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Assessment of inter-city transport of particulate matter in the Beijing-Tianjin-Hebei region

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Abstract

The regional transport of $PM_{2.5}$ plays an important role in the air quality over the Beijing-Tianjin-Hebei (BTH) region in China. However, previous studies on regional transport of $PM_{2.5}$ are mainly at province level, which is insufficient for the development of an optimal joint $PM_{2.5}$ control strategy. In this study, we calculate $PM_{2.5}$ inflows and outflows through the administrative boundaries of three major cities in the BTH region, i.e. Beijing, Tianjin and Shijiazhuang, using the WRF (Weather Research and Forecasting model) -CMAQ (Community Multiscale Air Quality) modelling system. The monthly average inflow fluxes indicate the major directions of $PM_{2.5}$ transport. For Beijing, the $PM_{2.5}$ inflow fluxes from Zhangjiakou (on the northwest) and Baoding (on the southwest) constitute 57% of the total in winter, and Langfang (on the southeast) and Baoding constitute 73% in summer. Based on the net $PM_{2.5}$ fluxes and their vertical distributions, we find there are three major transport pathways in the BTH region: the Northwest-Southeast pathway in winter (at all levels below 1000 m), the Southeast-Northwest pathway in summer (at all levels below 1000 m), and the Southwest-Northeast pathway both in winter and in summer (mainly at 400 – 1000 m). In winter, even if surface wind speeds are small, the transport at above 400 m could still be strong. Among the three pathways, the Southwest-Northeast happens along with $PM_{2.5}$ concentrations 30% and 55% higher than the monthly average in winter and summer, respectively. Analysis of two heavy pollution episodes in January and July in Beijing show a much stronger (8-16 times) transport than the monthly average, emphasizing the joint air pollution control of the cities located on the transport pathways, especially during heavy pollution episodes.

Key words: $PM_{2.5}$ flux; inter-city transport; CMAQ model; Beijing-Tianjin-Hebei region

1. Introduction

The Beijing-Tianjin-Hebei (BTH) region, one of the most developed regions in China, is suffering from severe pollution of particulate matter with diameter less than 2.5 μm ($PM_{2.5}$). According to the monitoring data from China National Environmental Monitoring Centre (http://www.cnemc.cn/), the average $PM_{2.5}$ concentrations of the BTH region in 2013, 2014 and 2015 were 106 μg/m^3, 93 μg/m^3 and 77 μg/m^3, respectively, which far exceeded the 35 μg/m^3 standard of China. The high $PM_{2.5}$ concentrations have adverse impacts on visibility (Zhao et al., 2011b) as well as human health (Zhang et al., 2013), and thus may cause a large economic loss

(Mu and Zhang, 2013). Therefore, it is urgent to reduce the $PM_{2.5}$ pollution in the BTH region.

Emissions from one city can substantially affect the $PM_{2.5}$ pollution in another city under particular meteorology conditions due to the transport of air pollutants. For example, some studies showed that emissions from outside Beijing can contribute 28-70% of the ambient $PM_{2.5}$ concentrations in Beijing (An et al., 2007; Streets et al., 2007; BJEPB, 2015; Wang et al., 2014). A number of approaches have been applied to evaluate the inter-city transport of $PM_{2.5}$ and its effect on local air quality. The backward trajectory, such as the HYSPLIT model (Stein et al., 2015), is one of the most commonly used methods. This method can provide the most probable transport trajectory for the air masses that have arrived at a target location; however, it cannot quantify the inter-city transport of $PM_{2.5}$. Another commonly used method is the sensitivity analysis based on Euler 3-D models, such as CMAQ (Community Multiscale Air Quality model), which is done by calculating the change in concentration due to a change in emissions. This method includes the Brute Force Method (e.g. Wang et al., 2015), the decoupled direct method (DDM, (Itahashi et al., 2012)), and the Response Surface Model (RSM, (Zhao et al., 2015)). These methods are all based on a chemical transport model, so that the physical and chemical processes can both be well considered. However, there are gaps between sensitivities and the contribution of inter-city transport because of the non-linear relationships between emissions and concentrations (Kwok et al., 2015).

Based on the meteorology field and air pollutant concentrations simulated by air quality model, the inter-city transport of $PM_{2.5}$ can be easily evaluated by the $PM_{2.5}$ flux through city boundaries. Compared to the preceding methods, the flux approach can give direct and quantitative assessment of the transport of pollutants without a heavy calculation burden. This approach has been widely applied to assess the transport of air pollutants on a large scale, such as inter-continent transport (Berge and Jakobsen, 1998; In et al., 2007). There are also studies that evaluated the pollutant transport over a regional scale (Jenner and Abiodun, 2013; Wang et al., 2009); some of which focused on the BTH region (An et al., 2012; Wang et al., 2010). In those studies, the boundaries for flux calculations are at the province level. However, in China, the air pollution control strategy is formulated and implemented at the city level. Moreover, most previous studies regarding $PM_{2.5}$ transports in the BTH region have focused on Beijing. In recent years, however, under the policy of "integrating development of BTH region", the air quality in Tianjin and cities in Hebei Province are being increasingly emphasized. Therefore, a systematic assessment of the $PM_{2.5}$ flux at the city level in the BTH region is needed.

In this study, we select Beijing, Tianjin and Shijiazhuang as target cities, and calculate the inter-city $PM_{2.5}$ transport fluxes through the administrative boundaries between the target cities and the neighboring prefecture-level cities, based on the WRF (Weather Research and Forecasting model) –CMAQ modeling system. Further the $PM_{2.5}$ source for each city and the transport pathway in the BTH region are identified based on the $PM_{2.5}$ transport flux results.

2. Methodology

2.1. Emission inventory

A multiscale emission inventory is used in this study. For the regions outside China mainland, we use the MIX emission inventory (Li et al., 2017) for the year 2010. For the China mainland other than the BTH region, we adopt a gridded emission inventory of 2012 developed in our previous study (Cai et al., 2017). For the BTH region, we develop a bottom-up emission inventory in 2012. A unit-based approach is used for power plants, iron and steel plants, and cement plants (Zhao et al., 2008). Emission factor approach is used for other sectors (Fu et al., 2013; Zhao et al., 2013b). In particular, emissions in Beijing are updated from the bottom-up inventory developed by Tsinghua University and Beijing Municipal Research Institute of Environmental Protection (BJEPB, 2010; Zhao et al., 2011a). The emissions of major pollutants by city are shown in Table 1. Methods for the biogenic emissions, the VOC speciation and the spatial and temporal allocation of emissions are consistent with our previous studies (Zhao et al., 2013a). The spatial distributions of emissions are shown in Fig. S1 (see the Supplementary Information (SI)).

