Temporal and spatial characteristics of PM$_{2.5}$ transport fluxes of typical inland and coastal cities in China

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**Article Info**

**ABSTRACT**

Local pollution and the cross-boundary transmission of pollutants between cities have an inevitable impact on the atmosphere. Quantitative assessments of the contribution of transport to pollution in inland and coastal cities are necessary for the implementation of practical, regional, and joint emission control strategies. In this study, the Comprehensive Air Quality Model (CAMx), together with the Weather Research and Forecasting model (WRF), was used to simulate the contributions to pollution of different cities in 2016. The monthly inflow, outflow, and net flux from the ground to the extended layers served as the three main indicators for the analysis of the interactions of PM$_{2.5}$ transport between adjacent cities. Between inland and coastal cities, the magnitude of inflow and outflow are larger in the former than in the latter. The inflow flux in the inland cities (Beijing and Shijiazhuang) was 10.6 and 10.7 kt/day, respectively, while that in the coastal cities (Tianjin, Shanghai, Hefei, Nanjing, and Hangzhou) was 9.1, 3.3, 5.8, 4.4, and 3.7 kt/day, respectively. In terms of variation over the year, the strongest inflow in the BTH region occurred in April, followed by October, July, and January, while that in the coastal cities in YRD occurred in January, followed by October, April, and July. Therefore, based on the flux intensity calculations and the transport flux pathways, effective joint control measures can be provided with scientific support, and a better understanding of the evolutionary mechanism among inland and coastal cities can be acquired.

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Introduction

Over the past decades, China has undergone rapid economic growth, urbanization, and industrialization. In connection with these, the country experiences regional ambient air pollution, which occurs frequently in the autumn and winter (Miao et al., 2018; Yang et al., 2020). Aside from local pollutant emissions, inter-city transport (both in inland and coastal cities) could also contribute to the production of PM$_{2.5}$ pollution over large, distributed regions (Cai et al., 2017). Beijing–Tianjin–Hebei (BTH) and Yangtze River Delta (YRD) are two typical city clusters, which became significant driving forces in the economic development of China; however, the annual average PM$_{2.5}$ concentrations in these city clusters often exceeds the National Ambient Air Quality Standard (NAAQS) for annual PM$_{2.5}$ (35 μg/m$^3$) (Cheng et al., 2013a; Cheng et al., 2013b; Song et al., 2016; Zhang et al., 2018). To reduce the frequency of haze episodes, the "Atmospheric Pollution Prevention and Control Action Plan Police" was created in September 2013.

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Thus, a deep understanding of the variation of the regional transport of air pollutants between inland and coastal cities is essential for the air quality management of the megacity and the development of regional-scale control measures.

Recognizing the import role of transport on PM$_{2.5}$ pollution, recent studies have been conducted in different cities in China using various qualitative and quantitative methods. Based on the theories of Lagrangian and Eulerian, those two methods have been derived kinds of methods to explore the characteristics of PM$_{2.5}$ transport and diffusion. As for qualitative analysis, some researches explored the Hybrid Single Particle Lagrangian Integrated Trajectory Model to calculated and investigate the air mass trajectories. And coupled with cluster analysis (Wang et al., 2009), potential source contribution function (Wei et al., 2019) or concentration weighted trajectory (Kang et al., 2020), different pollution is associated with trajectories to identify the possible source areas and transport pathways reaching the receptor site. Without regional influence and lacking pollution emission information restriction, qualitative methods are widely applied both inland (Wang et al., 2015a) and coastal cities (Fu et al., 2016). However, the analytical methods consider only atmospheric dynamics and neglect chemical reactions, which dissipate the requirement of quantifying and assessing regional transport contributions to the concentrations of PM$_{2.5}$.

