RC1:

This paper applied the WRF-CMAQ model to simulate the air quality over the Beijing-Tianjin-Hebei area and calculate the trans-boundary fluxes to Beijing, Tianjin, and Shijiazhuang. This paper used a new method that assessed the pollutants transport by vertical surface flux calculation instead of usually used scenario analysis.

Author's reply: We feel encouraged to receive the reviewer's recognition for our work. We treasure the valuable comments raised by the referee and have followed them in revising the manuscript. Please check the below for the point-to-point responses.

(1) Using this method, it is easily to understand the pollutants inflows from each direction or each surrounding city to the objective city. But the inflows from one city didn't necessarily mean those pollutants were from that city. It might be generated from the upflow cities. Did the authors consider about that? And is there any consideration about solving this?

Authors' reply: We appreciate for the valuable comment. Indeed, the transport flux itself could not tell whether the pollutants are from the neighbor city or from other upstream cities, and this problem is one of the major disadvantages of the flux method. However, the goal of using the flux method is mainly focused on the transport from different directions, rather than the contribution of different cities. By putting together the transport characteristics of different cities as is done in our study (see Fig. R1), we have obtained a general transport feature in the BTH region, which can facilitate a qualitative understanding of where the fluxes are mainly from.

As shown in Fig. R1, in January, the $PM_{2.5}$ that flows into Beijing from Baoding on the southeast mainly happens at a higher level, so the $PM_{2.5}$ may origin from a larger area upstream. Then we can track backward along the dark arrow, and the inflow may come from Baoding, Shijiazhuang or even Xingtai. In July, the transport directions between Baoding and Shijiazhuang are different at the lower and the upper level. We can infer that the inflowing $PM_{2.5}$ into Shijazhuang mainly come from Baoding rather than farther regions to the northeast.

We admit that the flux approach cannot quantitatively evaluate the contribution from each city, and we hope that future studies can combine the flux method with other methods such as tagging models to quantitatively assess the contribution of different cities or regions on the transport pathways identified in the current study.

We have included the preceding discussions on the limitations in the revised manuscript. (Page 15, Line 401-407)

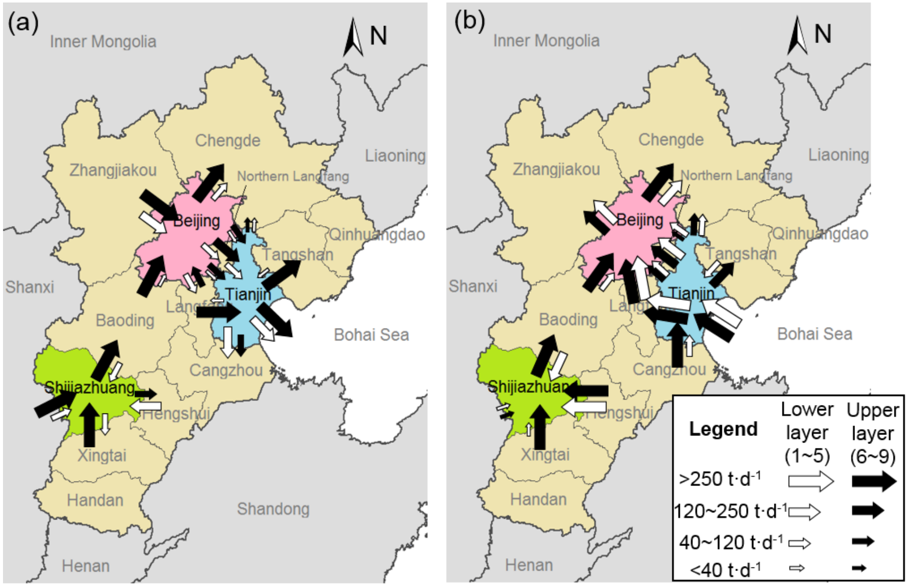

Figure R1 The transport fluxes through each boundary segment of the three target cities in January (a) and July (b). The size of the arrows represents the amount of the fluxes, while white and black arrows denote fluxes at the lower (layer 1-5 in the model, from the ground to about 310 m) and upper (layer 6-9 in the model, from about 310 m to about 1000 m) layers, respectively.

(2) Wind directions are very important to calculate the pollutants fluxes (Figure 3 and Equation 1). But in the model evaluation section (S2), the authors only evaluate the wind speed, temperature, and humidity. I would suggest the authors to evaluate the wind direction in the simulation results.

Authors' reply: We thank the referee for the good suggestion. We evaluated the wind direction using the same method as the other meteorology parameters. The results are shown in Table R1. The bias of wind direction at 10m (WD10) falls within the benchmark, but the gross error exceeds the benchmark for both January and July. The larger gross error is partly caused by the lower precision of the observation data. The WD10 observations only have 16 different values, while the simulation could have any value between 0 and 360. For example, if the real WD10 is 125 degree, the value will be reported as 140 degree. Even if the simulation is exactly 125 degree, an additional gross error of 15 degree will be introduced. In addition, compared to other similar simulation studies in China (e.g., Hu et al., 2016; Zhao et al., 2013), the gross error of WD10 falls in a similar range. Therefore, we still believe that the simulated meteorology field shows a reasonable agreement with the observation.
We have added the data and the discussion above in our SI (Page 3, Line 37 – 50).

Table R1 Comparison of simulated and observed wind direction

Parameter	Index	Unit	Benchmark[a]	Jan-2012	Jul-2012
Wind direction (WD10)	Observation Mean	deg	-	203.9	175.2
	Simulation Mean	deg	-	222.4	174.8
	Bias	deg	≤±10	-2.64	-1.47
	Gross error	deg	≤30	43.23	43.7

a. The benchmarks used in this study are suggested by Emery (2011).

RC2:

This paper analyzed the flux flow between cities in BTH area, northern plain in China, with a commonly used transport model WRF-CMAQ. It is an important issue for policy makers to understand the regional transport of air pollution, and would be helpful in decision of emission control strategy. The paper is clearly written and easy to follow. I suggest its publication when the following issues are further stressed or discussed.

Author's reply: We appreciate the reviewer's valuable comments which help us improve the quality of the manuscript. We have carefully revised the manuscript according to the reviewers' comments. Below is our point-to-point responses to the issues raised by the reviewer.

(1) Language. There are some grammar errors in the manuscript and the language should be polished.

Authors' reply: We sincerely apologize for the deficiency in language. We have gone through the text carefully and corrected the grammar errors.

(2) Lines 56-57, Page 3. It is not quite persuasive, since the non-linear relationship is considered in the DDM and RSM methods.

Authors' reply: We thank the referee for pointing out the problem. The expression in the manuscript was not quite accurate and might cause misunderstanding. We believe that all methods based on chemical transport models, including the DDM and RSM methods, are able to consider the non-linear relationship between emissions and concentrations. However, the sensitivity of $PM_{2.5}$ concentrations to emission perturbation, which DDM and RSM aim to quantify, is different from the inter-city transport of $PM_{2.5}$. Assuming that the emission reduction in the source region leads to a 30% reduction in $PM_{2.5}$ concentration in the target region, the transboundary transport of $PM_{2.5}$ may not be 30% because of the nonlinearity in the emission-concentration relationships.

Zhao et al (2017) found that during winter time, the $PM_{2.5}$ could response negatively to NOx reduction. Therefore, if we use sensitivity approach such as brute-force method,

DDM and RSM to assess the regional transport of $PM_{2.5}$, the result may deviate (probably underestimate) from the real case.
We changed our expression in the manuscript for a better understanding of the insufficiency of the DDM and RSM methods for our study (Page 3, Line 55-57).

(3) Lines 128-129, Page 6. The authors stated that the biases of simulated meteoro logical field and $PM_{2.5}$ concentrations fall in a reasonable range. For meteorological field, the statement could be supported by Table S1, with the evidence by Emery et al., 2001. For $PM_{2.5}$ concentrations, however, we could not think it is "reasonable" as no further information is given. The bias could be quite large in some case, and some major components such as SOC were largely underestimated as indicated by the authors. Therefore, I suggest that the authors provide the evidence or criterion to justify the model performance., or describe the current simulation progress (model performance) in BTH region.

Authors' reply: We thank the referee for the valuable suggestion. We calculated additional indices including Mean Fractional Bias (MFB) and Mean Fractional Error (MFE), and compared them with the benchmark values suggested by Boylan and Russell (2006). The definition of the two indices can also be found in the research of Boylan and Russell (2006). The simulation results of the $PM_{2.5}$ concentration are well within the model performance criteria. We have added the new statistical results as well as the benchmark values in Table 2 in the manuscript.
Due to the limitation of monitoring sites, the benchmark is not applicable for the evaluation of $PM_{2.5}$ components. Therefore, we compare the model performance with other studies in the BTH region. The results are summarized in Table R2. All of the studies underestimate the sulfate concentrations. The underestimation ranges between 9% and 79%, and most of them are larger than 30%. The nitrate simulation results vary in different studies, but the majority of the studies tend to overestimate its concentration. The concentration of EC is usually much lower than the other four components, which may contribute to the large discrepancy in the simulation results in different studies. For OC, although some studies overestimate the concentration, more studies exhibit a lower concentration than observation. Generally speaking, the biases of the $PM_{2.5}$ components in the current study have similar magnitude to other recent studies in the BTH region.
We have included the discussion in the revised SI (Page 7, Line 69-80)

Table R2 Summary of the $PM_{2.5}$ component simulation results for the BTH region in recent studies

Time	Site	SO_4^{2-} NMB (%)	NO_3^- NMB (%)	NH_4^+ NMB (%)	EC NMB (%)	OC NMB (%)	Reference
2005 annual	Tsinghua, Beijing	-14	13	10	-24	-36	Wang et al., 2011
2005 annual	Miyun, Beijing	-36	62	9	-17	-52	Wang et al., 2011
14 Jan – 8 Feb, 2010	Beijing	-72	-32	-5	124	26	Liu et al., 2016
14 Jan – 8 Feb, 2010	Shangdianzi, Beijing	-78	-24	-13	36	-7	Liu et al., 2016
Jan, 2010	Peking university, Beijing	-39	85	33	101	-2	Liu et al., 2016
14 Jan – 8 Feb, 2010	Shijiazhuang, Hebei	-79	-35	-7	81	38	Liu et al., 2016
14 Jan – 8 Feb, 2010	Chengde, Hebei	-78	48	-10	-39	-50	Liu et al., 2016
14 Jan – 8 Feb, 2010	Tianjin	-72	0	9	149	85	Liu et al., 2016
11-15 Jan, 2013	Beijing	~ -73	~ -43	-	-	-	Wang et al., 2014
Jan 2013	Handan, Hebei	-9	33	-11	50	37	Wang et al., 2015
Jul 2013	Handan, Hebei	-32	-3	8	96	30	Wang et al., 2015
Oct – Nov, 2014	7 sites in the BTH region	-48	16	-25	87	-37	Zhao et al., 2017
22 Jul – 23 Aug, 2012	Xiong County, Hebei	-52	95	2	120	-25	This study
22 Jul – 23 Aug, 2012	Ling County, Shandong	-57	79	-14	117	-1	This study

(4) Section 3.2, Page 9. The authors described the difference in flux pattern between Jan and July. However, the reasons for the difference is not further discussed, and the seasonal mechanisms in pollution transport remained unclear. More information should be provided here.

