Vertical distribution of PM$_{2.5}$ transport flux in summer and autumn in Beijing

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**ABSTRACT**

Temporal and spatial distribution characteristics of PM$_{2.5}$ transport fluxes between Beijing and its adjacent cities in July and October 2016 were estimated quantitatively using the cross-boundary transport flux method based on the WRF-CMAQ model in this study. Vertical distribution of PM$_{2.5}$ net fluxes was revealed, and hourly evolution characteristics of PM$_{2.5}$ transport fluxes in typical heavy pollution processes were illustrated. We verified significant seasonal differences in PM$_{2.5}$ transport fluxes between Beijing and its adjacent cities during the study period. PM$_{2.5}$ total inflow and outflow fluxes in autumn were 3187 t/d and -2721 t/d, which were significantly higher than that of 2134 t/d and -2172 t/d in summer. In autumn, Beijing received more PM$_{2.5}$ from adjacent cities, while the results in summer were opposite. Maximum net fluxes appeared at 600 - 800 m and 1000 - 1260 m above the ground in summer and autumn, respectively. The vertical distribution characteristics of pollution days and clean days were consistent, both of which show a net inflow from Baoding and Langfang to Beijing, while Beijing was in a state of net outflow to Chengde. During the whole heavy pollution process, the evolution characteristics of PM$_{2.5}$ flux at low and high altitudes were consistent, and the intensity of the latter was 2.15~5.30 times of the former. Meanwhile, local discharging was more likely to cause extreme PM$_{2.5}$ heavy pollution, and a better peak clipping effect can be achieved when emission reduction measures are initiated 1 to 2 days before heavy pollution. The results can provide scientific support to put forward effective joint control measures and obtain insights into the evolutionary mechanism.
of haze episodes in Beijing.

Keywords
meteorology-air quality coupling model system (WRF-CMAQ), PM$_{2.5}$ transport flux, heavy pollution, vertical distribution, seasonal difference, Beijing

1. INTRODUCTION
Currently, China's air pollution presents regional compound pollution characterized by PM$_{2.5}$ and O$_3$ pollution. Its essence is the coupling of multiple atmospheric environmental problems caused by the emission of a variety of air pollutants (Hua et al., 2021; Yan et al., 2020). Affected by the atmospheric circulation, multiple pollutants were transported across the boundary, resulting in a series of atmospheric pollution (Zhang et al., 2021), such as the rise of PM$_{2.5}$ and O$_3$ concentration levels in contiguous areas, haze, photochemical smog, etc. PM$_{2.5}$ has small particle sizes, which are rich in a large number of toxic and harmful chemical components (Zhang et al., 2022). In addition, PM$_{2.5}$ can stay in the atmosphere for a long time (Wang et al., 2021) and be transported over a long distance (Li et al., 2021). Therefore, it has a significant impact on air quality (Elizabeth et al., 2017), atmospheric visibility (Cheng et al., 2021), human health (Qi et al., 2016), climate changes (Zhong et al., 2019), and other aspects. China's air quality has improved significantly since 2013 when the Chinese government implemented the “Air Pollution Prevention and Control Action Plan”. However, regional heavy pollution incidents dominated by PM$_{2.5}$ still occurred frequently, especially in the Beijing-Tianjin-Hebei region (Li et al., 2016), Yangtze River Delta (Shu et al., 2017),
Pearl River Delta (Liu et al., 2017), and Sichuan-Chongqing (Li et al., 2017a), which have attracted widespread attention from domestic and foreign scholars. Therefore, it is imperative to quantitatively identify the quantitative characteristics of transboundary transport of PM$_{2.5}$ pollution and to elucidate the contribution of local emissions and regional transport.