In contrary, quantitative methods, named three-dimensional Eulerian chemical transport models could solve larger study domain and is ideal for multiple sources appointment, handling complex atmospheric chemistry mechanism and even forecasting pollutant trend. The models are consisted of Community Multi-scale Air Quality (CMAQ), the Comprehensive Air Quality Model with Extension (CAMx), the Weather Research and Forecasting model coupled with Chemistry (WRF/chem), the Nested Air Quality Predicting Modeling System (NAQPMS) and the rest. Wu et al (2017) applied WRF-chem model to evaluate the contributions of trans-boundary transport to the air quality in Beijing and figured out that the air quality in receptor city is generally determined by the trans-boundary during a persistent air pollution episode. For coastal cities, such as YRD region (Ding et al., 2013), numerical studies always paid more attention to the transported dust particles contributed to the mean surface layer concentrations of PM concentration. But neglect the different altitudes transport characterize and make unclearly transport flux among each boundary surrounding the target cities. Based on the Particulate Source Apportionment Technology (PSAT) and the Integrated Source Apportionment Model (ISAM), Chang et al. (2019) established the Transport Flux Calculation (TFC) method which can investigate the evolutionary trend of PM$_{2.5}$ transport flux at different altitudes and effectively distinguish the inflow and outflow flux between local and adjacent regions. Zhang et al. (2019b) applied the same method to calculate the transport flux across districts, cities, and regions in northern China. However, affected by the monsoon circulation and underlying surface, inland and coastal cities may have different variation on the boundary transport or different altitudes, the previous researches focus on the single areas transport flux characterizes. In a word, no matter inland and coastal cities, there are insufficient for investigating the spatial and temporal distribution of PM$_{2.5}$ flux in both BTH and YRD regions, especially for their difference.

In this study, we applied the PSAT method and the TFC method to explore the characteristics of the PM$_{2.5}$ transport flux in four aspects. First, a comprehensive discussion on the spatial and temporal variations in the PM$_{2.5}$ transport flux in the BTH and YRD regions is conducted, which focus more on the variation in cross-inland and cross-costal transport. Second, inflow, outflow, and net flow are taken as the three principal indexes for the quantification of the PM$_{2.5}$ flux balance, and net flow is used to identify the PM$_{2.5}$ transport pathway. Finally, the transport contributions of the prefecture level cities, both inland and costal, are assessed to support the development of a comprehensive joint control strategy.

1. Materials and methods

1.1. Study areas

The BTH and the YRD regions are metropolitan areas and the economic centers in China. Seven capital cities were considered to study the temporal and spatial variation in the PM$_{2.5}$ concentrations and transport flux. In the BTH region, we selected Beijing (BJ), Tianjin (TJ), and Shijiazhuang (SJZ); in the YRD region, we chose Hangzhou (HZ), Shanghai (SH), Hefei (HF), and Nanjing (NJ). According to the Multi-resolution Emission Inventory for China, the total PM$_{2.5}$ emission from different anthropogenic sources was 8.1 Tg in 2016, in which the BTH and YRD regions accounted for 8.5% and 14.3% of the total amount, respectively (http://meicmodel.org, Liu et al., 2015). Among the cities with the highest amount of PM$_{2.5}$ emissions in China, Hebei in the BTH region and Anhui in the YRD region ranked the second and fourth, respectively. In this paper, the study area was set from 25° N to 45° N and 105° E to 120° E, covering all cities of the two regions, as shown in Fig. 1.

1.2. WRF/CAMx-PSAT model system

1.2.1. Model configuration

In this study, we employed the WRF/CAMx-PSAT modeling system to investigate the flux transport of anthropogenic emission sources in 2016 over the study regions. A two-level nested-grid modeling domain was established for the simulation, with the first five days being considered as model spin-up to minimize the impact of initial chemical files, as shown in Fig. 1. Domain 1 covered most of eastern and central China, with a spatial resolution of 36 km x 36 km (120 rows and 171 columns). Domain 2 covered the two regions, with a spatial resolution of 12 km x 12 km (223 rows and 163 columns). The simulation of Domain 1 was regarded as the time-varying boundary conditions for Domain 2, while a fixed profile available in CAMx-PSAT was used for itself. Both Domains 1 and 2 were set with 28 layers vertically extending from the surface to an altitude of 20 km aboveground, which were consistent with the WRF model. The configuring details and the input data of the modeling system are summarized as follows:

Emission data. The input emission files required by CAMx were prepared using the SMOKE model. The emission data
used in this study mainly include (1) the BTH region emission inventory developed by Zhou et al. (2014), which was calculated through the categories of activities and their emission coefficients; (2) the YRD region emission inventory developed by Sun et al. (2018); and (3) the emission inventory outside BTH and YRD of anthropogenic sources in 2016 from Tsinghua University, with a spatial and time resolution of 0.25° × 0.25° and a one-month interval.