Authors' reply: We appreciate for the valuable comment. What determine the seasonal transport flux are mainly two factors, the wind speed and the $PM_{2.5}$ concentration in the upstream areas. The $PM_{2.5}$ concentration is related to both the meteorology condition and the emission, with the upstream emissions being the most important factor. If we understand the roles of emissions and winds in the transport, we can answer the question of the seasonal mechanisms. Therefore, we combined the response of this comment with the fifth one below.

(5) Related with Q4, the paper described the pattern of pollution transport between cities, which is helpful for policy making. For scientific issue, however, the main factors influencing the transport were not sufficiently discussed. Could the author explain the roles of emissions and meteorological condition on the transport using the cases presented in the paper?

Authors' reply: We combine the response of this comment with the fourth one. To better understand how the wind and concentration affect the transport fluxes, we have made several wind rose plots for different cities, different seasons and different heights. Besides the traditional wind rose plot that displays wind direction with wind speed frequencies, we also made plots that display the wind direction with $PM_{2.5}$

concentration frequencies. We chose the ground layer and the 7th layer to represent the lower layer and the upper layer respectively. The plots for Beijing are shown in Fig. R2 as an example. The plots for the other two cities are displayed in the SI (Fig. S3 and Fig. S4).

In January, the dominant wind directions near the ground ranges from northwest to northeast. The NNE wind has the highest frequency, while the NW wind has the highest wind speed (Fig. R2(a)). The dominant northern winds reflect the winter monsoon. Although the concentration coming with the northern winds are relatively low because of the low emission rate on that direction (Fig. R2(b)), the high frequency and wind speed also cause an overall strong transport from the northwest to the southeast. Wind directions and the corresponding concentrations are quite different at the upper layers (Fig. R2(c), (d)). The prevalent northern wind remains (though the dominant directions shift slightly from NNE to NW), and the frequency of southwestern winds is much higher than that at lower layers. Moreover, the $PM_{2.5}$ concentrations that come with southwestern winds are much higher than the other directions. The strong emission in southern Hebei (which lies on the southwest direction of Beijing), especially the elevated sources may be responsible for the high concentration from the southwest. Therefore, in January, the dominant northwestern winds account for the Northwest-Southeast pathway at both lower layers and upper layers, while the large emissions on the southwest direction mainly caused the Southwest-Northeast pathway at upper layers.

In July, the dominant wind directions at the lower layer are the southeastern directions, reflecting the summer monsoon (Fig R2(e)), and coincidentally the highest concentrations also come along with the southeastern winds (Fig R2(f)). Emissions from Tianjin, Langfang, and Tangshan may influence Beijing by the southeastern winds. The emission and the wind direction both contribute to the Southeast-Northwest pathway at the lower layers. The high frequency wind directions shift clockwise to the southern directions at the upper layers in July, as is shown in Fig. R2(g), and the southwest wind and the southeast wind are both important. Moreover, the directions with high concentrations also shift to both the southwest and the southeast directions (Fig. R2(h)). Therefore, in July, the dominant southeastern winds and the emissions on the southeast directions caused the Southeast-Northwest pathway at both the upper and the lower layers. The Southwest-Northeast pathway is a combination result from the southern winds and the emissions, which is different from that in January.

Similar analysis can be made to the plots for the other two cities. Due to the length limitation, we put the plots into the SI. The plots in Fig. R2 and the discussions above has been included in our revised manuscript (Page 10-11, Line 262-287).

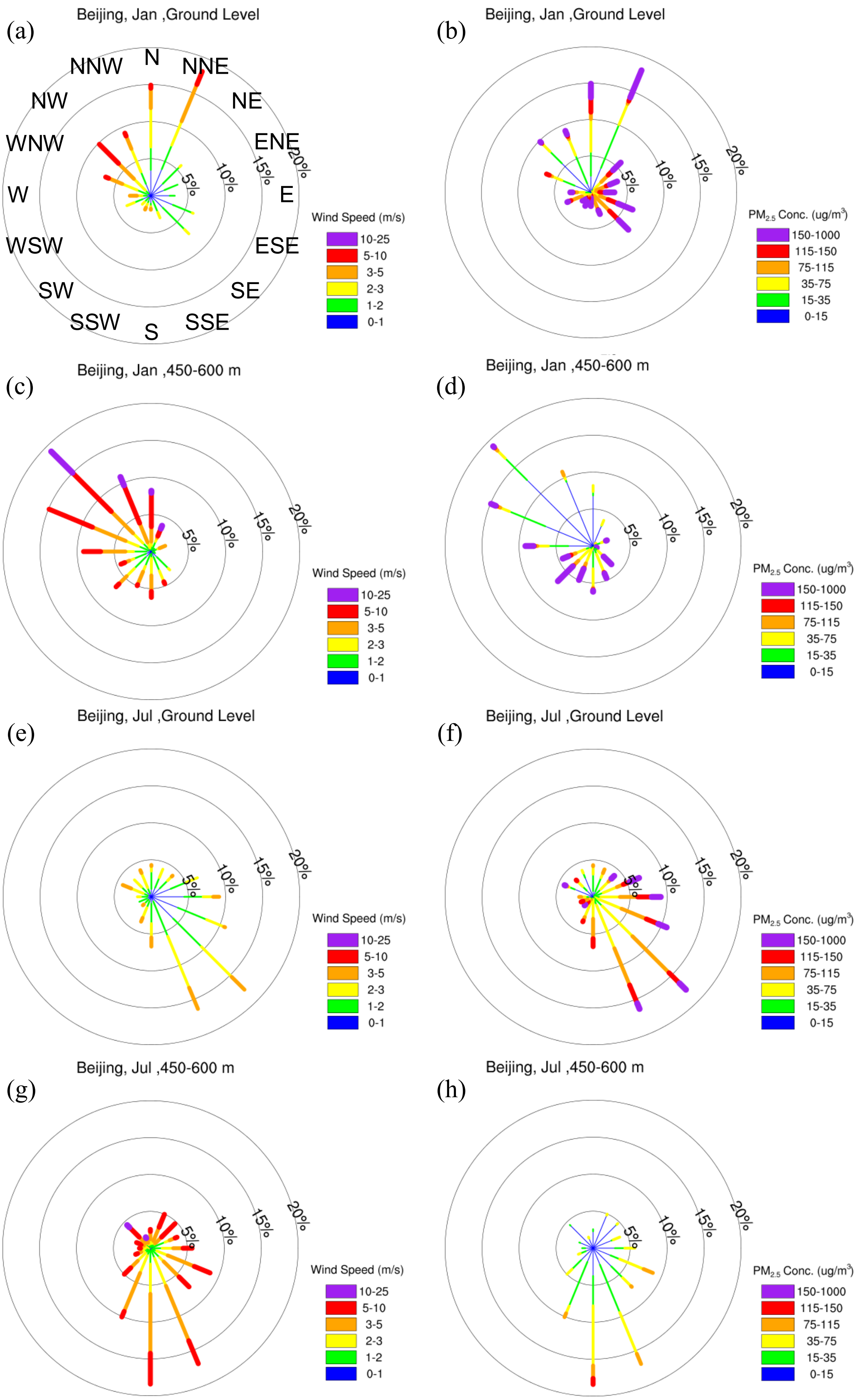

Figure R2 The wind rose plots showing the frequency of wind speed (a, c, e, g) and $PM_{2.5}$

concentration (b, d, f, h) at different wind directions for Beijing. The ground level and the 7th level (about 450-600 m) in the model are chosen to represent lower levels and upper levels. The percentages denote the frequency.

SC1:

The manuscript is meaningful for the prevention and control of regional pollution in north China. It is absolutely worth of publishing as the study itself is extremely interesting. However, some improvements are suggested.

Authors' reply: It is our honor to receive the valuable comments from Dr. Tang. We have revised the manuscript carefully according to these comments. Please see below for our point-to-point responses.

In the manuscript, the authors found the southwest-northeast transport pathway. Actually, it is the most important pathway in North China Plain, especially during the heavy polluted episodes. Tang et al. (2015) and Zhu et al. (2016) found aerosols transported from the southwest between 500-1200 m (in the upper boundary layer) using ceilometer observations, which were the same with your simulations. However, the transport just emerged during the initial periods of the heavy pollution episodes. With the in crease of the aerosols, the PBL decreases (below 500m) and the transport effects weaken during the heavy polluted periods. Could you please quantify the transport in different pollution degrees?

Authors' reply: The reviewer raised a very useful question. The transport fluxes vary with different meteorology conditions in different days. Following this suggestion, we calculated the flux for individual days in January and July, and sorted the data into groups based on different pollution levels (see Fig. R3). Taking Beijing as an example, in January, the simulated concentration ranges from 11 $\mu g/m^3$ to 271 $\mu g/m^3$, while in July, the range is from 6 $\mu g/m^3$ to 94 $\mu g/m^3$. We set 6 groups for January and 5 groups for July. The separating points are chosen to be near the 30, 55, 75, 85 and 95 percentiles in January, and the 30, 60, 80, 90 percentiles in July. The groups are denser at higher concentrations to better reveal the details before and after heavy pollution periods.