Technical means of studying PM$_{2.5}$ cross-boundary transport at home and abroad mainly included mathematical statistics, stereo observation, and modeling simulation. Ge et al. (2018) developed a mathematical-statistical approach to calculate the relative contributions of minimal local emission (MinLEC) and the maximum regional transport (MaxRTC), which is based on the measurement results of several air pollutants concentrations near the surface of the North China Plain. They estimated impact of local emissions and regional transport on air quality, and illustrated that the average annual MinLEC (MaxRTC) contributions of NO$_2$, SO$_2$, PM$_{2.5}$, and CO in the North China Plain were 61.7% (30.7%), 46.6% (48%), 52.1% (40.2%) and 35.8% (45.5%), respectively. Lv et al. (2017) have studied heavy haze on the sixth ring road in Beijing based on a vehicle-based mobile lidar and found that the average inflow flux from the southwest on the day of heavy pollution was about 308$\mu$g/m$^2$·s. This showed a significant transport path from the southwest to the northeast, demonstrating a greater impact on the formation of this heavy pollution along the southwest transport path. In addition, the air quality model has become one of the most widely used and most important methods for studying the laws of cross-boundary transport of pollution, which can reproduce the physical and chemical processes of atmospheric
pollutants in the atmosphere, such as the transmission, transformation, diffusion, and
sedimentation. At present, scholars at home and abroad have quantitatively identified
the contribution of atmospheric PM$_{2.5}$ pollution transport near the surface in multiple
regions, indicated the importance of regional transport to the formation of heavy
PM$_{2.5}$ pollution in multiple cities/regions, based on the source apportionment
techniques in numerical models, such as the Particulate Matter Source Apportionment
Technology (PAST) in CAMx (Comprehensive Air Quality Model with Extension)
model (Wang et al., 2017a), the Integrated Source Apportionment Model (ISAM) in
CMAQ (Community Multiscale Air Quality) Model (Dong et al., 2020), the on-line
tracer-tagged technology in NAQPMS (a three-dimensional regional Eulerian
chemical transport model developed by the Institute of Atmospheric Physics)
model (Yang et al., 2020), the source sensitivity (brute force zero-out) approach (Baker
et al., 2016), and the process analysis (Zhang et al., 2019a). However, the
above-mentioned studies mostly focused on the contribution of near-surface PM$_{2.5}$
local emissions and regional transport and lacked the quantitative characteristics of
the transport of PM$_{2.5}$ pollution at different ground clearances in the vertical direction.

Transport flux method has been widely used to study the characteristics of air
pollutants cross-boundary transport and can quantitatively evaluate the transportation
of pollutants between neighboring cities and regions by calculating the transport flux
of pollutants at different vertical heights, based on wind field and pollutant
concentration outputted by meteorological and air quality models (Yao et al., 2021). At
present, most studies focused on the vertical distribution characteristics of PM$_{2.5}$
transport flux in typical seasons (Chang et al., 2018). However, there were relatively few studies focusing on typical heavy pollution processes, elucidating the hourly evolution laws and vertical distribution characteristics of PM$_{2.5}$ transport flux, and the difference of PM$_{2.5}$ transport flux between clean days and heavy pollution days need to be further investigated.

Based on the implementation of regional air pollution prevention and control policies by the Chinese government, the PM$_{2.5}$ pollutions had been greatly improved during 2016-2022. However, heavy pollution has not been completely eliminated, especially in autumn when the wind is high and the air is crisp, heavy pollution events still occur occasionally. Therefore, we quantitatively identified the vertical distribution characteristics of PM$_{2.5}$ transport flux between Beijing and neighboring cities during July and October 2016, based on the WRF-CMAQ model, analyzed the difference in flux intensity under different seasons and pollution levels, and clarified the hourly evolution characteristics of PM$_{2.5}$ transport flux in typical heavy pollution processes. Flux intensity calculations can provide scientific support to formulate scientific and effective air pollution prevention and control strategies and PM$_{2.5}$ heavy pollution response plans in China.

2. MATERIALS AND METHODS

2.1 Model configuration

In this study, the Weather Research and Forecasting model (WRF, version 3.7.1) (Yang et al., 2018) and the Comprehensive Air Quality Model with Extension model (CMAQ, version 5.0.1) (Zhang et al., 2019b) were employed. The study period
was from July 5th to 30th and October 5th to 30th, 2016, with the simulation time interval of 1h. Among them, the parameter setting of WRF model can be found in Table 1. It is noteworthy that the cumulus cloud parameterization scheme is adopted for the domain 1 and 2, but not for the domain 3. δ coordinate represents the height parameter set in the vertical direction by the WRF and CMAQ model. Both models set 28 air pressure layers in the vertical direction of the simulation area, with the corresponding δ coordinates of 1.000, 0.994, 0.988, 0.981, 0.969, 0.956, 0.944, 0.926, 0.902, 0.881, 0.852, 0.828, 0.796, 0.754, 0.704, 0.648, 0.589, 0.546, 0.495, 0.445, 0.387, 0.287, 0.187, 0.136, 0.091, 0.061, 0.020 and 0.000. The corresponding height of each δ coordinate have been summarized in Table 2.