2.2. WRF-CMAQ model configuration

One-way, triple nesting domains are used in WRF-CMAQ model to simulate the meteorology and air pollutant fields, as shown in Fig. 1. Domain 1 covers mainland China and part of East Asia and Southeast Asia at a grid resolution of 36 km × 36 km; Domain 2 covers the eastern China at a grid resolution of 12 km × 12 km; Domain 3 covers the BTH region at a grid resolution of 4 km × 4 km, which is the target area of this study. The simulation periods are January and July 2012, representing the winter and summer time, respectively. For the WRF (version 3.7) model, 23 sigma levels are selected for the vertical grid structure with the model top pressure of 100 mb at approximately 15 km. The National Center for Environmental Prediction (NCEP)'s

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Final Operational Global Analysis data are used to generate the first guess field with a horizontal resolution of 1°×1° at every 6 h. The NCEP's Automated Data Processing (ADP) data are used in the objective analysis scheme. The major physics options are the Kain-Fritsch cumulus scheme, the Pleim-Xiu land surface model, the ACM2 planetary boundary layer (PBL) scheme, the Morrison double-moment scheme for cloud microphysics, and the Rapid Radiative Transfer Model (RRTM) longwave and shortwave radiation scheme. The Meteorology-Chemistry Interface Processor (MCIP) version 3.3 is applied to process meteorological data into a format required by CMAQ.

We subsequently use CMAQv5.0.2 to simulate the air quality field. The CMAQ model is configured with the AERO6 aerosol module and the CB-05 gas-phase chemical mechanism. The default profile is used to generate the boundary condition of the first domain, and the simulation results of the outer domains provide the boundary conditions for the inner domains. The simulation begins five days ahead to minimize the impact of initial condition.

The model predicted meteorology and $PM_{2.5}$ concentrations are compared with observation data. The results are shown in the Supplementary Information (SI). The simulations agree well with observations, with all the indices within the benchmarks suggested by Emery et al. (2001). We evaluate simulated $PM_{2.5}$ concentrations against observations at 5 sites located in Domain 3, i.e., Beijing, Shijiazhuang, Xianghe, Xinglong and Yucheng (see Fig. 1), as shown in Table 2. The time series of simulated and observed $PM_{2.5}$ concentrations are shown in Fig. 2. It can be seen that the variation trends of $PM_{2.5}$ are well reproduced both in January and in July for all 5 sites. The average $PM_{2.5}$ concentrations are slightly underestimated in January, while the underestimation is larger in July in most sites, especially in Beijing and Xinglong. To understand the reason of the biases, it is required to evaluate the simulation results of major components of $PM_{2.5}$. Given that we have no observations of $PM_{2.5}$ components in 2012 in the BTH region, we additionally simulate the air quality in July and August in 2013, and compare with $PM_{2.5}$ component observations at several sites (see details in the SI). Generally, the underestimation of total $PM_{2.5}$ in the summer time mainly comes from the underestimation of organic carbon (OC) and sulfate. The default CMAQ tends to underestimate secondary organic aerosol to a large extent, especially in summer when photochemical reactions are active, which is a common problem of most widely used chemical transport models (Simon and Bhave, 2012; Heald et al., 2005; Zhao et al., 2016). The lack of aqueous oxidation of SO_2 by NO_2 (Wang et al., 2016), and SO_2 oxidation at dust surface (Fu et al., 2016) may partly account for the underestimation in sulfate. The underestimation in sulfate also partly explains the overestimation in nitrate. In conclusion, the biases of simulated meteorological field and $PM_{2.5}$ concentrations fall in a reasonable range. The modelling results can be used for further studies.

2.3. $PM_{2.5}$ flux calculation

The $PM_{2.5}$ flux in this study stands for the mass of $PM_{2.5}$ flowing through a particular vertical surface in a particular period of time. The vertical surface extends from the ground to a particular vertical level along the boundary of two regions (Fig. 3(a)). However, the models can only provide a three-dimensional discrete wind field and $PM_{2.5}$ concentration field. Therefore, the vertical surface through which the flux is calculated is discretized to several vertical grid cells, as is illustrated in Fig. 3(b) and detailed in the next paragraph. In this case, the expression of $PM_{2.5}$ flux can be written as

$$Flux = \sum_{i=1}^{h} \sum_{l} L H_i c \vec{v} \cdot \vec{n} \qquad (1)$$

where l is the boundary line; h is the top layer; L is the grid width; H_i is the height between layer i and $i-1$; c is the concentration of $PM_{2.5}$ on the vertical grid cell; $\vec{v}$ is the wind vector, and $\vec{n}$ is the normal vector of the vertical grid cell. The variables in the expression can be obtained from the output of the models. We choose the 9^{th} layer (about 1000 m) from the ground as the top layer in flux calculation, because most of the $PM_{2.5}$ transport between regions happens inside the boundary layer (Shi et al., 2008). Even though the transport could happen above the boundary layer, the influence of such transport on the near ground concentrations are less important because of weaker vertical mixing above the boundary layer.

Beijing and Tianjin are two most important and developed megacities in the BTH region. Shijiazhuang is the capital city, and also one of the most developed and polluted cities in Hebei province. Therefore, we choose these three cities as the target cities for flux calculation. In order to accurately distinguish the transport from different adjacent cities and to understand the net $PM_{2.5}$ inflow of a city as a whole, all the administrative boundaries between the target city and the adjacent cities are chosen as the boundary line. The boundary lines are separated to different segments by neighbor cities, and the fluxes are calculated separately for each segment of the boundary. The locations of the three target cities and their neighbors are shown in Fig. 1. Note that because there is a small area which is surrounded by Beijing and Tianjin but belongs to the city of Langfang, the boundaries between Beijing and Tianjin, Beijing and Langfang, and Tianjin and Langfang are all separated into two segments. To distinguish them, we add the relative location of the boundary to the neighbor city's name, like "Beijing (N)" and "Beijing (S)".