In January, the transport becomes stronger when the concentration is higher, but the transport flux decreases in turn when the concentration is the highest. The inflow from Baoding and outflow to Chengde, which are the indicator of the Southwest-Northeast pathway, also experience a gradual rise followed by a sudden decline. In July, the situation is similar, though the decrease is less significant. Such result is consistent with Tang et al. (2015) and Zhu et al. (2016) that the Southwest-Northeast transport pathway is more significant during the rising phase of a heavy pollution period, but fades when the pollution reaches the peak.

Inspired by these results, we also conducted a day-to-day analysis on the two heavy pollution episodes described in Section 3.3, which occur in January and July, respectively (Fig. R4). We find the "flowing in and accumulating" phenomenon for both episodes. For the episode in January, the inflow (especially from southwest) is strong in January 18th, while the inflow declines rapidly in January 19th, the day with the highest concentration. The phenomenon is more significant during the episode in July. In July 18th and 19th, the inflow flux is very strong, while in July 20th which has the highest concentration, the flux decreases for more than one order of magnitude. This finding emphasizes the importance of early temporary control before heavy pollution occurs. We have revised our manuscript to include the above results and discussions. (Page 11-12, Line 294-306)

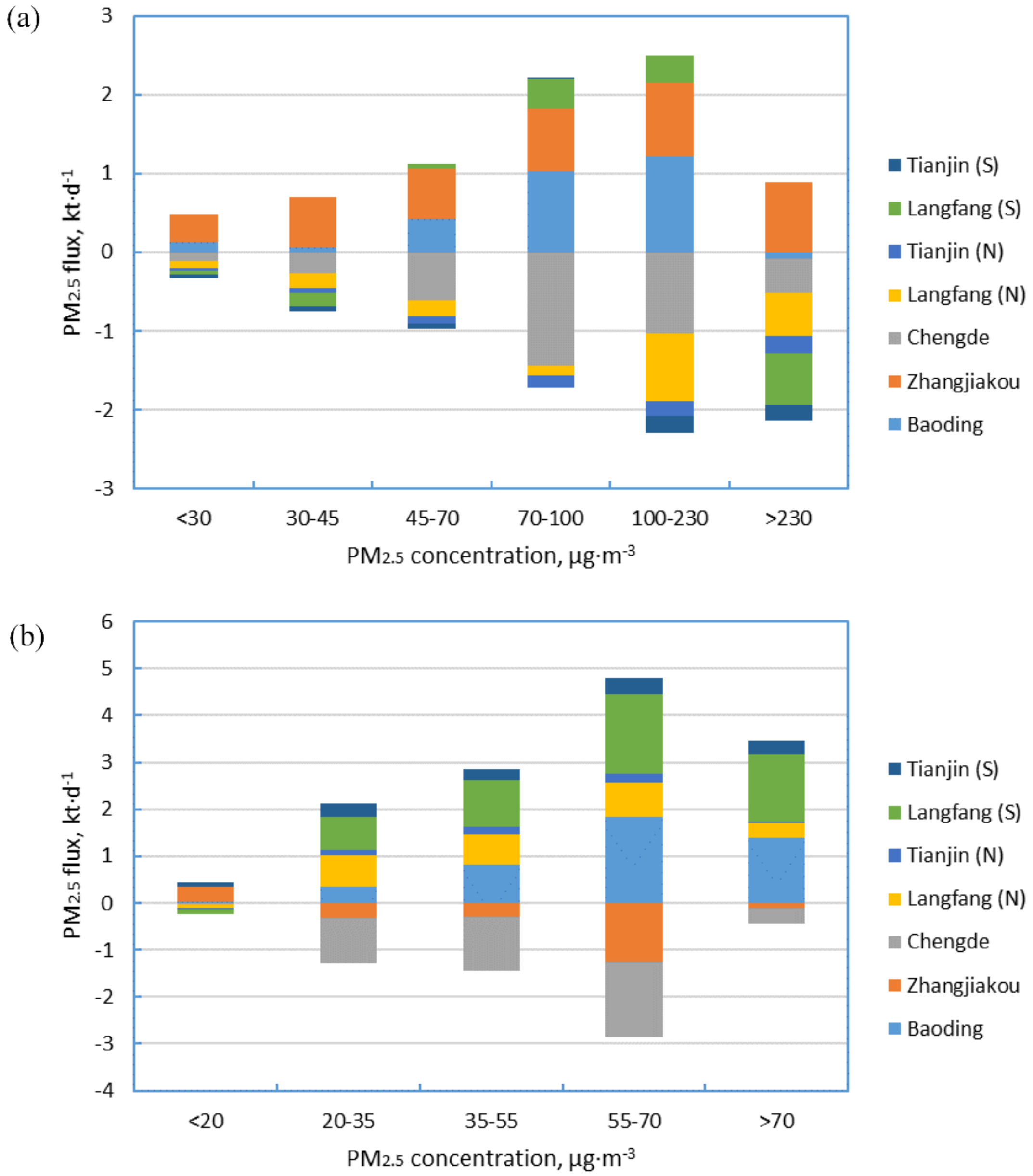

Figure R3 $PM_{2.5}$ average flux between Beijing and its neighboring cities in different pollution degrees in (a) January and (b) July.

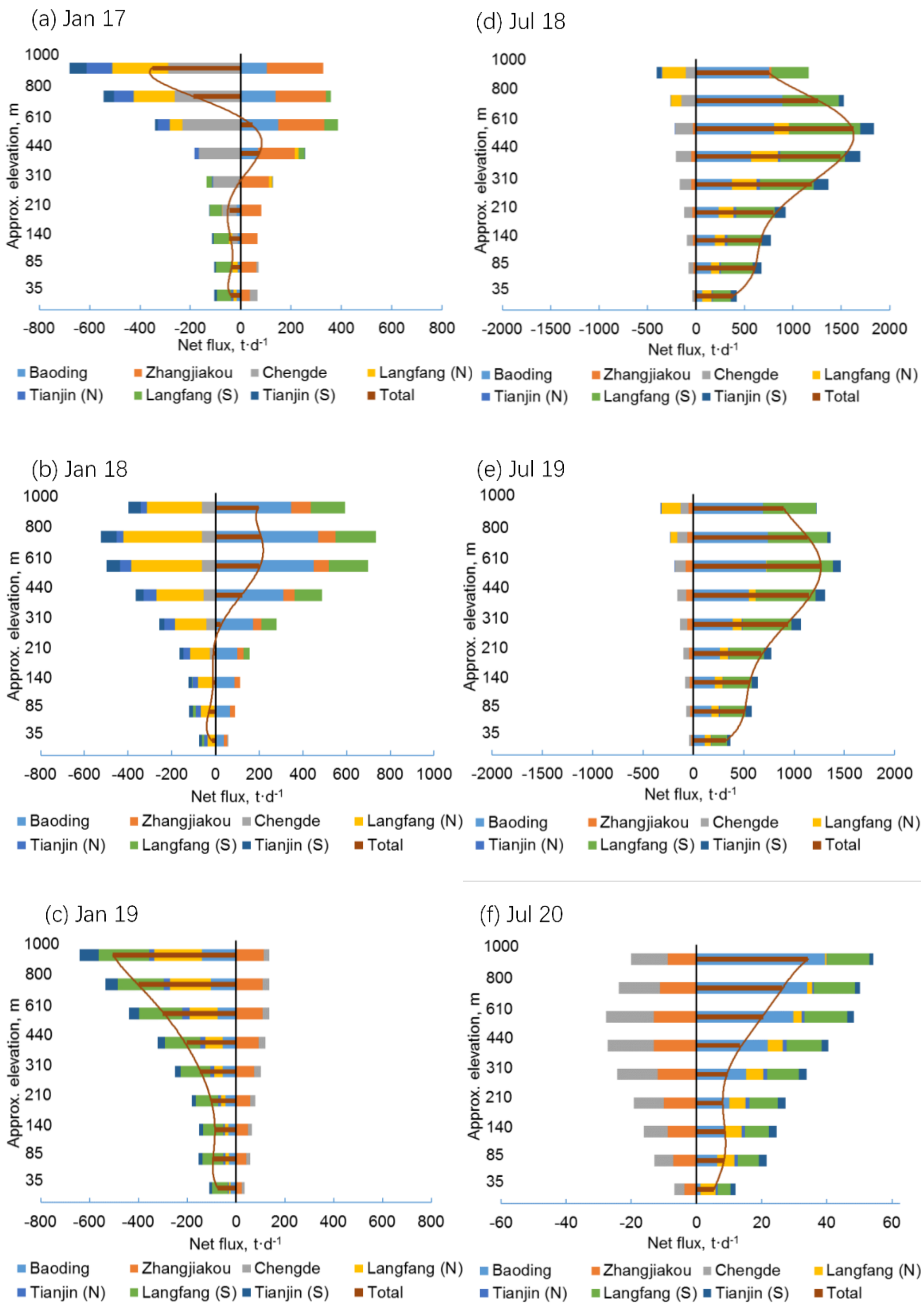

Figure R4 $PM_{2.5}$ fluxes during heavy-pollution days in Beijing in January and July: (a) January 17th, (b) January 18th, (c) January 19th, (d) July 18th, (e) July 19th and (f) July 20th.

In addition, some precursors were also transported in the initial periods. Afterwards, the precursors will react and form particles. Could you please quantify the contributions of the particles and the precursors' transport?

Authors' reply: The transport of precursors that may transform into particles is indeed an important factor. However, only if the precursors are tracked in all the physical and chemical reactions can we quantify the contribution of the precursor's transport to the $PM_{2.5}$. The flux approach is not able to account for this issue, which is one of the main shortages. Nevertheless, the flux approach can capture the transport features of all primary and most of the secondary $PM_{2.5}$. We have some discussion on this shortage in our manuscript, and we hope that future study can combine the tracer model with the flux approach to overcome this shortage.

What's more, without the passage of large- or medium-scale meteorological system, the local mountain-plain winds emerges in North China Plain (Tang et al., 2016, Fig. 10). The alternation between the mountainous (northeast) winds that begin at 03:00 LT at night and the plain (southwest) winds that begin at 12:00 LT in the afternoon occurs. Therefore, air pollutants will transport to the northeast direction in the afternoon and then transport back during latter of half of the night. Could you please clarify the transport circulations combined with the influences of the mountain-plain winds?