Table 1 Parameterization schemes of WRF model

<table>
<thead>
<tr>
<th>Parameterization process</th>
<th>Related parameters</th>
<th>Settings</th>
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</thead>
<tbody>
<tr>
<td>Microphysics</td>
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<td>Lin</td>
</tr>
<tr>
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<td>RRTM</td>
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<tr>
<td>Short-wave radiation</td>
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<tr>
<td>Land surface process</td>
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<td>Noah</td>
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<tr>
<td>Cumulus cloud parameterization (for domain 1 and 2)</td>
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<td>Grell-3D</td>
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<tr>
<td>Boundary layer</td>
<td>bl_pbl_physics</td>
<td>YSU</td>
</tr>
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</table>

Table 2 δ coordinate and its corresponding height in the vertical direction

<table>
<thead>
<tr>
<th>No.</th>
<th>δ</th>
<th>height/m</th>
<th>No.</th>
<th>δ</th>
<th>height/m</th>
</tr>
</thead>
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<tr>
<td>1</td>
<td>1.000</td>
<td>0</td>
<td>15</td>
<td>0.704</td>
<td>2701.70</td>
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<tr>
<td>2</td>
<td>0.994</td>
<td>48.23</td>
<td>16</td>
<td>0.648</td>
<td>3306.99</td>
</tr>
<tr>
<td>3</td>
<td>0.988</td>
<td>95.68</td>
<td>17</td>
<td>0.589</td>
<td>3988.72</td>
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<tr>
<td>4</td>
<td>0.981</td>
<td>153.50</td>
<td>18</td>
<td>0.546</td>
<td>4518.36</td>
</tr>
<tr>
<td>5</td>
<td>0.969</td>
<td>251.62</td>
<td>19</td>
<td>0.495</td>
<td>5188.10</td>
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<tr>
<td>6</td>
<td>0.956</td>
<td>358.99</td>
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<tr>
<td>7</td>
<td>0.944</td>
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<td>8</td>
<td>0.926</td>
<td>611.10</td>
<td>22</td>
<td>0.287</td>
<td>8595.12</td>
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<tr>
<td>9</td>
<td>0.902</td>
<td>817.34</td>
<td>23</td>
<td>0.187</td>
<td>10895.43</td>
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<tr>
<td>10</td>
<td>0.881</td>
<td>1001.27</td>
<td>24</td>
<td>0.136</td>
<td>12388.83</td>
</tr>
</tbody>
</table>
The horizontal simulation grids of the two models used the Lambert projection coordinate system, with three nested domains set up, and the grid resolution remained the same. The boundary layer of the outer grid adopted the default setting of the CMAQ model, and the boundary layer of the inner grid was determined by outer layer simulation. The central point for the CMAQ model was set at the coordinate (39.02°N, 116.47°E). Domain 1 covered most of eastern and central China, with a horizontal resolution of 36 km; Domain 2 covered the Beijing-Tianjin-Hebei (BTH) region, Henan, Shanxi, Shandong, parts of the Inner Mongolia Autonomous Region, Jiangsu, Hubei, Shaanxi, Anhui, Liaoning, Zhejiang, and Shanghai, with a horizontal resolution of 12 km; Domain 3 covered Beijing and neighboring cities, with a horizontal resolution of 4 km, as shown in Fig. 1. Since the initial conditions used in the inner and outer layer simulations were all default values, the actual simulation period starts on the 1st of the month. The first four days were to ensure that the model simulation was initialized and was not used for subsequent analysis.
2.2 Data collection

In this study, the hourly meteorological parameters at the surface, including temperature (T), wind speed (WS), wind direction, and relative humidity (RH), were obtained from the Chinese MICAPS (Meteorological Information Combine Analysis and Process System) (Wang et al., 2017b). The hourly data of surface PM$_{2.5}$ mass concentration were acquired from the website (https://www.aqistudy.cn/historydata) provided by the China Air Quality Online Monitoring and Analysis Platform, which were used to verify the accuracy of WRF and CMAQ model simulations. The initial weather field and boundary conditions adapted global troposphere analysis data based on the NCEP (National Centers for Environmental Prediction), with a resolution of 1° ×1° and a temporal resolution of 6 h. We employed a high-resolution county-level pollution emission inventory for the BTH region, which was developed by the Beijing University of Technology (Zhou et al., 2017). Emissions data outside the BTH region were obtained from the multi-resolution emission inventory for China (http://www.meicmodel.org/). The pollutants in the emission inventory include SO$_2$, CO, NO$_x$, NH$_3$, VOCs, PM$_{2.5}$, PM$_{10}$, and others.