The flux through a boundary varies every now and then, depending on the wind direction. The polluted air mass may flow in, affect the local air quality and flow out subsequently in a short time, so that the flux may offset each other during the integration. Therefore, to characterize the intensity of interactions between two regions as well as the general impact of $PM_{2.5}$ transport, three indices are chosen in regard to the flux calculation, that is the inflow flux, outflow flux and net flux.

3. Results and discussion

3.1. Characteristics of the inter-city $PM_{2.5}$ transport in January

The monthly inflow, outflow and net fluxes through each boundary segment of the three target cities are shown in Fig. 4, from which we can get an overview of the transport in a relatively long period of time. We treat the fluxes as positive if $PM_{2.5}$ flows into the target cities, and vice versa. Therefore, the positive total net fluxes in Beijing and Shijiazhuang reveal that the $PM_{2.5}$ flows into these two cities generally exceed the flows out, and that these cities act as a "sink" of $PM_{2.5}$. This is possibly due to the unique terrain of Beijing and Shijiazhuang. These two cities are both half-surrounded by western and northern mountains, while major emissions of $PM_{2.5}$ lie to the south and east. Consequently, pollutants are easily trapped in the bulging part of the plain if there is a weak wind from the south or the east. The trapped pollutants are either scavenged by wet deposition without flowing out, or diluted by strong vertical convection due to the strong northwestern wind brought by the cold front and thus flow out of the boundary layer. In contrast, Tianjin behaves as a "source" of $PM_{2.5}$ flux. Furthermore, a probe into the detailed inflow, outflow and net fluxes through each boundary segment of the three cities may help to understand to what extent the cities interact with their neighbors. For Beijing, in winter, the inflow fluxes mainly come from Zhangjiakou (on the northwest) and Baoding (on the southwest), and the outflows go to Chengde (on the northeast) and Langfang (on the southeast) more than others. For Tianjin, Langfang (on the northwest) and Tangshan (on the northeast) contribute most of the inflow fluxes, and the Bohai sea (on the southeast) and Tangshan again receive the major outflow fluxes. Shijiazhuang acts differently from Beijing and Tianjin. The inflow and outflow fluxes through all the four boundary segments are considerably strong, with Xingtai (on the south) and Baoding (on the northeast) contributing relatively more to inflow and outflow fluxes, respectively.

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$PM_{2.5}$ fluxes may vary with height. We calculate the vertical distribution of net flux through each boundary segment to see at what level the transport mainly occurs. The results are shown in Fig. 5 (a), (c) and (e). The fluxes are shown for each vertical layer of the CMAQ model, with the approximate elevation marked to the left. Generally, the total flowing intensity is stronger at higher levels for all three cities, while the major contributor varies with layer. If we add up the net fluxes through all boundary segments (shown by the narrow bars with an envelope line), we can see that the "sink" behavior of Beijing is mainly contributed by the total net fluxes at 400 to 600 m where contribution from Baoding (on the southwest) exhibits a rapid increase with height. Similarly, total net flux for Tianjin peaks near 600 m where Tangshan (on the northeast) receives much more outflow than it does near the ground. Total net flux for Shijiazhuang peaks near 400 m where Hengshui (on the east) and Xingtai (on the south) make dominant contributions.

In order to better understand the general image of the transport characteristics in the BTH region, we use arrows to represent the net transport fluxes on a map. The result of January is shown in Fig. 6(a). The size of the arrows represents the amount of the fluxes, while white and black arrows denote fluxes at the lower (layer 1-4 in the model, from the ground to about 400 m) and upper (layer 5-9 in the model, from about 400 m to about 1000 m) layers, respectively. From the map we can identify two key $PM_{2.5}$ transport pathways in the BTH region in January: the Northwest-Southeast pathway (Zhangjiakou -> Beijing -> Langfang -> Tianjin -> The Bohai Sea) and the Southwest-Northeast pathway (Xingtai -> Shijiazhuang -> Baoding -> Beijing -> Chengde). The former one is related to the prevailing wind direction brought by winter monsoon in the BTH region, and happens at both lower layers and higher layers. The latter one happens mainly at higher layers.

According to the Ekman Spiral, wind speed is much higher at the upper level of the boundary layer (Holton and Hakim, 2012), so that pollutants can travel a longer distance during their lifetime. Assuming that the emission height of each city is similar, we believe that the higher altitude $PM_{2.5}$ flows in, the farther the source it comes from. From this point of view, the $PM_{2.5}$ flow of the Southwest-Northeast pathway at higher levels may consist of a relatively long range transport. In winter time, the southwest wind field usually occurs after the passage of a cold high pressure, when the wind speed is low and the sky is clear. Such air condition traps less upward infrared radiation at night, which helps to enhance the air stability, or even causes temperature inversion. Moreover, the southwest wind also brings moisture, leading to the formation of fog, which may enhance the aqueous reaction to form more particles. Therefore, southwest wind is usually accompanied by pollution. The Southwest-Northeast transport pathway should be intensely considered in the winter time of the

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BTH region. In contrast, the northwest wind usually comes during the passage of a cold high pressure, with relatively high wind speed both at lower and higher levels, bringing dry, cold and clean air from the non-polluted area. The large fluxes from northwest are more likely due to the strong winds rather than the high $PM_{2.5}$ levels.

3.2. Characteristics of the inter-city $PM_{2.5}$ transport in July

We conduct the same calculation in July to probe into the transport characteristics in summer. The monthly average inflow, outflow and net fluxes are shown in Fig. 4. Similar to January, total net fluxes are positive (more inflow than outflow) for Beijing and Shijiazhuang, and negative (more outflow than inflow) for Tianjin, though the magnitude is much higher than that in January. In detail, the inflow fluxes of Beijing mainly come from Langfang (on the southeast) and Baoding (on the southwest), and the outflow fluxes are mainly to Chengde (on the northeast) and Zhangjiakou (on the northwest). For Tianjin, Bohai sea (on the east) and Tangshan (on the northeast) contribute a large part of inflow, and Langfang (on the northwest) and Tangshan receive most of the outflow fluxes. The transport directions for Beijing and Tianjin in July are quite different from those in January. However, for Shijiazhuang, all of the four directions (Shanxi, Baoding, Hengshui and Xingtai) still contribute comparable amount of inflow and outflow fluxes, with slightly larger inflows from Xingtai (on the south) and Hengshui (on the east).