The anthropogenic emissions in the inventory were divided into point sources, line sources, and area sources, which include industrial, fugitive dust, VOC, residential road and non-road mobile, and biomass burning sources, among others. Biogenic emissions include those from straw, firewood, livestock feces burning, and forest and grassland fires, whose dataset was found in the China Statistical Yearbook (http://www.stats.gov.cn/), and the estimation formula was obtained from a previous study (Xing et al., 2018). The pollutants in the emission inventory include of PM$_{2.5}$, PM$_{10}$, SO$_2$, NOx, VOC, CO, and NH$_3$.

**Meteorological data.** The meteorological fields required by CAMx were simulated using the WRF model, which utilize final (FNL) operational global analysis data from the Global Forecast System developed by the National Center for Environmental Prediction (http://rda.ucar.edu/datasets/ds083.2), with a spatial resolution of 1° × 1° and a temporal resolution of 6 h, which serve as the meteorological initial and boundary conditions.

**PSAT method.** PSAT is an extension tool of CAMx, which was employed in the pollution diagnosis of source appointments, zonal markings, and chemical species (Wagstrom et al., 2008). Using this method, we could mark the source regions and select the coordinates of the receptor points, as well as trace particles and precursors. In this study, CAMx-PSAT was utilized to simulate the specified grid point pollution concentration and calculate the transport flux of PM$_{2.5}$ over the surface and vertical of cities.

**Model configurations.** Version 3.5.1 of the WRF model was used in this study. Purdue Lin, Yonsei University, and Noah and Grell-Devenyi were chosen as the Microphysics, Planetary Boundary Layer (PBL), and Land-Surface and Cumulus schemes, respectively. The radiation schemes used were the Goddard shortwave radiation and Rapid Radiative Transfer Model longwave radiation. Version of 6.3.0 of the CAMx model was employed. Its vertical resolution was configured with 28 layers, which was consistent with the WRF model. The gaseous and aerosol modules used were the Carbon Bond-05 mechanism with chlorine and updated toluene chemistry (Sarwar et al., 2008) and the sixth-generation modal CAMx aerosol mode with extensions for sea salt emissions and thermodynamics (Whitten et al., 2010; Yarwood et al., 2010), respectively.

The anthropogenic sources emission was adopted to identify the impact of PM$_{2.5}$ flux transport over the two city clusters. The target periods of the simulation were set to January, April, July, and October of 2016, which represent winter, spring, summer, and autumn, respectively.

1.2.2. **Model evaluation**

For the simulation performance of the WRF and CAMx model, the correlation coefficient (COR), mean fractional bias (MFB), mean fractional error (MFE), normalized mean bias (NMB), and normalized mean error (NME) were selected as statistical parameters. Moreover, the fraction of predictions within a factor of two of observations (FAC2) used by Hanna (1993) and the index of agreement (IOA) used by Duveiller et al. (2016) were also utilized in this study. The detailed functions are found in **Supplement Appendix A.** The meteorological parameters were acquired from the Chinese Meteorological Information Comprehensive Analysis and Process System, while the surface PM$_{2.5}$ concentration data were obtained from China National Environmental Monitoring Center (CNEMC; http://106.37.208.233:20035/). We compared the observation
and simulation of the PM$_{2.5}$ concentrations and the major meteorological parameters for January, April, July, and October. As shown in Tables S1 and S2, T2 was well reproduced, with the CORs being generally greater than 0.70. For WS10, the NMB ranged from -39.90% to -15.94%, and the NME ranged from 15.84% to 48.52%. In terms of RH, the simulations were universally underestimated, agreeing well with the observations; CORs were from 0.43 to 0.68, the NMB ranged from -15.63% to 34.52%, and NME ranged from 11.04% to 45.91%. Overall, the simulated meteorological field corresponds with the observations and reproduces the characteristics of temporal variations and magnitudes.