2.3 PM$_{2.5}$ flux calculation

The PM$_{2.5}$ flux stands for the mass of PM$_{2.5}$ that flows through a special vertical surface during a certain period (Zhang et al., 2019c), which was used to quantitatively analyze the characteristics of PM$_{2.5}$ cross-boundary transport at different heights between the target region and the surroundings. We took Beijing as the target city,
determined the boundary line and its corresponding vertical section according to China’s administrative divisions, whose adjacent cities consisted Zhangjiakou, Baoding, Langfang (S), Langfang (N), Tianjin, and Chengde. The WRF and CMAQ model belongs to the Euler three-dimensional grid mode, the simulation area can be regarded as a three-dimensional box composed of multiple grids (Tin et al., 2021). Each grid has a fixed three-dimensional position and generates the wind field and PM$_{2.5}$ concentration in the corresponding grid. Therefore, the outflow data of the model can be used to calculate the transport flux of PM$_{2.5}$ on a specific section, after the section to be calculated was discretized according to the meshing method in the CMAQ model. The PM$_{2.5}$ inflow flux stands for the mass of PM$_{2.5}$ that flows into the target city from surrounding cities during a certain period, with represents of "+". The PM$_{2.5}$ outflow flux stands for the mass of PM$_{2.5}$ that flows from the target city to surrounding cities during a certain period, with represents of "-". The PM$_{2.5}$ net flux stands for the vector sum of the two fluxes. The calculation formula of PM$_{2.5}$ flux is indicated as below (Eq. 1):

$$\text{Flux} = \sum_{k=1}^{h} \sum_{l} L \cdot H_{k} \cdot c \cdot v \cdot n \cdot 8.64 \times 10^{-8}$$  \hspace{1cm} (1)$$

where Flux is the PM$_{2.5}$ transport flux (t/d); $h$ is the top height, with set to 12 layers in this study; $l$ is the boundary line between two adjacent cities; $L$ is the boundary length (m); $H_{k}$ is the altitude difference between layer $k$ and $k+1$(m); $c$ is the PM$_{2.5}$ concentration($\mu$g·m$^{-3}$); $v$ is the wind vector($^\circ$ and m·s$^{-1}$); and $n$ is the normal vector corresponding to the vertical grid cell; $8.64 \times 10^{-8}$ is the unit conversion coefficient that converts $\mu$g/s into t/d.
2.4 Model evaluation

To evaluate the performance of the WRF and CMAQ model, we selected correlation coefficient (COR), normalized mean bias (NMB), and normalized mean error (NME) as statistical parameters, which refer to U.S. Environmental Protection Agency (EPA)’s evaluation standards (US EPA, 2007), calculated as follows (Eq. 2; Eq. 3; Eq. 4):

\[ COR = \sqrt{\frac{\sum_{i=1}^{n} (Sim_{(i)} - Obs_{(i)})^2}{n}} \]  
\[ NMB = \frac{\sum_{i=1}^{n} (Sim_{(i)} - Obs_{(i)})}{\sum_{i=1}^{n} Obs_{(i)}} \]  
\[ NME = \frac{\sum_{i=1}^{n} |Sim_{(i)} - Obs_{(i)}|}{\sum_{i=1}^{n} Obs_{(i)}} \]

Where \( Sim_{(i)} \) stands for the simulations of meteorological factors or PM$_{2.5}$ mass concentration, \( Obs_{(i)} \) stands for the observations of meteorological factors or PM$_{2.5}$ mass concentration, \( n \) stands for the number of samples.