Fig. 5 (b), (d) and (f) display the vertical distributions of monthly average net fluxes with respect to the three cities in July. For Beijing, the total net fluxes are positive at all levels, which are different from those in January. The major contributor Baoding and Langfang show different behaviors. Net flux from Baoding is nearly zero near the ground, but increases rapidly with height, while the net flux from Langfang (including both Langfang (N) and Langfang (S)) is significant at all levels, and is largest at medium height. These phenomena are tied to the wind speed and direction at different heights in the BTH region in summer. The dominant wind direction near ground is from southeast. Within the boundary layer, the wind will rotate clockwise and become stronger with height, according to Ekman Spiral (Holton and Hakim, 2012). Langfang and Baoding are located to the southeast and southwest of Beijing, respectively. The increase of wind speed and the rotation of wind direction will constantly enhance the $PM_{2.5}$ transport from southwest, but could contribute oppositely to the transport from southeast, causing a local maximum in middle layers. For Tianjin, the overall outflow happens mainly at levels below 600 m, where the outflow flux mainly goes to Langfang. The inflow flux is dominated by the

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Bohai Sea at all heights, indicating a cross-sea transport from Shandong or other areas. The vertical distribution of net fluxes for Shijiazhuang is quite similar to that in January, except that Shanxi does not contribute a considerable amount of inflow flux any more.

We also show the general transport characteristics in the BTH region with arrows on the map, as is shown in Fig. 6(b). Compared with that in winter, the transport at lower layers becomes stronger. We can also figure out two major transport pathways in BTH in July: the Southwest-Northeast pathway (Xingtai -> Shijiazhuang -> Baoding -> Beijing -> Chengde), and the Southeast-Northwest pathway (Bohai -> Tianjin -> Langfang -> Beijing -> Zhangjiakou, and Hengshui -> Shijiazhuang). The latter pathway, which is caused by the summer monsoon, is significant at both lower and upper layers. The pathway from southwest to northeast is only obvious at upper layers. Note that in summer the vertical mixing is stronger. Therefore, although the Southwest-Northeast pathway is only active at higher levels, the transport may still affect the near-ground concentration remarkably.

If we put together the transport characteristics in winter and summer, we can see that, aside from the opposite transport pathways brought by the monsoon in different seasons, there is a steady transport pathway from southwest to northeast in the BTH region regardless of the season. This pathway has also been found in some other studies. Wu et al. (2017) analyzed the regional persistent haze events in the BTH region during 1980-2013, and found that southwestern wind field at 925 hPa (~800 m) is a typical meteorology condition. Backward trajectory studies by Zhao et al. (2017) also found a southerly transport pathway during pollution periods in the BTH region. Therefore, the Southwest-Northeast pathway is indeed important in the BTH region. The monthly transport characteristics could bring us inspiration on how the joint control of different cities should be applied. The transport pathway that happens at lower layers suggests that we should primarily control nearby low-level emission sources, while the pathway at upper layers calls for the control over a larger region to the upstream direction.

3.3. Characteristics of $PM_{2.5}$ transport during heavy-pollution episodes

Here we present the net $PM_{2.5}$ fluxes of Beijing during two heavy-pollution episodes in January and July of 2012 as examples. In January, we choose 17^{th} and 18^{th}, which are the two most polluted days (the simulated $PM_{2.5}$ daily average concentrations 211 $\mu g/m^3$ and 271 $\mu g/m^3$) throughout the month. In July, we also choose

the period with the highest concentration, i.e. 18th to 20th. The results are shown in Fig. 7 (a-b).
The magnitude of net fluxes in the two days in January (-14 kt/day and 17 kt/day) is much higher than the monthly average value. For 17th Jan, there are some weak outflows mainly to Langfang at lower levels, while stronger inflows from Baoding and Zhangjiakou occur at 300-600m. On 18th Jan, fluxes at lower level remain relatively small though the inflow and outflow directions reverse. However, strong inputs from Baoding and Langfang at above 400m become significantly strong. It can be seen that although the fluxes near the ground are small, the inflow transport can be quite strong at levels above 300 m. Coincidentally, the elevation of the mountains in the northwest of Beijing are commonly higher than 300 m, making it harder for the inflowing $PM_{2.5}$ to flow out. The large amount of $PM_{2.5}$ inflows can only be efficiently blown out to the northeast direction (Chengde, Langfang (N) and Tianjin (N)). These results are consistent with Jiang et al. (2015), who also found a strong southerly input at a high level during a haze episode in winter. Therefore, the Southwest-Northeast pathway is of great importance during this heavy pollution period.
For the day with the highest concentration in July (July 20th), the vertical distribution does not show much difference from the average of July (Fig. 5 (b)), except for the magnitude. The fluxes are about 1/5 of the monthly average, or less than 1/10 of that in the heavy-pollution period in January. This result suggests that the heavy pollution in Beijing in 20th July is not dominated by the inter-city transport during that very day. However, situations are totally different on 18th July (Fig. 7 (c)), the first day when the simulated $PM_{2.5}$ concentration reaches a high level in this pollution episode. The magnitude of fluxes is about 6 times larger than the monthly average, or some 30 times larger than that on 20th July. More importantly, the outflow flux is much smaller than the inflow flux contributed mainly by Baoding and Langfang, which correspond to the Southwest-Northeast and Southeast-Northwest pathways respectively. Therefore, we can draw an image about how the $PM_{2.5}$ transport affects the air quality in Beijing during this pollution episode. On 18th July, the $PM_{2.5}$ start to flow into Beijing through the Southeast-Northwest and Southwest-Northeast pathways with a very strong flux, but very few of them flow out, causing the accumulative increase of $PM_{2.5}$ concentration. On 20th July, the wind field become stable and the transport weakened, but the $PM_{2.5}$ that have flowed in before accumulate to form the heavy pollution. This result indicates that both the Southeast-Northwest and the Southwest-Northeast pathway are important for Beijing during this polluted period, and the emission from outside Beijing should be controlled at least 2 days in advance to reduce the peak concentration.
From the discussions above, we can see that $PM_{2.5}$ transport plays an important role in the heavy-pollution

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periods in Beijing. We further analyze the $PM_{2.5}$ flux data of the three cities day by day, and try to identify the presence of transport pathway for each day in Beijing, based on whether the inflow flux from a certain direction is significantly larger than the others. Finally, 8 days in January and 4 days in July are subject to the transport of Southwest-Northeast pathway, 22 days in January are subject to the transport of Northwest-Southeast pathway, and 8 days in July are subject to the transport of the Southeast-Northwest pathway. In July, there are other 8 days subject to both the Southeast-Northwest pathway and the Southwest-Northeast pathway ("SE-NW + SW-NE" for short). Moreover, some days do not show a clear transport direction, which are referred to as "unclassifiable days". We calculate the average simulated concentration for each transport pathway. The results are shown in Table 3.