Authors' reply: We thank the reviewer very much for this useful comment. In our original study, we calculated daily $PM_{2.5}$ fluxes, so that the mountain-plain winds (which is a diurnal variation feature) is not taken the into consideration. Following the reviewer's comment, we tried to probe into the diurnal wind and flux pattern in Beijing. The simulated average diurnal wind patterns at 100 m height in January and July in Beijing are shown in Fig. R5(b). We also put the observation results from Tang et al., (2016) in Fig R5(a) as a reference. We find that the simulated wind pattern is consistent with the observation. In January, the mountain-plain winds are presented as the change in wind speed, but the wind direction does not change significantly during the whole day. In July, there is a significant wind direction shift, similar to the description of the reviewer. The mountainous wind (northeast) begins at 2:00 LT, and is taken over by the plain wind (southeast) at about 10:00 LT, and the mountainous wind is much weaker than the plain wind. A circulation of mountain-plain wind may have influence on the transport of $PM_{2.5}$ in July.

Considering that the mountain-plain wind circulation mainly happens at the foot of the mountains, we calculated the fluxes through the boundaries between Beijing and its three neighboring cities on the south/southeast (Baoding, Langfang and Tianjin) during mountainous wind hours and the plain wind hours in July separately (Fig. R6). During the plain wind hours, all the boundaries on the southwest and southeast of Beijing have positive net fluxes, which is due to the relatively strong southerly plain winds. During the mountainous wind hours, however, there is no significant direction change of the fluxes except for the boundary of Baoding and Southern Langfang at levels below 200 m. The sign of fluxes mostly remaines unchanged because the mountain-plain wind circulation is weaker at higher levels, and the wind speed of the mountainous wind is even weaker at the southernly boundaries which has limited effect to alter the sign of the flux. Nevertheless, the magnitude of fluxes is significantly smaller than the plain wind hours, which is partly attributed to the mountain-plain wind circulation. Therefore, the summertime mountain-plain wind circulation in Beijing does not significantly alter the sign of inter-city $PM_{2.5}$ fluxes but does have considerable impact on their magnitude. We have included the discussion on the mountain-plain wind in our revised manuscripts (Page 14, Line 365-369 in the main text, and Page 12-13, Line 102-131 in SI).

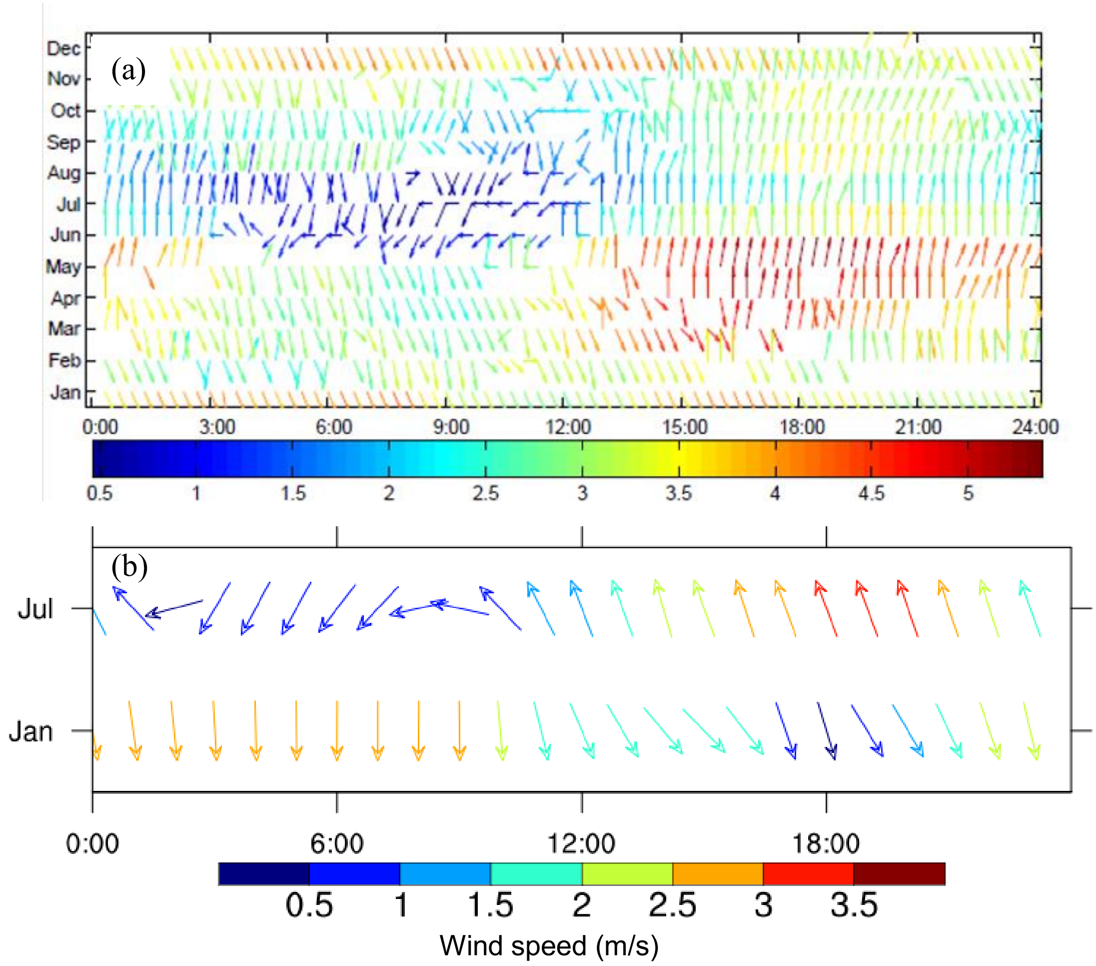

Figure R5 The observed and simulated monthly average diurnal variation of winds in Beijing in July. (a) The observation results from Tang et al. (2016). (b) The simulation results in this study.

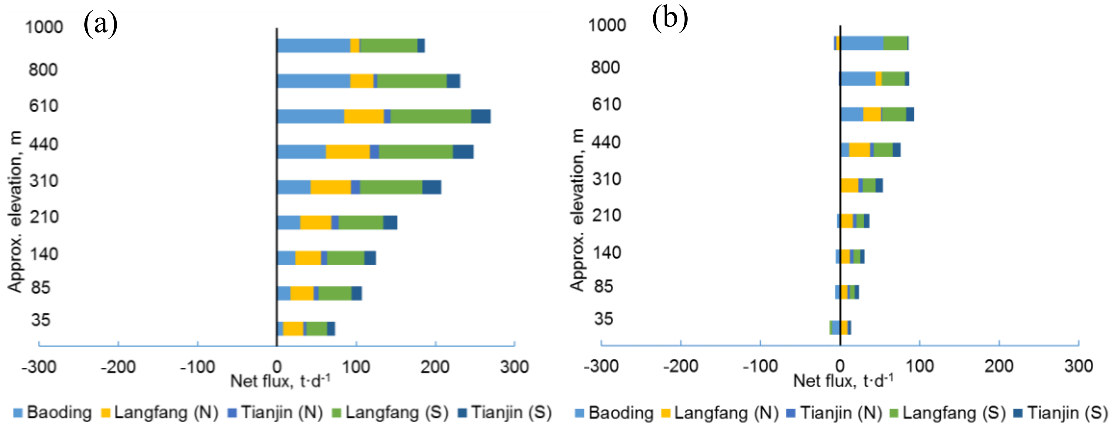

Figure R6 The transport fluxes in July between Beijing and its neighboring cities during (a) plain wind hours (11:00 – 1:00 (+1 day) LT) and (b) mountainous wind hours (2:00 – 10:00 LT)

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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 pollution of the Beijing-Tianjin-Hebei (BTH) region in China. However, previous studies on regional transport of $PM_{2.5}$ mainly aim 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 300 – 1000 m). In winter, even if surface wind speeds are low, the transport at above 300 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 $\mu g/m^3$, 93 $\mu g/m^3$ and 77 $\mu g/m^3$, respectively, which far exceeded the 35 $\mu g/m^3$ standard in 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 $PM_{2.5}$ concentration in the BTH region.

Emissions from one city can substantially affect the $PM_{2.5}$ pollution in another city under particular meteorology conditions by the transport process. For example, some studies showed that emissions from outside Beijing can contribute to 28-70% of the ambient $PM_{2.5}$ concentration in Beijing (An et al., 2007; Streets et al., 2007; BJEPB, 2015; Wang et al., 2014b). 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 of the air mass before it arrives 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, the sensitivity of $PM_{2.5}$ concentration in the target city to emissions from the source city is not necessarily the same as the contribution of transport process, because of the non-linear relationships between emissions and concentrations(Kwok et al., 2015). Based on the simulated meteorology field and air pollutant concentrations, the inter-city transport of $PM_{2.5}$ can be simply expressed 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 large scale transport of air pollutants, such as inter-continent transports (Berge and Jakobsen, 1998; In et al., 2007). There are also studies that evaluated the pollutant transport on 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 focused on Beijing. In recent years, however, under the policy of "integrating development of BTH region", the air quality in Tianjin and the 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. The $PM_{2.5}$ 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 region outside China mainland, we use the MIX emission inventory (Li et al., 2017) of 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 of 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 in each 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 study (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

We establish a one-way, triple nesting domain in the 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. The top layer pressure is 100 mb at approximately 15 km. The National Center for Environmental Prediction (NCEP)'s Final Operational Global Analysis data with a horizontal resolution of 1°×1° at every 6 h are used to generate the first guess field. 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 cloud microphysics scheme, and the Rapid Radiative Transfer Model (RRTM) longwave and shortwave radiation scheme. The Meteorology-Chemistry Interface Processor (MCIP) version 3.3 is applied to convert the WRF output data to a format required by CMAQ.

We 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 of each month 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. Most of the indices are 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. However, the MFB and MFE indices in January and July for the domain all fall inside the "criteria" benchmark value suggested by Boylan and Russell (2006). To understand the reason of the underestimation, it is necessary 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 it with the $PM_{2.5}$ 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 of sulfate. The underestimation of sulfate also partly explains the overestimation of nitrate. Moreover, the biases of $PM_{2.5}$ major components in the current study fall in a similar range with other studies in the BTH region (Wang et al., 2015; Wang et al., 2014a; Wang et al., 2011; Zhao et al., 2017; Liu et al., 2016) (See details in the SI). 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 $PM_{2.5}$ that flow 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 model can only provide 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

$$Flux = \sum_{i=1}^{h} \sum_{l} LH_i c\vec{v} \cdot \vec{n} \quad (1)$$

where l is the boundary line of two regions; 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 $PM_{2.5}$ at 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 9th layer above the ground (about 1000 m) as the top layer, 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 is less important because the vertical mixing above the boundary layer is weaker.