The meteorological observation data of two first-level stations in Beijing and Shijiazhuang were selected to verify the accuracy of the WRF model, including temperature at 2m (T2), relative humidity (RH), and wind speed at 10m (WS10). The verification results are shown in Fig. 2 and Fig. 3. On the whole, the simulations of T2, RH, and WS10 were relatively consistent with the observations, with a significantly positive correlation. For T2, the simulations of the two cities in July and October were generally higher than observations, with CORs of 0.74~0.93, NMBs ranging from
0.05% to 1.35% and NMEs ranging from 0.35% to 1.35%, so as to explain the WRF model can better reproduce the temporal and spatial distribution characteristics of air temperature. In terms of RH, the simulations were underestimated, with NMBs of -43.09%~18.99%, NMEs of 21.27%~43.09%, but the CORs were both above 0.54. For WS10, the CORs of Beijing (Shijiazhuang) in July and October were 0.41 (0.63) and 0.67 (0.58), respectively, which were lower than the CORs of T2 and the CORs of RH. Simulations were generally higher than observations, the simulation deviations of Shijiazhuang were relatively greater than that of Beijing. NMBs of Beijing (Shijiazhuang) during July and October were 14.33% (47.46%) and 26.91% (74.87%), respectively, and NMEs of Beijing (Shijiazhuang) during July and October were 30.86% (49.63%) and 42.63% (75.73%). Notably, the simulations of near-surface wind speed were generally greater than previous model studies (Chen et al., 2017), with reasons that may include differences in the local terrain, insufficient data resolution of the urban underlying surface, etc (Han et al., 2018).

For PM2.5 mass concentration, the simulations and the observations of the two cities had better consistency, with CORs both above 0.67. And the simulations were generally lower than the observations, with NMBs and NMEs varied between -35.74%~5.44% and 31.98%~40.36%, respectively. There were varying degrees of underestimation of PM2.5 mass concentration during heavy pollution days, which was similar to the other studies and may be the main reason for the larger overall simulation error. This may be related to many factors, such as the uncertainty of the emission inventory, the imperfect secondary pollutants conversion mechanism in the
model, the horizontal and vertical resolution, and the WRF simulation error (Gao et al., 2016; Liu et al., 2017). In summary, WRF and CMAQ models could accurately reproduce the evolution characteristics of various meteorological elements and PM$_{2.5}$ concentration in the simulation region, and the simulation error was within an acceptable range, which could be further used for the simulation study of PM$_{2.5}$ transport flux.

**Fig. 2** Comparison of the simulations and observations of temperature at 2 m (T2), relative humidity (RH), wind speed at 10 m (WS10), and PM$_{2.5}$ mass concentration in Beijing during July (a, b, c, and d) and October (e, f, g, and h), 2016. The statistical parameters of SIM, OBS, COR, NMB, and NME are included.
3 RESULTS

3.1 PM$_{2.5}$ total transport flux

3.1.1 Seasonal difference

There were significant differences in the total transport flux of PM$_{2.5}$ between Beijing and adjacent cities in July and October 2016, as shown in Fig. 4. Overall, based on the analysis of PM$_{2.5}$ and wind speed simulation error, the PM$_{2.5}$ concentration in July was underestimated more than that in October, meanwhile the wind speed simulations in July were less overestimated than those in October. These simulation errors may have increased the difference in the intensity of the inflow and outflow flux between July and October. The PM$_{2.5}$ average daily total inflow flux
(2134 t/d) was equivalent to the total outflow flux (-2172 t/d), resulting in a small total net flux (-38 t/d), which showed that the transport between Beijing and surrounding cities was basically in balance. The total inflow flux from neighboring cities to Beijing was in the order of Baoding > Langfang (N) > Langfang (S) > Zhangjiakou > Tianjin > Chengde, the total outflow flux from Beijing to surrounding cities was in the order of Chengde > Zhangjiakou > Langfang (N) > Baoding > Langfang (S) > Tianjin. The PM$_{2.5}$ total net flux in October was relatively greater (467 t/d), which was about 12 times that in July. As for July, Beijing showed a net output to Chengde, while Baoding and Langfang showed a net input to Beijing. In October, both the total inflow flux (3187 t/d) and outflow flux (-2721 t/d) were greater than the flux in July. As in July, the total inflow flux from Baoding to Beijing was the strongest in October, and the total outflow flux from Beijing to Chengde was the strongest in October.