The days with Southwest-Northeast pathway shows the highest $PM_{2.5}$ average concentrations among all days in both January and July. Therefore, the Southwest-Northeast pathway should be the focus of control strategies. In contrast, the Northwest-Southeast pathway tends to happen along with the lowest concentrations in both seasons. Note that in January, the day with the highest concentration (January 19th) is coincidentally identified as the Northwest-Southeast pathway. That day is on the eve of the rapid clearing by the northwest wind (Fig. 2(a)). While the cold front is passing, the heavy polluted air mass is forced to move from northwest to southeast, which cause a significant transport. However, since the pollution brought by such transport usually happen with a strong cold front, the $PM_{2.5}$ concentration will soon become very low (Jia et al., 2008). If we exclude January 19th from the Northwest-Southeast pathway days, the average concentration will be only 48.5 $\mu g/m^3$. In July, the Southeast-Northwest pathway and the Southwest-Northeast pathway happen simultaneously for 8 days. The average concentration is 47.4 $\mu g/m^3$, the second highest in July, which further emphasizes the importance of the transport from the southwest. In summary, the Southwest-Northeast pathway should be taken great consideration both in January and July, followed by the Southeast-Northwest pathway in July.

4. Conclusions

By calculating $PM_{2.5}$ inflow and outflow fluxes through the boundaries between each two prefecture-level cities, this study has shown the major $PM_{2.5}$ input and output directions in winter and summer for Beijing, Tianjin, and Shijiazhuang. For Beijing, the inflow fluxes mainly come from northwest and southwest in winter, and southeast and southwest in summer. For Tianjin, the inflow fluxes are mostly from northwest and northeast in winter, and east and northeast in summer. In Shijiazhuang, however, the four neighboring regions contribute comparable amount of inflow fluxes both in winter and summer.

By analyzing the net $PM_{2.5}$ fluxes and their vertical distribution, we identify several major transport pathways and the height they occur: the Northwest-Southeast pathway in winter (at all levels below 1000 m, but stronger at levels above 400 m), the Southeast-Northwest pathway in summer (at all levels below 1000 m), and the Southwest-Northeast pathway both in winter and in summer (at levels between 400 m and 1000 m). Although the third pathway does not happen as frequently as the other two in corresponding seasons, it is accompanied by quite high $PM_{2.5}$ concentrations in both seasons. Additionally, the relatively large transport height of this pathway suggests the importance of the long-range transport of $PM_{2.5}$ on air quality. Specially, in winter, even if the wind speed near the ground is low, which we often refer to as "steady" conditions, the transport above 400 m, which is primarily associated with long-range transport, could still be strong. These findings suggest that the joint control for cities on the Southwest-Northeast pathway should be emphasized both in winter and summer.

We also find that inter-city transport during heavy-pollution episodes could be stronger than the monthly average, at least for the two polluted periods investigated in this study. In the heavy pollution episode in summer, $PM_{2.5}$ flows into Beijing and accumulates for two days, leading to a heavy pollution. Therefore, mitigating emissions from a larger area may be essential for the control of ambient $PM_{2.5}$ in Beijing. Moreover, it appears important to control the upstream sources several days ahead to mitigate the $PM_{2.5}$ accumulations, rather than only taking actions when the pollution is already heavy. However, we must note that the two episodes we studied may not represent the general characteristics of heavy-pollution episodes, which requires a more systematic analysis in the future.

The current study has several limitations. We only quantify the $PM_{2.5}$ transport at the outer boundary of the city, ignoring the inter-city transport of gaseous precursors that may be converted to secondary $PM_{2.5}$ in the target city. Additionally, we cannot distinguish whether the $PM_{2.5}$ inflows through the administrative boundary come from the neighbor city or upstream areas. Future studies may combine the flux calculation with the tracer or tagging methods to overcome these defects. Despite these limitations, the flux approach has indeed proved to be a powerful tool to visually assess the inter-city transport of pollutants.

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Table 1 Summary of the emissions of major pollutants in Beijing, Tianjin and 11 prefecture-level cities in Hebei in 2012

Emissions (kt/year)	NOx	SO_2	$PM_{2.5}$	PM_{10}	BC	OC	NMVOCs	NH_3[a]
Beijing	202	120	75	177	9	9	381	52
Tianjin	392	287	113	151	17	26	287	45
Hebei	1620	1079	875	1172	141	221	1346	628
Shijiazhuang	270	198	149	203	23	33	230	87
Chengde	84	45	37	49	6	10	56	34
Zhangjiakou	112	52	41	54	7	11	56	35
Qinhuangdao	71	39	30	40	5	8	51	22
Tangshan	266	145	100	135	15	24	181	68
Langfang	79	71	63	86	10	14	100	35
Baoding	158	123	118	155	20	33	202	89
Cangzhou	149	121	109	148	17	25	164	67
Hengshui	79	66	62	84	10	15	92	50
Xingtai	140	105	77	102	13	21	113	60
Handan	213	115	89	117	15	26	148	82

481 **Table 2 Comparison of the simulated and observed $PM_{2.5}$ concentrations at five sites.**

Indices		**Mean OBS**	**Mean SIM***	**NMB**	**NME**
Unit		$\mu g \cdot m^{-3}$	$\mu g \cdot m^{-3}$	%	%
January, 2012	Beijing	86.0	65.2	-24.2	32.2
	Shijiazhuang	193.9	170.8	-11.9	45.3
	Xianghe	132.3	85.6	-35.3	44.5
	Xinglong	39.4	38.6	-2.0	42.7
	Yucheng	140.9	124.1	-11.9	31.2
July, 2012	Beijing	68.2	35.6	-47.8	49.5
	Shijiazhuang	70.3	79.8	+13.6	37.6
	Xianghe	61.3	47.2	-23.0	35.6
	Xinglong	48.9	24.6	-49.6	53.8
	Yucheng	77.3	55.2	-28.6	39.6

482 *Average of the days only when observations are available.