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.

The locations of the three target cities and their neighbors are shown in Fig. 1. Note that there is a small area surrounded by Beijing and Tianjin that belongs to the city of Langfang, so the boundaries between Beijing and Tianjin, Beijing and Langfang, and Tianjin and Langfang are each 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 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 $PM_{2.5}$ inflows of these two cities generally exceed the outflows, 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 us understand the extent to which 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 the 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, where Xingtai (on the south) and Baoding (on the northeast) contributing relatively more to inflow and outflow fluxes, respectively.

$PM_{2.5}$ 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 of each vertical layer in the CMAQ model are shown separately, and the approximate elevation of each layer is marked on the left. Generally, the total flowing intensity is stronger at higher levels for all three cities, while the major contributor varies with layers. 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, the total net flux for Tianjin shows a peak value near 600 m where Tangshan (on the northeast) receives much more outflow than it does near the ground. Total net flux for Shijiazhuang shows a peak value near 400 m where Hengshui (on the east) and Xingtai (on the south) have dominant contributions.

In order to better understand the general image of the transport characteristics in the BTH region, we display the net flux results on a map, using arrows to represent the net flux direction and intensity. The result of January is shown in Fig. 6(a). Bigger arrow represents larger flux, and white and black arrows denote fluxes at the lower (layer 1-5 in the model, from the ground to about 310 m) and upper (layer 6-9 in the model, from about 310 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 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 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 a $PM_{2.5}$ inflow at higher altitude origins more probably from a farther source. 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 during the winter time in the BTH region. In contrast, the northwest wind usually comes during the passage of a cold high pressure, with a 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 for Beijing mainly come from Langfang (on the southeast) and Baoding (on the southwest), and the outflow fluxes mainly go 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 the 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, where inflows from Xingtai (on the south) and Hengshui (on the east) are slightly larger.

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 the ground is from the southeast. Within the boundary layer, the wind will rotate clockwise and become stronger at higher levels 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 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 no more contribute a considerable amount of inflow flux.

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. Considering that in summer the vertical mixing is stronger, 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. To better understand how the wind and concentration affect the transport fluxes, we calculate the frequency of wind directions and the corresponding wind speed and $PM_{2.5}$ concentration, and plot them as "wind rose" plots. We show the plots of Beijing in Fig. 7 as an example. The plots for the other two cities can be found in SI.

In January, the dominant wind directions near the ground ranges from northwest to northeast. The NNE wind has the highest frequency, while the NW wind has the highest wind speed (Fig. 7(a)). The dominant northern winds reflect the winter monsoon. Although the concentration coming with the northern winds are relatively low because of the low emission rate on that direction(Fig. 7(b)), the high frequency and wind speed also cause an overall strong transport from the northwest to the southeast. Wind directions and the corresponding concentrations are quite different at the upper layers (Fig. 7(c), (d)). The prevalent northern wind remains (though the dominant directions shift slightly from NNE to NW), and the frequency of southwestern winds is much higher than that at lower layers. Moreover, the $PM_{2.5}$ concentrations that come with southwestern winds are much higher than the other directions. The strong emission in southern Hebei (which lies on the southwest direction of Beijing), especially the elevated source may be responsible for the high concentration from the southwest. Therefore, in January, the dominant northwestern winds account for the Northwest-Southeast pathway at both lower layers and upper layers, while the large emissions on the southwest direction mainly caused the Southwest-Northeast pathway at upper layers.

In July, the dominant wind directions at the lower layer are the southeastern directions, reflecting the summer monsoon (Fig 7(e)), and coincidentally the highest concentrations also come along with the southeastern winds (Fig 7(f)). Emissions from Tianjin, Langfang, and Tangshan may influence Beijing by the southeastern winds. The emission and the wind direction both contribute to the Southeast-Northwest pathway at the lower layers. The high frequency wind directions shift clockwise to the southern directions at the upper layers in July, as is shown in Fig 7(g), and the southwest wind and the southeast wind are both important. Moreover, the directions with high concentrations also shift to both the southwest and the southeast directions (Fig 7(h)). Therefore, in July, the dominant southeastern winds and the emissions on the southeast directions caused the Southeast-Northwest pathway at both the upper and the lower layers. The Southwest-Northeast pathway is a combination result from the southern winds and the emissions, which is different from that in January.

The monthly transport characteristics could bring us inspiration on how the joint control of different cities should be applied. The transport pathway 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. The daily characteristics of $PM_{2.5}$ transport in Beijing

In addition to the monthly characteristics of $PM_{2.5}$ transports discussed in Section 3.2, we analyzed the daily characteristics in this sector, taking Beijing as an example. Firstly, since different $PM_{2.5}$ concentration may be caused by different meteorology condition, and may also result in different transport flux characteristics in different days, we first calculate the $PM_{2.5}$ flux during different pollution levels (Fig. 8). We sort the daily data into 6 groups in January and 5 groups in July. The separating points are chosen to be near the 30, 55, 75, 85 and 95 percentiles in January, and the 30, 60, 80, 90 percentiles in July. The groups are denser at higher concentrations to better reveal the details around heavy pollution periods.

In January, the transport becomes stronger when the concentration is higher, but the transport flux decreases in turn when the concentration is the highest. The inflow from Baoding and outflow to Chengde, which are the indicator of the Southwest-Northeast pathway, also rise gradually, followed by a sudden decrease. In July, the situation is similar, though the decrease is less significant. Such result is consistent with Tang et al. (2015) and Zhu et al. (2016) that the Southwest-Northeast transport pathway is more significant when the pollution is still rising.

To reveal the daily characteristics comprehensively, 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 17th - 19th, which are the most polluted days in January (the simulated $PM_{2.5}$ daily average concentrations all exceed 200 μg/m^3). In July, we also choose the period with the highest concentration, i.e. 18th to 20th. The results are shown in Fig. 9.

The magnitude of net fluxes in January 17th and 18th (-590 t/day and 688 t/day) is much higher than the monthly average value (139 t/day). 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 300 m 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.

However, in 19th January when the concentration reaches the peak, the inflow transport becomes weaker than the previous days, especially for the southwest inflow. The $PM_{2.5}$ experienced a significant inflow from the southwest followed by an accumulation period with little inflow. Therefore, the Southwest-Northeast pathway is of great importance during the first days of 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 (f)), 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 and 19th July (Fig. 7 (d,e)), the days when the simulated $PM_{2.5}$ concentration reaches a high level but is still rising 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 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 that are 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 show 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 in 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.

Besides the daily variability of $PM_{2.5}$ transport, we also analyzed the diurnal variability brought by the "mountain – plain wind cycle" in summer times in Beijing (Tang et al., 2016). However, because the average plain wind is much stronger than the mountainous wind which is only obvious below 200 m, the fluxes brought by the mountainous wind is much weaker than that by plain wind (Fig. S6). The diurnal variation of winds does not have a significant influence on the direction of the transport fluxes.

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 300 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 300 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 300 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.

By analyzing daily transport fluxes in Beijing, we also find that the flux during the days with higher $PM_{2.5}$ concentration is generally higher, but the flux during the top 10% polluted days is smaller. The flux during heavy-pollution episodes is stronger than the monthly average 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. First, we only quantify the transport of $PM_{2.5}$ at the boundary of the city, which is not the only way by which transport process may influence the $PM_{2.5}$ concentration in the target city. Other processes include the inter-city transport of gaseous precursors that remain in gaseous phase at the boundary but may convert to secondary $PM_{2.5}$ in the target city. Secondly, the $PM_{2.5}$ transported through the outer boundary is a mixture of different sources that does not only from the neighbor city itself. Although we have obtained a general transport feature in the BTH region which can facilitate a qualitative understanding of where the fluxes are mainly from, the flux approach cannot quantitatively evaluate the contribution from each city in the upstream areas. If we want to overcome these disadvantages, a life-time tracing during the emission, transportation, reaction and deposition processes of $PM_{2.5}$ and its gaseous precursors is needed. Therefore, future studies may combine the flux calculation with the tagging models 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 3 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

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

Indices		Mean OBS	Mean SIM[a]	NMB	NME	MFB	MFE
Unit		$\mu g \cdot m^{-3}$	$\mu g \cdot m^{-3}$	%	%	%	%
	Beijing	86.0	65.2	-24.2	32.2		
	Shijiazhuang	193.9	170.8	-11.9	45.3		
January, 2012	Xianghe	132.3	85.6	-35.3	44.5	-19.6	19.6
	Xinglong	39.4	38.6	-2.0	42.7		
	Yucheng	140.9	124.1	-11.9	31.2		
	Beijing	68.2	35.6	-47.8	49.5		
	Shijiazhuang	70.3	79.8	+13.6	37.6		
July, 2012	Xianghe	61.3	47.2	-23.0	35.6	-35.1	40.2
	Xinglong	48.9	24.6	-49.6	53.8		
	Yucheng	77.3	55.2	-28.6	39.6		
"Criteria" benchmark[b]		-	-	-	-	≤±60	≤75
"Goal" benchmark[b]		-	-	-	-	≤±30	≤50

a. Average of the days only when observations are available.

b. Benchmarks are suggested by Boylan and Russell (2006).

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

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 $PM_{2.5}$ 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 (a) and July (b). The size of the arrows represents the amount of the fluxes, while white and black arrows denote fluxes at the lower (layer 1-5 in the model, from the ground to about 310 m) and upper (layer 6-9 in the model, from about 310 m to about 1000 m) layers, respectively.

Figure 7. The wind rose plots showing the frequency of wind speed (a, c, e, g) and $PM_{2.5}$ concentration (b, d, f, h) at different wind directions for Beijing. The ground level and the 7th level (about 450-600 m) in the model are chosen as the representation of lower levels and upper levels. The percentages denote the frequency.

Figure 8. $PM_{2.5}$ average flux in different pollution degrees in (a) January and (b) July.

Figure 9. $PM_{2.5}$ fluxes during heavy-pollution days in Beijing in January and July: (a) January 17th, (b) January 18th, (c) January 19th, (d) July 18th, (e) July 19th and (f) July 20th..