Fig. 4 Total transport flux distribution of PM$_{2.5}$ between Beijing and adjacent cities in July and October 2016
3.1.2 Analysis of the difference between clean days and polluted days

In this study, the periods when the average daily concentration of PM$_{2.5}$ was not higher than 75μg/m$^3$ were defined as clean days, the periods when the average daily concentration of PM$_{2.5}$ was higher than 75μg/m$^3$ were defined as pollution days. On the whole, the simulation errors of PM$_{2.5}$ concentration in clean days and wind speed in polluted days were relatively smaller. While PM$_{2.5}$ concentration in polluted days were underestimated and wind speed in clean days were overstated. The above results may lead to the increase of inflow and outflow flux intensity difference between clean days and polluted days. As shown in Fig. 5, the total inflow flux from Langfang (N) and Tianjin to Beijing were relatively larger, with the flux of 994 and 771 t/d, respectively. The total outflow flux from Beijing to Chengde and Zhangjiakou was relatively larger, with the flux of -914 and -1080 t/d, respectively. This showed that the transport contribution of high concentrations-PM$_{2.5}$ from external sources in Beijing in summer was mainly affected by the southeast monsoon climate, and the transport pathway along the southeast-northwest direction was relatively significant. In addition, the contributors of the total inflow and outflow flux of clean days in July were different from the pollution days. The major contributors were Baoding and Chengde, respectively, with the total net flux of 414 and -737 t/d, respectively, which indicated that PM$_{2.5}$ in Beijing was mainly transported along the southwest-northeast direction during clean days in July.

Contrary to the above, Baoding and Langfang (S) became the main contributors of the total inflow flux during polluted days in October, with the flux of 1645 and
1310 t/d, respectively. Chengde became the main receiver of the total outflow flux, with the flux of -2478 t/d, while the main contributors of the total net flux were Baoding, Langfang (S), and Chengde. Thus higher concentration may be affected by the southwest-northeast transport pathway in October in Beijing. Compared to the polluted days, the total inflow flux, outflow, and net flux during clean days were significantly lower, were 0.20–0.46 times the total flux of polluted days, and fluxes of the main contributing cities were relatively close.

Fig. 5 Total transport flux distribution of PM$_{2.5}$ on polluted days and clean days between Beijing and adjacent cities in July (a) and October (b) 2016
3.2 Vertical distribution of PM$_{2.5}$ transport flux

3.2.1 Seasonal difference

The vertical distribution characteristics of PM$_{2.5}$ net flux in Beijing in July and October 2016 are shown in Fig. 6. On the one hand, it showed that the PM$_{2.5}$ net flux varied with the height above the ground. On the other hand, it also revealed that there were significant differences in the direction of net transport flux in different seasons. The total net flux of PM$_{2.5}$ at each height above the ground in July was negative, except for those below 153m, revealing Beijing became a “source” that released more outflow than the inflow from the surroundings. Contrary to July, the total net flux of PM$_{2.5}$ at each height above the ground in October was positive, indicating Beijing acts as a role of “convergence” city that received the inflow from surroundings exceeded the outflow released by itself. In July, the PM$_{2.5}$ total net flux below 153m was expressed as a net inflow and mainly from Langfang (N) and Tianjin, with flux between 16 t/d~20 t/d and 16 t/d~18 t/d, respectively. The PM$_{2.5}$ total net flux that ranged from 153m to 1261m expressed as a net outflow appeared as a bimodal distribution and reached the maximum (~21 t/d) at approximately 600 – 800 m. Langfang and Baoding were the main contributors of the net inflow flux. The net flux transported from Baoding to Beijing increased significantly with height (from -1 t/d to 68 t/d), so as revealed the significant influence of pollutants transportation from Baoding to Beijing in the southwest transport path. In addition, Beijing had a continuous net outflow flux to Chengde and Zhangjiakou, especially Chengde, where received the net outflow flux reached -120 t/d.
In October, the PM$_{2.5}$ total net flux below 459m changed relatively stable (20 – 36 t/d), then the flux increased significantly with height and reached the maximum (70 t/d) at approximately 1000 – 1260 m. Similar to July, Langfang and Baoding were the main contributors of the net inflow flux, and Chengde was the main receiver of the net outflow flux. Generally, Beijing was greatly affected by regional transport from Baoding and Langfang, which had a greater influence on the outflow to Chengde.

**Fig. 6** Vertical distribution of PM$_{2.5}$ net fluxes between Beijing and its adjacent cities during July (a) and October (b) 2016

### 3.2.2 Difference between clean and polluted days

The differences in vertical distribution characteristics of PM$_{2.5}$ net flux during the clean days and polluted days are shown in Fig. 7. As shown in Fig. 7(c,f), the PM$_{2.5}$ total net flux in July and October was shown in the vertical direction as follows: near-surface (+), mid-altitude (-), high altitude (+), and the sum of flux in polluted days was 1.71 times, 6.80 times, and 2.04 times that of the clean days, respectively.