483

Table 3 The mean and maximum simulated $PM_{2.5}$ concentrations in Beijing for all days in January and July and for the days that belong to particular transport pathways.

Month	Pathway type	Days	Mean $PM_{2.5}$ conc in Beijing, μg/m^3	Max $PM_{2.5}$ conc in Beijing, μg/m^3
Jan	All days	31	65.2	270.7
	Southwest-Northeast	8	85.1	211.5
	Northwest-Southeast	22	58.6	270.7
	Unclassifiable day(s)	1	53.0	53.0
Jul	All days	31	35.0	94.4
	Southwest-Northeast	4	54.2	94.4
	Northwest-Southeast	5	15.4	30.9
	Southeast-Northwest	8	29.4	53.2
	SW-NE + SE-NW	8	47.4	71.0
	Unclassifiable day(s)	9	29.3	79.7

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Figure Captions

Figure 1. The simulation domains used in this study (left) and the map of the Beijing-Tianjin-Heibei region (right). The highlighted cities are the target cities for flux calculation. The red circles show the sites with $PM_{2.5}$ observations. The two sites with green circles have observations of $PM_{2.5}$ chemical components in 2013.

Figure 2. Time series of the simulated and observed PM2.5 concentrations in (a) Beijing, (b) Shijiazhuang, (c) Xianghe, (d) Xinglong, and (e) Yucheng.

Figure 3. An example of the vertical surface for flux calculation (a) before discretization, and (b) after discretization.

Figure 4. The inflow, outflow and net fluxes in January and July for (a) Beijing, (b) Tianjin, and (c) Shijiazhuang.

Figure 5. Vertical distribution of net fluxes in January (left) and July (right) for (a-b) Beijing, (c-d) Tianjin, and (e-f) Shijiazhuang

Figure 6. The transport fluxes through each boundary segment of the three target cities in January (left) and July (right). The size of the arrows represents the amount of the fluxes, while white and black arrows denote fluxes at the lower (layer 1-4 in the model, from the ground to about 400 m) and upper (layer 5-9 in the model, from about 400 m to about 1000 m) layers, respectively.

Figure 7. PM2.5 fluxes during two heavy-pollution days in Beijing in January and July: (a) January 17th, (b) January 18th, (c) July 18th and (d) July 20th.

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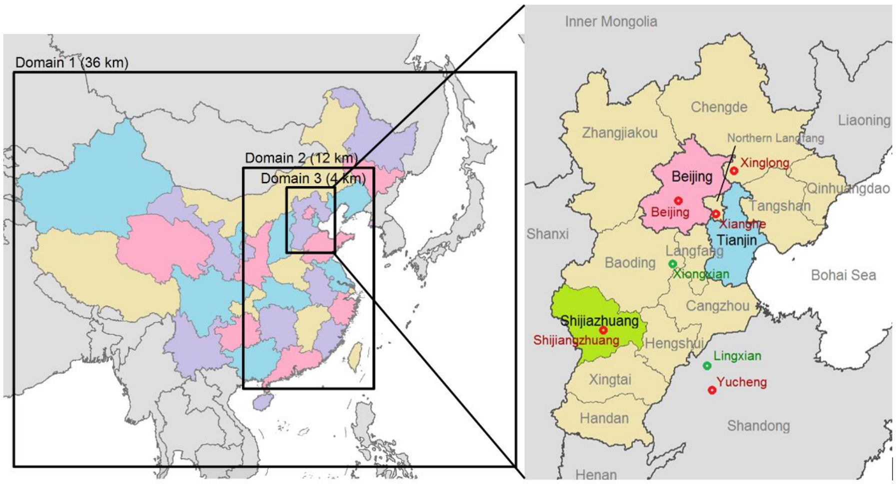

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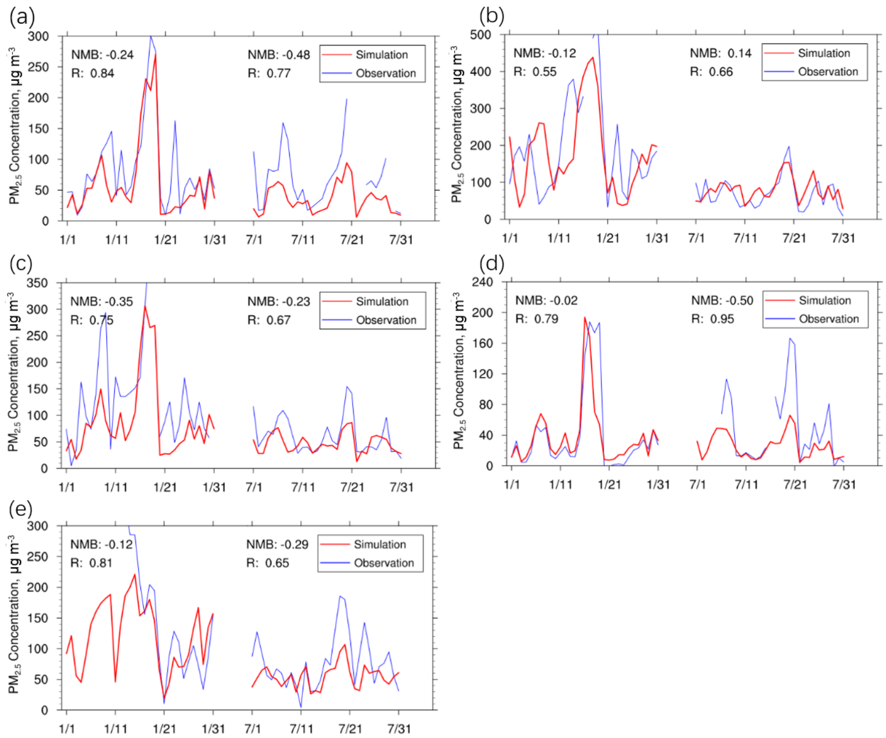
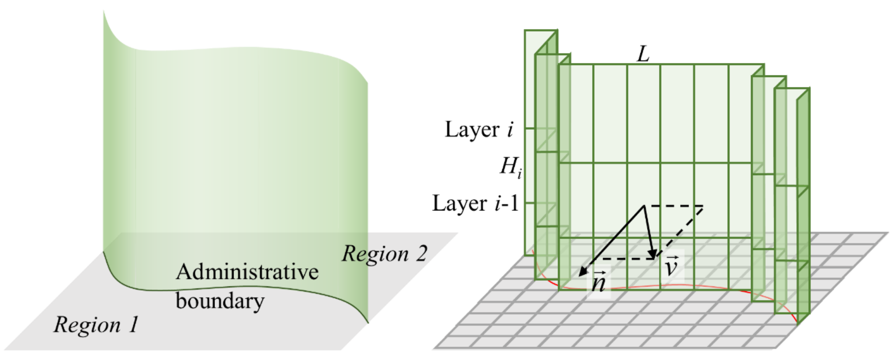