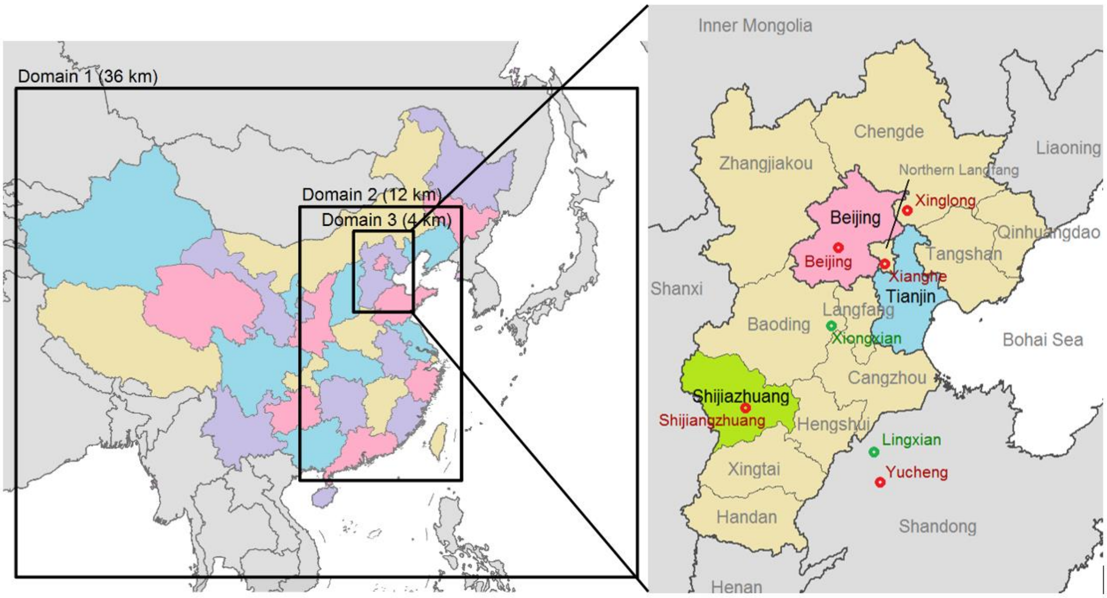


Figure 1 The simulation domains used in this study (left) and the map of the Beijing-Tianjin-Heibei region (right). The highlighted cities are the target cities for flux calculation. The red circles show the sites with $PM_{2.5}$ observations. The two sites with green circles have observations of $PM_{2.5}$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

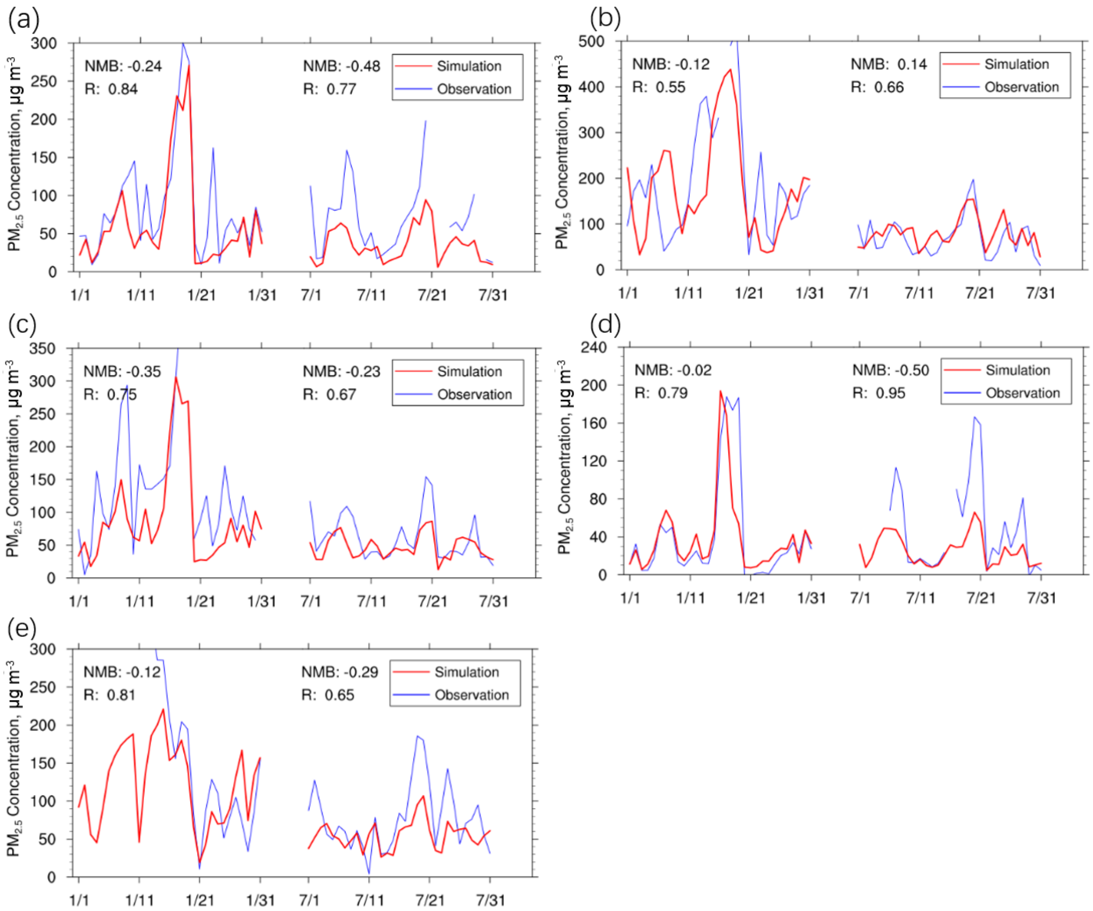
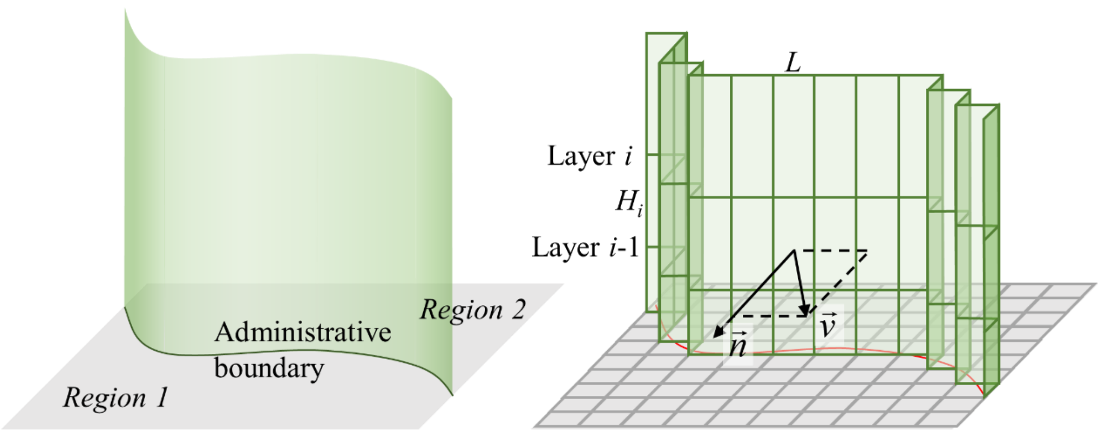


Figure 2 Time series of the simulated and observed $PM_{2.5}$ concentrations in (a) Beijing, (b) Shijiazhuang, (c) Xianghe, (d) Xinglong, and (e) Yucheng



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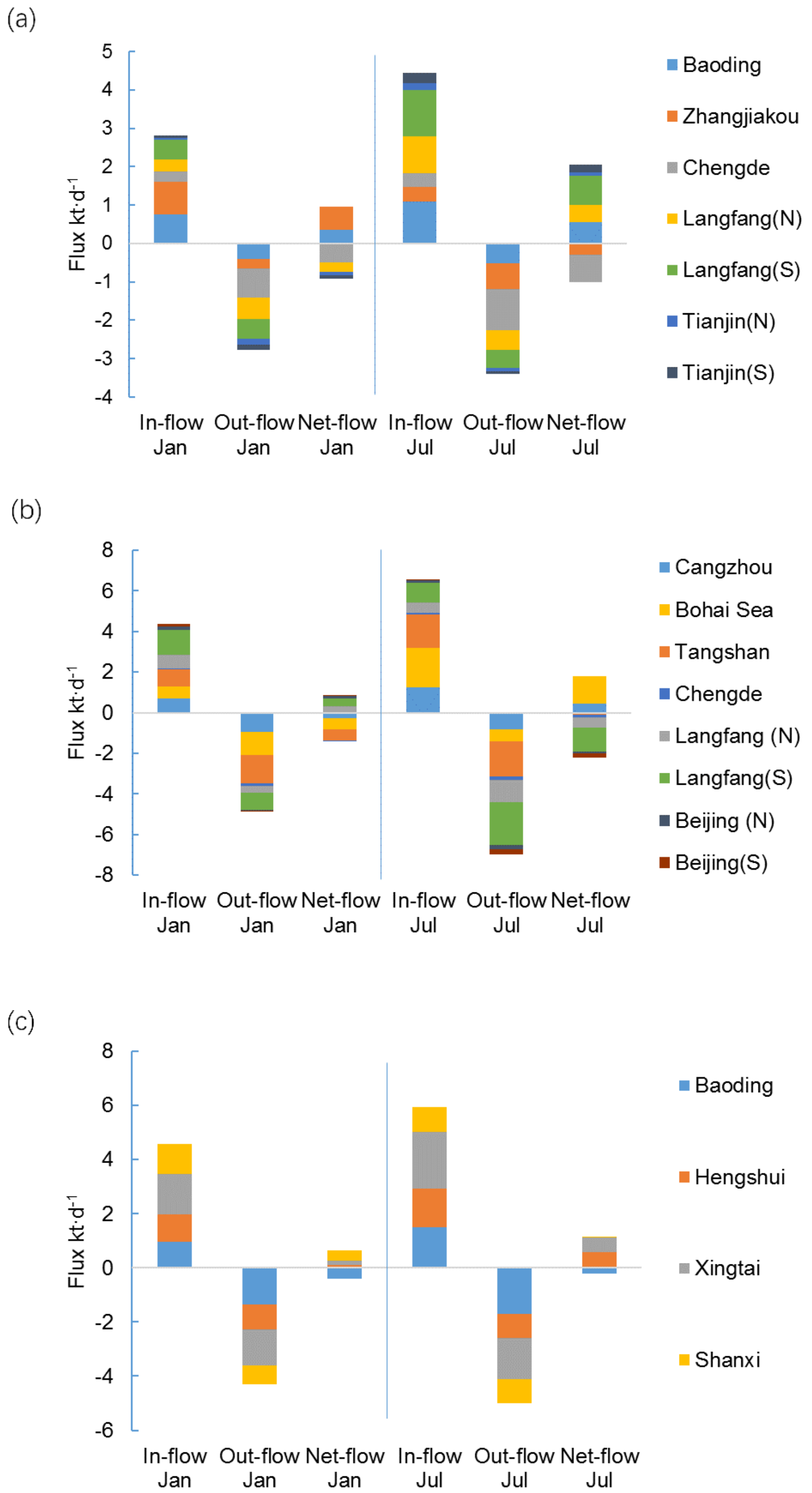