Tables 1 and S3 display the comparison of the month average simulations and observations at the two main city clusters located in Domain 2. For January, the CAMx model showed a better performance, with the COR being higher than 0.8 and having an NMB of 10.3%, NME of 31.8%, MFB of -15.3%, MFE of 39.0%, FAC2 of 0.97, and IOA of 87.1%. For April, July, and October, the CAMx model also yielded a better performance, with CORs ranging from 0.5 to 0.7, and the variations in NMB, NME, MFB, and MFE were in the range of 21.3% to 13.7%, 37.4% to 41.3%, -23.7% to -40.9%, and 44.9% to 53.3%, respectively. Obviously, there is a bigger margin of error compared with the observation in January. This resulted from the slightly underestimated PM$_{2.5}$ concentrations compared to observations during the haze episodes, especially in the autumn and winter (Li and Han, 2016). Numerous studies attributed the underestimation of PM$_{2.5}$ concentrations to several reasons, such as uncertainties in the emission inventory (Zhou et al., 2015), underestimation of RH and WS10 meteorological variables (Wang et al., 2015b), and drawbacks in the model mechanism drawback leading to major components biases (Zhao et al., 2016), among others.

Thus, in order to obtain a comprehensive and in-depth understanding of the characteristics of the variation, we observed PM$_{2.5}$ levels based on URG samplers in the JJJ region (BJ and SJZ). The detailed sampling and testing information can be found in our previous studies (Zhang et al., 2019a). Due to the shortage of observational data on PM$_{2.5}$ components in the YRD regions, we evaluated the model performance of PM$_{2.5}$ components beyond the JJJ region by comparing the simulation results with the observational data from literature, as shown in Table 2. The coverage for the observations for SO$_2$$_2$-, NO$_2$$_2$-, NH$_4$$_4$+, and OM from literature was generally similar to that in this research in both duration and space. Although some observation periods do not exactly match the modeling periods, the results should still be comparable. It is noted that the simulation results generally agree with the observations. Considering these uncertainties and other studies concerning simulated results, the performance of CAMx for simulating the temporal and spatial distribution of the total major chemical components of PM$_{2.5}$ was reasonable for the study periods.

**1.3. PM$_{2.5}$ flux calculation**

In this study, PM$_{2.5}$ flux represents the mass of PM$_{2.5}$ that flows through a special vertical surface during a certain period of time. The position of the flux calculation was chosen as the intersecting grids along the boundary line between the target city and the adjacent cities extending up to a specified vertical height, which was consistent with Chang et al. (2018) and Zhang et al. (2019b). Therefore, the vertical surface through the flux was discretized into a series of vertical grid cells, with the WRF and CAMx model having the same number vertical layers in order to extract PM$_{2.5}$ concentrations and wind vectors on the corresponding vertical layer. The expression for PM$_{2.5}$ flux is as follows:

$$\text{Flux} = \sum_{h=1}^{H} \sum_{l} L \cdot H_h \cdot c \cdot v \cdot n$$

where, $H$ is the top layer; $l$ is the boundary line of two adjacent regions; $H_h$ is the vertical height between layer $h$ and $h+1$; $L$ is

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<th>Table 1 – Performance statistics for PM$_{2.5}$ concentrations over two regions of city clusters.</th>
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$^a$ NMB: Normalized Mean Bias.  
$^b$ NME: Normalized Mean Error.  
$^c$ MFB: Mean Fractional Bias.  
$^d$ MFE: Mean Fractional Error.  
$^e$ FAC2: Fraction of predictions within a factor of two of observations.  
$^f$ IOA: Index of Agreement.

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<th>Table 2 – Comparison of PM$_{2.5}$ components in two regions of city clusters (units: µg/m$^3$)</th>
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(Zhang et al., 2019c)
the width of grid cell; c is the concentration of PM$_{2.5}$ in the vertical grid cell; u is the wind vector; and n is the normal vector of vertical grid cell. Meanwhile, the 12$^{th}$ layer (approximately 1.8 km above the ground) was regarded as the top layer to investigate the vertical distribution of PM$_{2.5}$ flux. While the 13$^{th}$ layer is a higher vertical layer that could still transport flux, it was unimportant for the near-ground concentrations due to the weaker vertical mixing above the boundary layer. The BTH and the YRD regions are two of the most important and developed megacity clusters in China. Thus, we chose them as the target cities for the flux calculation. Three indicators, namely the inflow, outflow, and net fluxes, were used to describe the flow of PM$_{2.5}$ mass between adjacent cities. To quantitatively distinguish the transport between different adjacent areas and to obtain the three flux indexes, all administrative boundaries between the target city and the adjacent cities were considered as boundary lines. The locations of the seven target cities and their neighbors are shown in Fig. 1. Flux varies every time with the wind direction changes. Inflow flux is positive (+), and it indicates the flow of PM$_{2.5}$ from the adjacent cities into the target city; outflow flux is negative (-), and it represents PM$_{2.5}$ that flows out of the target city into the adjacent cities; and the net flux is the offset between the inflow and outflow flux. All of these indicators characterize the intensity of interactions between two regions and the general impact of PM$_{2.5}$ transport.