Figure 2 Time series of the simulated and observed $PM_{2.5}$ concentrations in (a) Beijing, (b) Shijiazhuang, (c) Xianghe, (d) Xinglong, and (e) Yucheng



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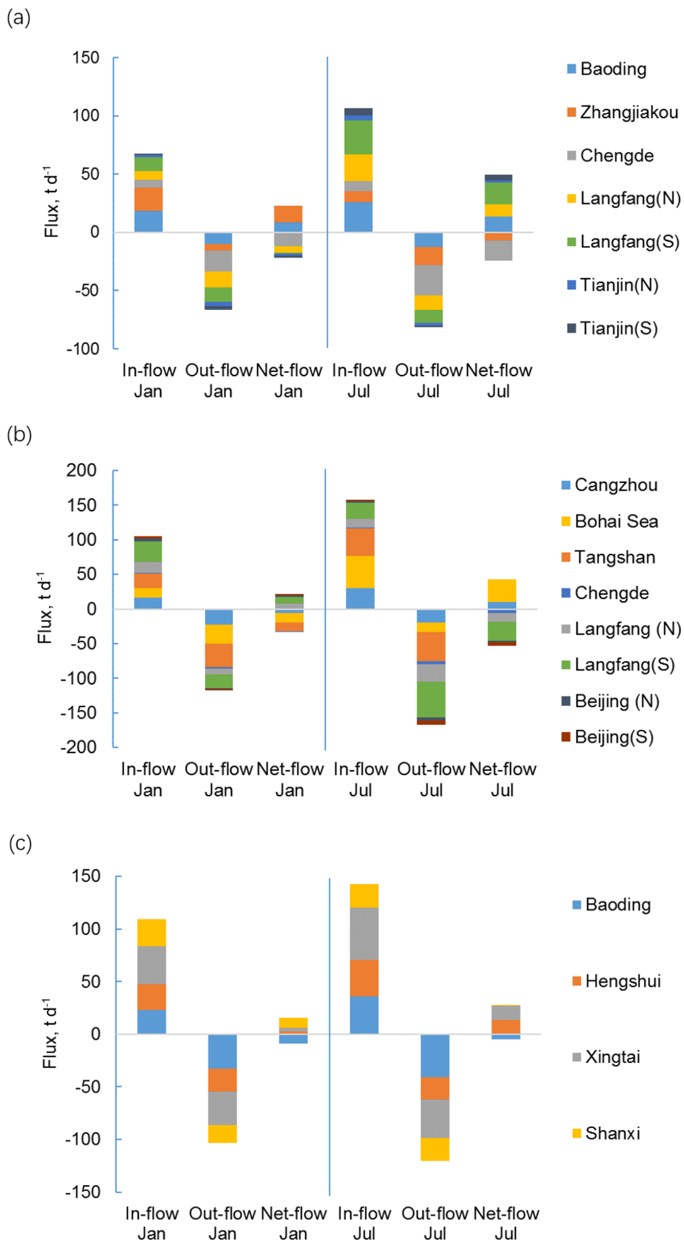

521

522 **Figure 4 The inflow, outflow and net fluxes in January and July for (a) Beijing, (b) Tianjin, and (c)**
523 **Shijiazhuang**

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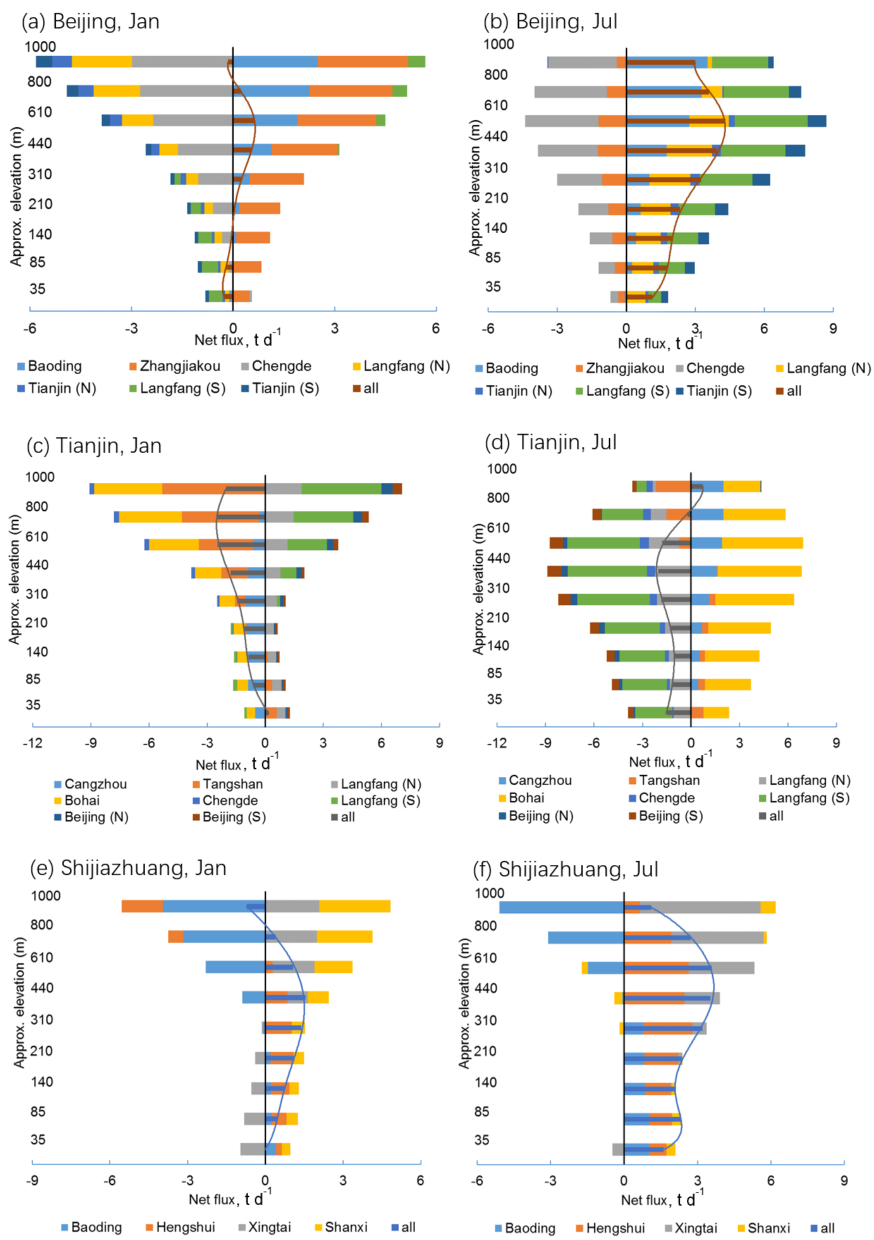