On the whole, the main contributor cities to the PM$_{2.5}$ net inflow flux and net outflow flux in polluted days and clean days in the vertical direction were similar. The net inflow flux mainly came from Baoding and Langfang, with the peak in polluted days...
(clean days) of 90 t/d (58 t/d) and 108 t/d (46 t/d), respectively. Chengde was the main receiver of the net outflow flux, with the peak in polluted days (clean days) of -224 t/d (-102 t/d).

For the polluted days in July, Langfang (N) and Tianjin were the main contributors of the PM$_{2.5}$ net inflow flux, and the net outflow flux was mainly transported to Zhangjiakou, Chengde, Langfang (S), and Baoding. The total net flux appeared at a relatively high level between 252 m – 817 m, ranging from -107 t/d to 125 t/d. But Beijing mainly received the net inflow from Baoding and Langfang and transported the net outflow to Chengde in clean days, with a relatively lower level of the total net flux (-3 – 25 t/d). In October, there was no significant difference in the net flux between polluted days and clean days. Chengde was the main receiving city of the net outflow flux, Baoding and Langfang were the main contributing cities of the net inflow flux. It is noteworthy that the net flux in polluted days in October was significantly higher than that in clean days. Taking Baoding and Chengde, the cities with the largest net inflow and net outflow respectively, as examples: the maximum net flux in the vertical direction of Baoding and Chengde in polluted days was 225 t/d and -331 t/d respectively, the corresponding maximum net flux in clean days was 47 t/d and -85 t/d respectively, and the former was 4.82 times and 4.63 times the latter, respectively, so as to explain that the mutual transportation between Beijing and surrounding cities during the PM$_{2.5}$ pollution period was relatively significant.
3.3 Hourly evolution of PM$_{2.5}$ transport fluxes during a heavy pollution process

To further clarify the evolution characteristics of the PM$_{2.5}$ transport flux at different stages of the heavy pollution process, we took a period in Beijing from 0:00 on the 17th to 23:00 on the 21st in October 2016 as an example and divided the heavy pollution into three stages: Stage I (0:00 on the 17th ~16:00 on the 18th), Stage II (17:00 on the 18th ~ 7:00 on the 20th), Stage III (8:00 on the 20th ~ 23:00 on the 21st).