Notably, it was difficult to directly acquire vertical observations of PM$_{2.5}$. We reviewed previous researches and found that Siosmos et al. (2017) established a function relationship between the mass concentration of PM$_{2.5}$ and atmospheric transmissivity and calculated the vertical distribution of PM$_{2.5}$ concentration inversion using a lidar and atmospheric transmission meter and a particle spectrometer. In this study, we collected fixed-point lidar data to inversely calculate the vertical distribution of PM$_{2.5}$ concentrations and coupled them with the corresponding altitude wind vector to calculate the observed transport flux. The fixed-point station is located in CNEMC (40.04° N, 116.42° E) from January 14 to 17. With the observations, we evaluated the simulation performance, which is described in detail in Section 2.5.

2. Results and discussion

2.1. Spatial-temporal variation of PM$_{2.5}$ in the study area

The seasonal average mass concentrations for PM$_{2.5}$ and PM$_{10}$ were 29.8–110.5 μg/m$^3$ and 49.4–170.8 μg/m$^3$ in the two city clusters (Fig. 2). They both varied significantly with seasonal variations. The highest values generally occurred in the winter at 89.0 ± 37.5 μg/m$^3$ and 135.6 ± 48.6 μg/m$^3$ for PM$_{2.5}$ and PM$_{10}$ in the BTH region and at 76.4 ± 15.7 μg/m$^3$ and 107.4 ± 23.1 μg/m$^3$ in the YRD region, respectively. The lowest values occurred in summer at 57.4 ± 13.1 μg/m$^3$ and 87.6 ± 15.5 μg/m$^3$ for PM$_{2.5}$ and PM$_{10}$ in the BTH region and at 29.8 ± 6.1 μg/m$^3$ and 49.4 ± 11.0 in the YRD region, correspondingly. Meanwhile, the average seasonal ratio of PM$_{2.5}$/PM$_{10}$ varied from 44.1% to 71.1% in the two regions. The order of the seasons, in decreasing average seasonal ratio, is winter (67.6% ± 8.4%), summer (59.4% ± 8.2%), autumn (57.9% ± 8.3%), and spring (51.2% ± 10.5%). This indicates that there is generally a greater amount of PM$_{2.5}$ than PM$_{10}$, especially in the winter. PM$_{2.5}$ and PM$_{10}$ concentrations reached the highest in winter because of poor atmospheric diffusion conditions (e.g. lower PBL height and higher and calmer winds) and increased emissions (e.g. coarse coal particles combustion in regions needing heat). On the other hand, better diffusion conditions, including the higher frequency of precipitation and stronger winds, occur in spring and summer, which help reduce the atmospheric PM$_{2.5}$ and PM$_{10}$ concentrations during these seasons (Shen et al., 2015).