Figure 5 Vertical distribution of net fluxes in January (left) and July (right) for (a-b) Beijing, (c-d) Tianjin, and (e-f) Shijiazhuang

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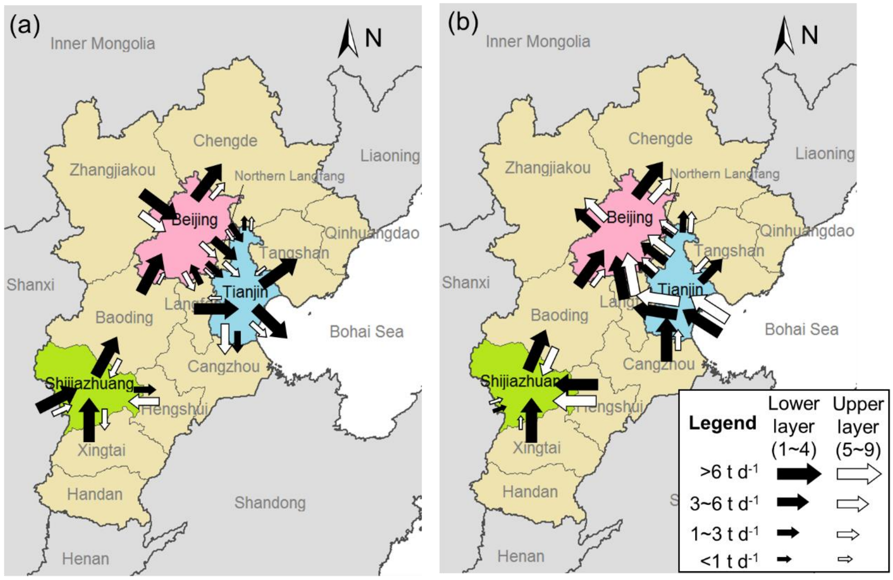
529 

530 **Figure 6 The transport fluxes through each boundary segment of the three target cities in January (a)**
531 **and July (b). The size of the arrows represents the amount of the fluxes, while white and black arrows**
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533 **9 in the model, from about 400 m to about 1000 m) layers, respectively.**

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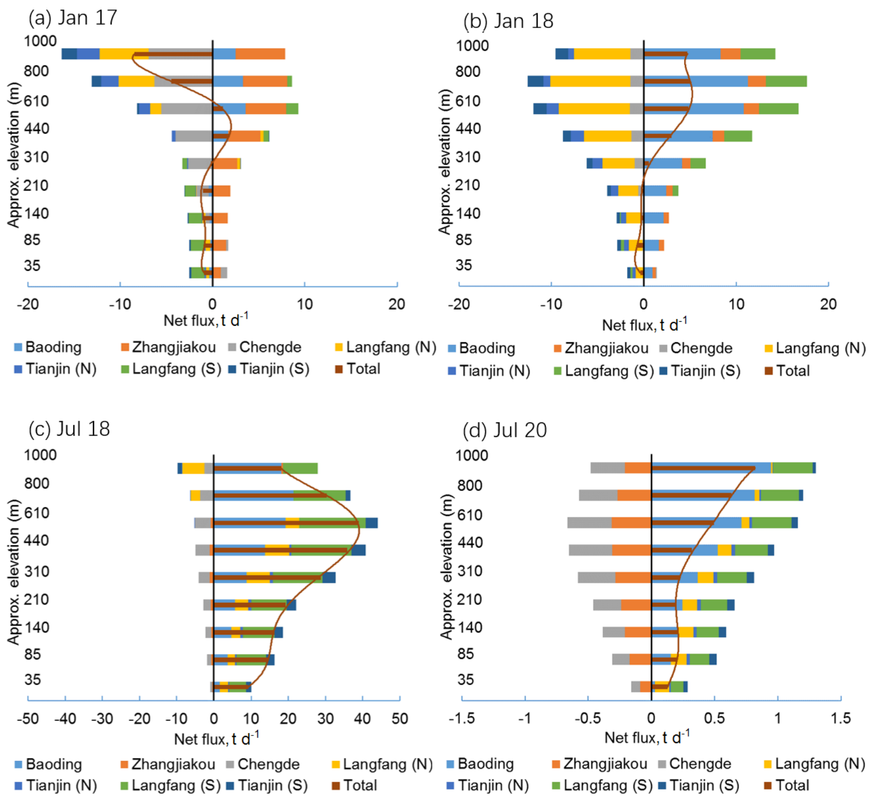

Figure 7 $PM_{2.5}$ fluxes during two heavy-pollution days in Beijing in January and July: (a) January 17th, (b) January 18th, (c) July 18th and (d) July 20th.