In the vertical direction, it is divided into low-altitude (below 400m) and high-altitude (above 400m). The time-wise changes of PM$_{2.5}$ inflow, outflow, and net flux in the three stages of heavy pollution were analyzed, as shown in Fig. 8. On the whole, the temporal-spatial evolution characteristics of PM$_{2.5}$ transport flux in low altitude and high altitude were relatively consistent. The total inflow, outflow, and net flux of high altitude PM$_{2.5}$ were 2.64, 2.15, and 5.13 times that of low altitude, respectively.
Results of the low altitude were analyzed as follows: the average flux of the total inflow, outflow, and net of PM$_{2.5}$ in the entire heavy pollution process was 1380 t/d, -1470 t/d, and -90 t/d, respectively. The total inflow and outflow fluxes were 1.32 and 1.61 times the monthly average flux of the low altitude in October, respectively, so as explained that the influence of the transportation during heavy pollution process has increased compared to the entire October. In the early stage of heavy pollution (Stage I), PM$_{2.5}$ total inflow and outflow flux increased slowly at a low level, with the average flux of 790 t/d and -610 kt/d, respectively. While Beijing accepted the inflow flux mainly from Baoding, it also continues to release outflow flux to Chengde. The total net flux was basically positive, indicating that adjacent cities had a continuous net input to Beijing, which led to a slow increase in Beijing’s PM$_{2.5}$ concentration, from 8 $\mu$g/m$^3$ at 0:00 on the 17th to 109 $\mu$g/m$^3$ at 12:00 on the 18th, which reached the first peak of the heavy pollution process.
Subsequently, PM$_{2.5}$ concentration remained at a high level into the Stage II of heavy pollution, and the PM$_{2.5}$ total inflow and outflow flux were significantly enhanced compared with Stage I, with the flux of 2720 t/d and -3050 t/d, respectively. However, the net flux was -330 t/d, so as indicated that the amount of PM$_{2.5}$ outputted from Beijing in stage II was higher than the amount inputted to itself by adjacent cities. From 20:00 on the 18th to 15:00 on the 19th, the PM$_{2.5}$ total net flux basically remained positive, with an average flux of 840 t/d, of which the net inflow flux mainly came from Baoding and Langfang (S), with an average flux of 701 and 752 t/d, respectively. The net outflow flux was mainly transported to Chengde with a mean
flux of -1221 t/d. As a result, PM$_{2.5}$ concentration maintained a continuous increase from 99 $\mu$g/m$^3$ to 183 $\mu$g/m$^3$, indicating that regional transportation played an important role in heavy pollution before it reached the extreme concentration. Subsequently, the PM$_{2.5}$ total net flux showed a high negative value, especially the net flux reached -8731 t/d at 3:00 on the 20th, where Baoding was the largest recipient of outflow flux with the highest net flux of -5700 t/d, and Tianjin and Langfang (N) were the largest contributors of inflow flux with the highest net flux of 2842 t/d. It is noteworthy that PM$_{2.5}$ concentration didn’t decrease due to the transportation (local to adjacent cities), but continued to rise, and reached the second (218 $\mu$g/m$^3$) and third concentration peaks (225 $\mu$g/m$^3$) at 19:00 on the 19th and 2:00 on the 20th respectively, thus indicating that this peak was mainly due to the contribution of local emissions in Beijing. In other words, local emissions have a greater potential to cause extreme PM$_{2.5}$ heavy pollution to occur, while external transport tends to have a significant impact before heavy pollution peaks, which was relatively consistent with the findings of Li et al.(2017b) and Tang et al.(2015). Therefore, in the 1~2 days before the PM$_{2.5}$ concentration reached the peak of heavy pollution, strengthening emergency emission reduction measures in neighboring cities will achieve a better peak shaving effect. After 2:00 on the 20th, influenced by northerly winds, clean air from Chengde transported into Beijing, and Beijing continued to export to Baoding and Langfang (S), and the average net flux of Baoding was as high as -4000 t/d, resulting in a sharp drop in Beijing PM$_{2.5}$ concentration to 42 $\mu$g/m$^3$ within 5 hours. In Stage III, PM$_{2.5}$ total inflow, outflow, and net flux were relatively lower, with mean
fluxes of 682, -831, and 149 t/d, respectively. PM$_{2.5}$ concentrations continued to
remain a low-level, basically below 40 μg/m$^3$, marking the basic end of this heavy
pollution.

Note that the characteristics of PM$_{2.5}$ flux at the higher–altitude layer were
almost similar to that of the lower–altitude layer (Fig. 8b). This demonstrated that
wind direction was more uniform at various altitudes, which were not conductive to
the vertical diffusion of haze pollution. Additionally, there were large positive inflows
from Zhangjiakou at the higher–altitude layer during stage II, which was mainly due
to the strong winds rather than high PM$_{2.5}$ concentration. On the whole, the total
inflow, outflow and net fluxes at the higher–altitude layer were probably two times
the average at the lower–altitude layer, indicating that transport contribution at the
higher–altitude layer may have a greater influence on regional air quality compared to
that of the lower–altitude layer.

4 CONCLUSION

Significant transport effects existed between Beijing and its adjacent cities in
July and October, and seasonal differences were evident, with both PM$_{2.5}$ total inflow
and outflow flux intensities higher in autumn than in summer. Beijing exported more
impact outward than received external inflow in summer and vice versa in autumn.
The vertical distribution of PM$_{2.5}$ net fluxes was significant, and maximum net fluxes
in July and October appeared at 817 m and 1261 m above the ground, respectively.
Both Baoding and Langfang showed a net inflow to Beijing in the vertical direction,
while Beijing showed a net outflow to Chengde.
Total average PM$_{2.5}$ net fluxes on polluted days and clean days in July and October were consistently distributed vertically, and the former was 1.71 to 6.80 times higher than the latter. Spatial and temporal distributions of PM$_{2.5}$ transport fluxes at low and high altitudes during heavy pollution were consistent, where the average PM$_{2.5}$ total inflow, outflow, and net fluxes at low altitude were 1380 t/d, -1470 t/d, and -90 t/d, respectively, and the flux at high altitude was 2.15 to 5.30 times higher than at low altitude. Meanwhile, we found that local discharging was more likely to cause extreme PM$_{2.5}$ heavy pollution, and a better peak clipping effect can be achieved when emission reduction measures are initiated 1 to 2 days before heavy pollution.

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