PM$_{2.5}$ and PM$_{10}$ concentrations exhibited similar fluctuations in the two regions (Fig. 2). Additionally, haze episodes at PM$_{2.5}$ concentrations > 150 μg/m$^3$ and non-haze episodes at
\(\text{PM}_{2.5}\) concentrations \(\leq 150 \mu g/m^3\) appeared alternatively. During haze days, there were high mass loadings at 206.2 ± 54.7 \(\mu g/m^3\), as opposed to non-haze days where mass loading was 56.6 ± 30.0 \(\mu g/m^3\). \(\text{PM}_{2.5}\) and \(\text{PM}_{10}\) concentrations were higher in the BTH region than in the YRD region. \(\text{PM}_{2.5}\) and \(\text{PM}_{10}\) concentrations in BTH was almost 1.4 times higher than those observed in YRD, due to significant increase in coal combustion during winter and higher density of population of the former. Furthermore, BTH is located in the east of Taihang Mountain and Yanshan Mountain, whose disadvantageous terrain conditions block the northwest wind, leading to pollutant accumulation and the aggravation of air pollution. Although \(\text{PM}_{2.5}\) and \(\text{PM}_{10}\) concentrations in YRD were relatively lower than those in BTH, they still do not comply with the Grade II level requirements of CNAQs. The causes of air pollution in these two regions are different. Air quality in BTH has always been greatly affected and challenged by the development of secondary industries, especially the iron and steel manufacturing industry and crude steel productions, which account for 26.08% of the national \(\text{PM}\) concentrations (Ren and Xu, 2018).

### 2.2. Regional transport contribution of surface \(\text{PM}_{2.5}\) to seven target cities

To investigate the variation of source apportionment from different regions to surface \(\text{PM}_{2.5}\) concentrations in BTH and YRD regions during the four seasons is shown in Supplement Appendix C. It can be found that no matter BTH and YRD regions, the local emissions are primary contribution source, with 56.28%-65.81% (for BTH) and 59.49%-67.72% (for YRD) during the four seasons, which suggested air quality improvement mainly need local pollution reduction. Especially in October, local emissions contribution is higher than other three seasons and the phenomenon are more obvious in the BTH region. One conceivable explanation regarding the various in those two regions was that the utilization rate of residential coal burning increases significantly cause by the temperature drop in winter, while in YRD region residential coal burning without permission by the policy. Apart from that, regional transport has closely related to the distance of the source region. For example, Shandong province, located between BTH and YRD regions, has basically same transmission contribution for each two regions, with 6.14%-11.42% for BTH and 9.23%-12.29% respectively. As for Inner Mongolia, the regional contribution has further difference, with 5.37% for BTH and 1.24% for YRD in October. This phenomenon indicated that both local and regional transport, especially from circumjacent regions played an important role in the air pollution.

### 2.3. Comparative analysis of flux across inland cities and coastal cities

#### 2.3.1. Overview of simulated \(\text{PM}_{2.5}\) transport flux across the cities

To obtain an in-depth understanding of the transport characteristics through each boundary segment of the seven target cities, \(\text{PM}_{2.5}\) fluxes were calculated based on Eq. (1). Fig. 3 shows the inflow, outflow, and net fluxes of the seven target cities during the four selected months. In general, the intensity of transport fluxes was associated with the characteristic of the local emission structure and the emission intensity from the dominant wind direction; the intensity of transport flux in inland cities were often higher in magnitude than that in coastal cities. For instance, the total four-month inflow flux intensity in the inland cities BJ and SJZ were 10.6 and 10.7 kt/day, respectively, while that in the coastal cities TJ, SH, HF, NJ, and HZ in the two regions were 9.1, 3.3, 5.8, 4.4, and 3.7 kt/day, respectively. As for the gross of outflow flux, the magnitudes were -10.1 to -10.3 kt/day for the inland cities and -3.8 to -11.1 kt/day for the coastal cities. The reason for this phenomenon is the frequent sea-land air exchange in the coastal cities, in which clean air from the sea dilutes the pollutant concentration and reduces the magnitude of the transport fluxes. Therefore, both inflow and outflow fluxes in the inland cities were 1.9–3.2 times and 1.6–2.6 times stronger than those in coastal cities as, respectively, due to the poor diffusion condition and heavy emission structure in the former. However, although TJ is a typical coastal city, it contributes 1.6–2.8 times more inflow flux and 1.7–2.8 times more outflow flux than other coastal cities, because it is surrounded by the Tangshan cities which produces more than 50% of the steel production capacity of Hebei province (Yang et al., 2019).