Figure 4 The inflow, outflow and net fluxes in January and July for (a) Beijing, (b) Tianjin, and (c) Shijiazhuang

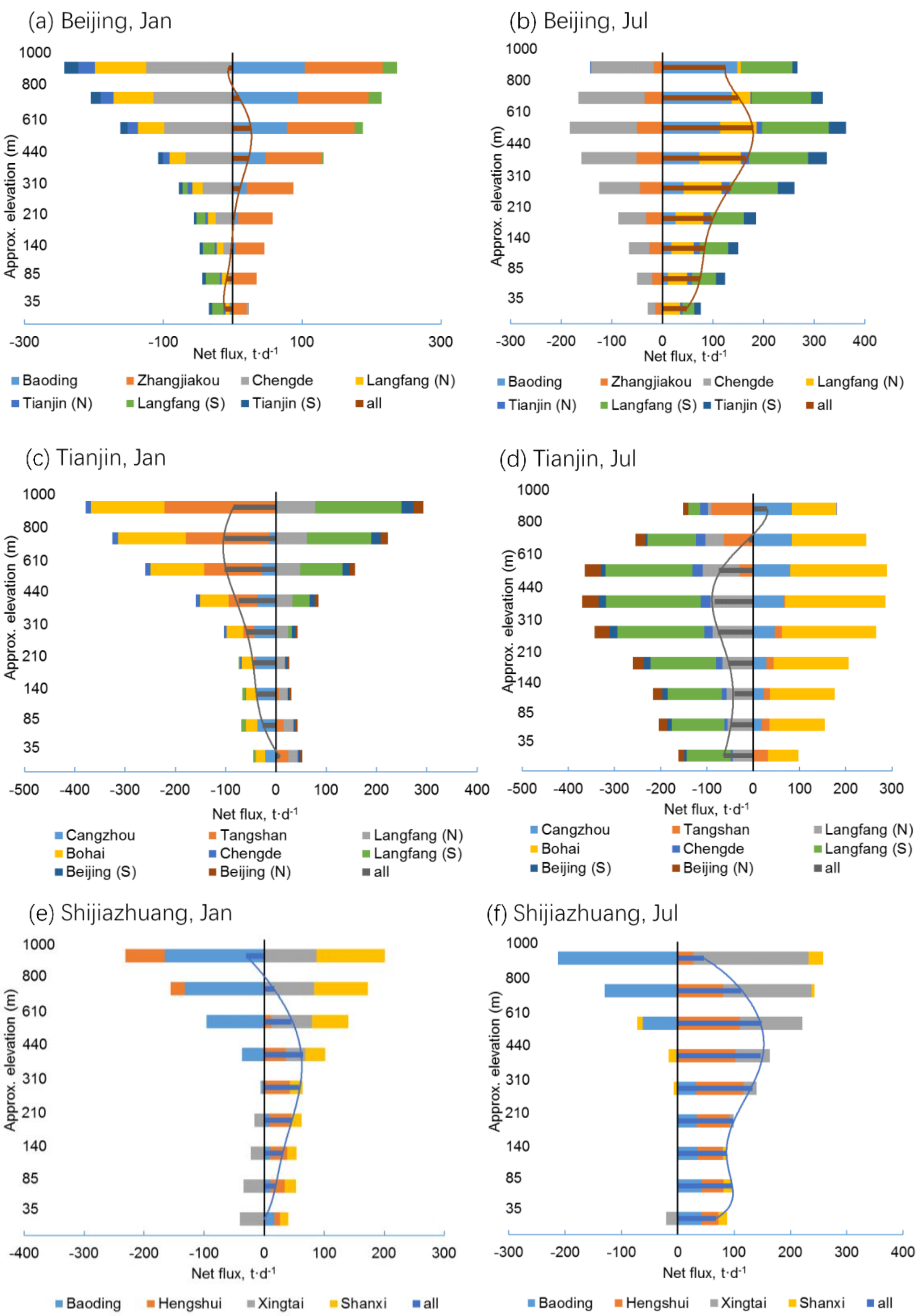

Figure 5 Vertical distribution of net fluxes in January (left) and July (right) for (a-b) Beijing, (c-d) Tianjin, and (e-f) Shijiazhuang

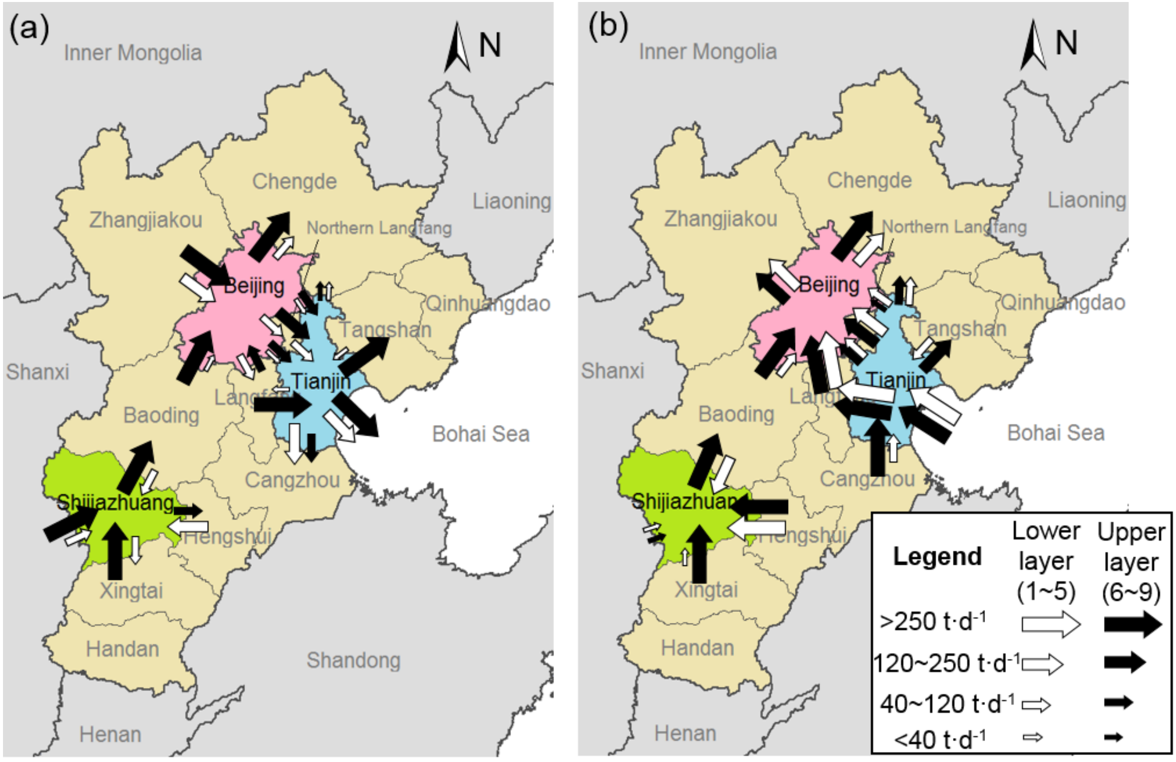

Figure 6 The transport fluxes through each boundary segment of the three target cities in January (a) and July (b). The size of the arrows represents the amount of the fluxes, while white and black arrows denote fluxes at the lower (layer 1-5 in the model, from the ground to about 310 m) and upper (layer 6-9 in the model, from about 310 m to about 1000 m) layers, respectively.

