributions from Tangshan, Tianjin and Baoding are comparable, and slight larger than those of the other cities. In terms of the contribution percent for $PM_{2.5}$ polluted episodes, twelve cities in Jing-Jin-Ji region can be classified into three groups. Tangshan and Tianjin are in the first big contribution group. And Shijiazhuang, Baoding, Langfang and Cangzhou are classified into the second big contribution group. The rest cities except Beijing in Jing-Jin-Ji region are in the third big contribution group. Fig. 17 shows that the first and second big contribution groups impact $PM_{2.5}$ concentrations in Beijing urban significantly, especially for $LCP < 50\%$ (non-local dominated) or $60\% > LCP > 50\%$. Therefore, among the twelve cities in Jing-Jin-Ji region, emission controls in Tangshan, Tianjin, Baoding, Langfang, Shijiazhuang and Cangzhou are important to reduce $PM_{2.5}$ pollution level and occurrence of haze episodes in Beijing.

4. Conclusions

To better understand the regional impact and the contribution of local emissions during heavy haze episodes in Beijing, we analyzed severe haze episodes that occurred January 6–23, 2013 to carry out a source apportionment simulation of $PM_{2.5}$ concentrations in central Beijing using CAMx with PSAT tool. Anthropogenic emission inventory used in CAMx modeling are based on 2012's emission data. The contributions from thirteen cities in the Jing-Jin-Ji region, parts of Henan and Shandong provinces, and the region outside the simulation domain as long-range transport were quantified. The formation of extremely high concentrations of $PM_{2.5}$ at certain times during the heavy haze episodes were discussed in terms of back trajectories generated by HYSPLIT model. In addition, evolution of transport contribution for local dominated and non-local dominated haze episodes were analyzed

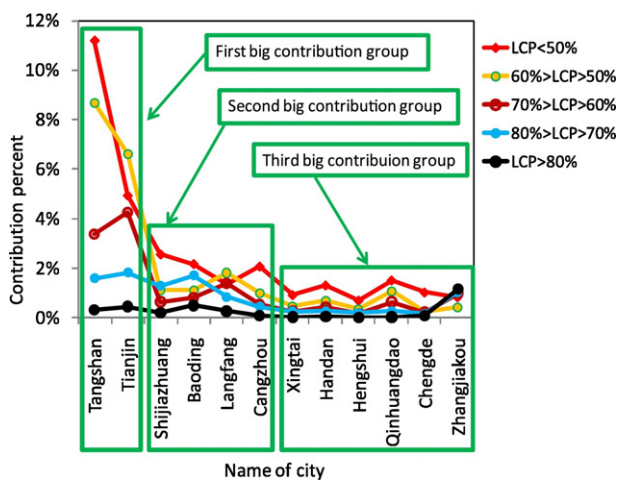


Fig. 17. Average contribution percent of each other city in Jing-Jin-Ji region to the $PM_{2.5}$ concentrations larger than $75\mu g m^{-3}$ in Beijing urban for the pollution periods with different ranges of local (Beijing) contribution percent (LCP) during January 6–23, 2013.

using the PSAT tool respectively. Finally, contribution percentages of each city in Jing-Jin-Ji region for PM_{2.5} polluted episodes with different local contribution percent in Beijing were discussed. Our conclusions are listed below.

On average, the local (Beijing) contribution to PM_{2.5} concentrations in central Beijing during January 6–23, 2013 accounted for 83.6% of the total, while regional transport from the areas within the simulation domain excluding Beijing accounted for 9.4%. Tianjin city and Hebei province contributed 1.3% and 5.2%, respectively. Long-range transport from areas outside the simulation domain contributed 7.0%. Overall, Beijing was the biggest contributor to average PM_{2.5} concentrations during the study period.

A local-dominated severe haze episode in Beijing is easily resulted from unfavorable meteorological conditions comprising a much lower PBL and a much smaller wind velocity than those that are found in non-local-dominated haze episodes. Compared to non-local contribution, local contribution is more easily to cause a sharp increase or sharp reduction of PM_{2.5} concentration in central Beijing because Beijing local has much stronger potential to form extremely heavy haze episodes. Therefore, it is most important to control local emissions to mitigate extremely heavy haze pollution such as occurred on January 12, 2013.

During a non-local-dominated haze episode in this study, the provinces of Hebei, Shandong and Henan greatly impacted PM_{2.5} concentrations in Beijing. Another big contribution came from the south boundary condition of this simulation domain, to the south of which are parts of Henan, Anhui and Jiangsu provinces, and many other distant cities. Among twelve cities in the Jing-Jin-Ji region excluding Beijing, the big contributors to PM_{2.5} concentrations in central Beijing during this non-local-dominated haze episode were Tangshan, Tianjin, Shijiazhuang, Cangzhou and Baoding.

Generally, cities in Jing-Jin-Ji region excluding Beijing can be divided into three groups in terms of the magnitude of contribution percentages to PM_{2.5} in Beijing urban for PM_{2.5} polluted episodes with different local contribution percentages. Tianjin and Tangshan can be regarded as the first big contribution group. Shijiazhuang, Baoding, Cangzhou and Langfang are the cities of the second big contribution group. And the rest cities are grouped into the third big contribution group. Overall, cities in the first and second big contribution groups impact PM_{2.5} concentration in Beijing urban significantly, especially for haze episodes.

Overall, controlling local emissions is a much more important measure to mitigate extreme haze episodes in Beijing, like that on the night of Jan 12, 2013. Emission control in Jing-Jin-Ji region, especially in Tianjin, Tangshan, Baoding, Langfang, Shijiazhuang and Cangzhou, as well as Henan and Shandong province, are important to alleviate the PM_{2.5} pollution level and reduce the occurrence of very severe haze episodes in Beijing.

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