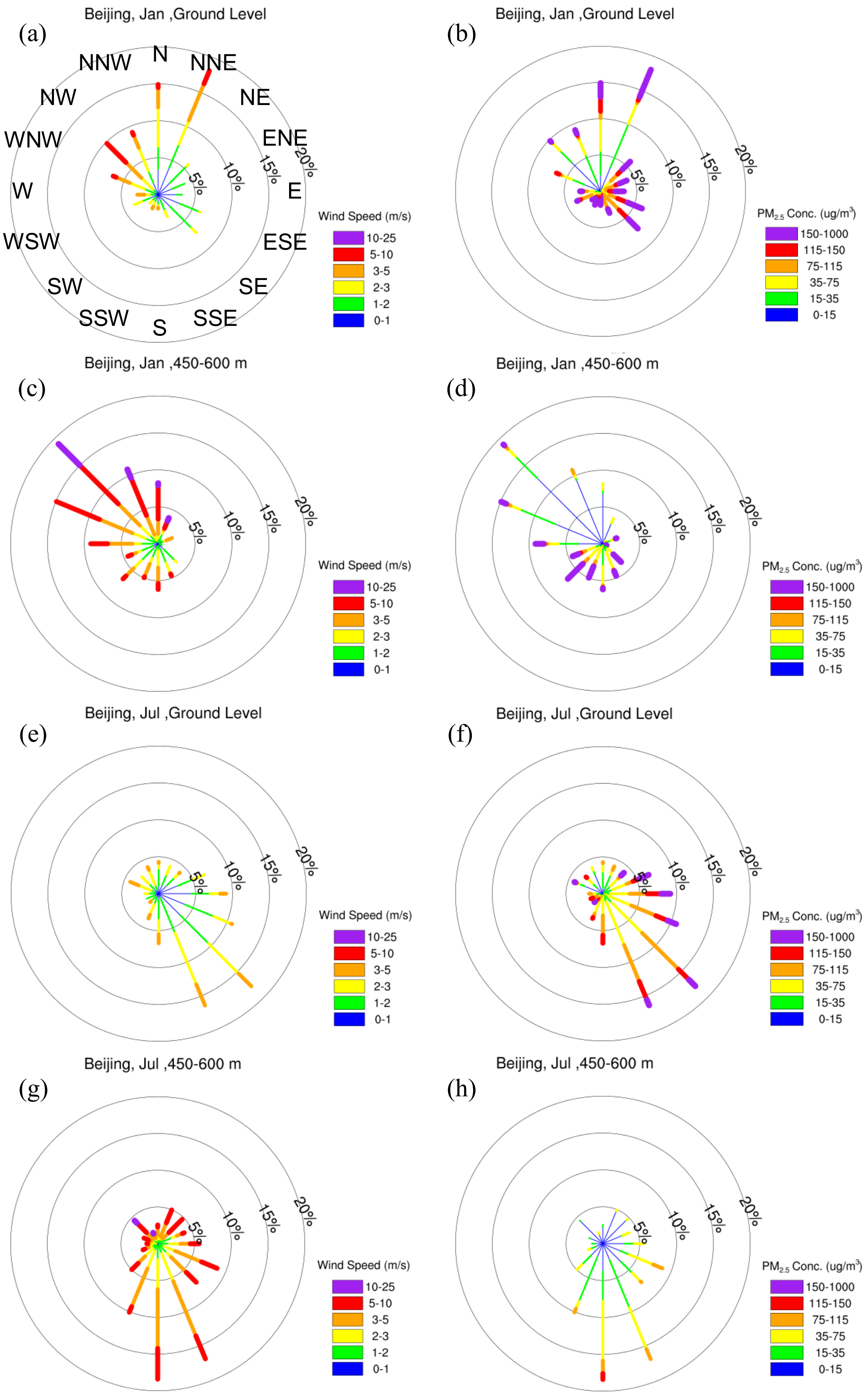

Figure 7 The wind rose plots showing the frequency of wind speed (a, c, e, g) and $PM_{2.5}$ concentration

(b, d, f, h) at different wind directions for Beijing. The ground level and the 7th level (about 450-600 m) in the model are chosen as the representation of lower levels and upper levels. The percentages denote the frequency.

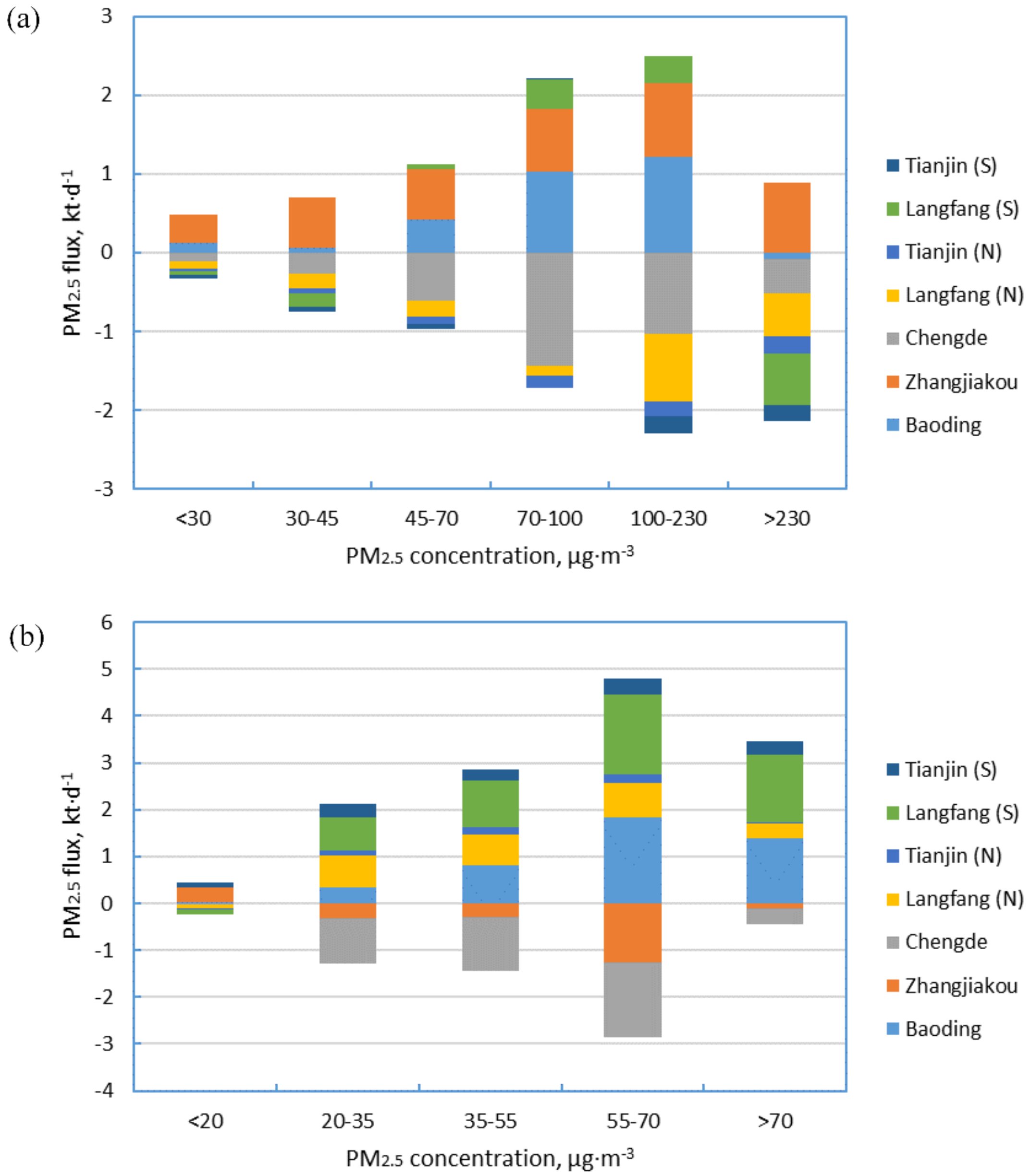

Figure 8 PM2.5 average flux in different pollution degrees in (a) January and (b) July.

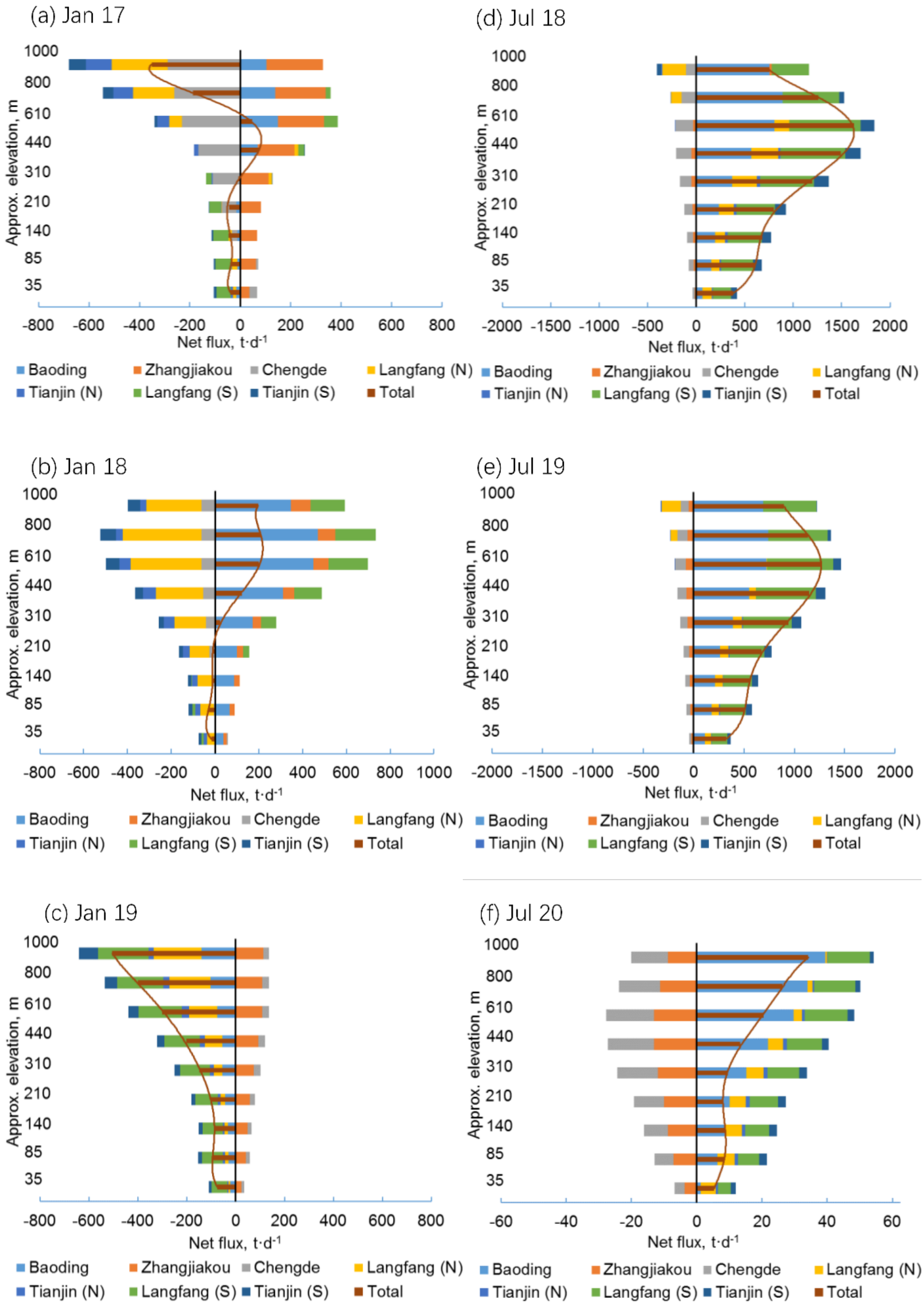

Figure 9 $PM_{2.5}$ fluxes during heavy-pollution days in Beijing in January and July: (a) January 17th, (b) January 18th, (c) January 19th, (d) July 18th, (e) July 19th and (f) July 20th.