We summarized the variation of flow characteristics based on the month to obtain an overview of the trends of \(\text{PM}_{2.5}\) transport during the representative months. It is clear that inflow flux and outflow flux have similar characteristics for each month, both for inland cities and coastal cities, whereas the net flux varies. For inflow flux, the order of the months in decreasing mean intensity was April, October, July, and January in the BTH region. The flow in the month with the strongest flux was 1.2–2.2 greater than that of the month with the weakest flux, although the inflow in SJZ in January was 1.3 times larger than that in July, which was consistent with the findings of Zhang et al. (2019b). However, dissimilar to the inland cities, the strongest inflow in the representative cities in YRD occurred in January, followed by October, April, and July, with inflows in January being 2.6–3.8 times greater than those in July. In terms of the magnitude of the average absolute outflow flux, the pattern of seasonal fluctuations was similar to that of the inflow, with the absolute values in the strongest month being 1.6–2.9 and 2.7–3.7 times greater than those of the weakest month in the BTH and YRD regions, respectively. The total net flux intensity between inland and coastal cities has a completely different seasonal variation. For inland cities, such as BJ and SJZ, inflow generally exceeded the absolute magnitude of outflow, indicating that the four cities act as \(\text{PM}_{2.5}\) “sinks.” In contrast, in the coastal cities, the absolute magnitude of outflow generally exceeded inflow, which implies that they serve as “source” cities. This phenomenon is possibly caused by the unique terrain of “convergence” cities such as BJ and SJZ, which are surrounded on the west and north by mountains and major emissions sources of \(\text{PM}_{2.5}\) in the south and east. Meanwhile, with the influence of the sea and land breeze in TJ and the coastal cities in the YRD, the mesoscale circulation in the lower atmosphere facilitates the transport and diffusion of local air pollutants (Lin et al., 2015). Based on the foregoing, we compared the flow characteristics of \(\text{PM}_{2.5}\) in the study areas to help us understand the extent at which cities interact with their neighbors during the different representative months (Fig. 3). For BJ, the majority of the inflow was con-
Fig. 3 – Simulated PM$_{2.5}$ inflow, outflow, and net fluxes in the two regions in January, April, July, and October.
tributed by Zhangjiakou and Baoding, while most of the outflow were conveyed to Chengde and Langfang in January and April. In contrast, Zhangjiakou and Chengde received major fluxes in July and October, while the inflow was similar to that in January and April. One possible reason for this is the favorable meteorological conditions in Zhangjiakou and Chengde that allow pollutants to diffuse more easily to their surrounding cities (Zhu et al., 2018). A similar pattern could be observed in both inland and coastal cities. In terms of large-scale circulation patterns associated with the East Asia winter monsoon, the North China Plain experience westerly winds, properties of which rely largely on the development of both the Siberian High and the Aleutian Low (Jhun and Lee, 2004).

2.3.2. Vertical distribution of PM$_{2.5}$ net flux across the cities

To explain the vertical variation of net flux at different altitudes across the city and determine the height range at which the vertical transport occurs, we calculated the vertical distribution of monthly net flux during the four representative months. The results are shown in Fig. 4.

Over all the vertical layers, the total PM$_{2.5}$ net flux had either one or three peaks; layers with larger net fluxes occurred at medium and high altitudes; and the major contributors vary with the approximate elevation of each layer. Influenced by topography and the effect of the monsoon climate, different cities served different roles with respect to their surroundings. The BTH region (except TJ), mainly including inland cities, generally played the role of a "source" for its adjacent cities. The vertical flux intensity was the strongest in October, followed by January, July, and April; the positive peak values of the total net fluxes were 29.13, 25.38, 53.59, and 48.93 ton/day, respectively, in BJ cities. In the YRD, a cluster of coastal cities, the net flux pattern is completely different from that of the inland cities. The cities principally acted as "convergence" cities for its surrounding. The negative peak values of the total net fluxes were -41.28, -27.36, -36.10, and -36.72 ton/day in SH; -51.05, -20.64, -13.64, and -36.45 ton/day in HF; -25.44, -8.79, -5.79, and -8.76 ton/day in NJ; and -49.30, -11.42, -13.96, and -26.18 ton/day in ZJ, respectively.

According to the seasonal variation, the main inflow contributor for BJ was the steam from Zhangjiakou (11.96–97.64 t/day), and the total net fluxes peaked at the height of 200–800 m. In April, the net flux alternates between positive and negative values in BJ, with the variation of the net amplitude in BJ.
Fig. 4 – Vertical distribution of PM$_{2.5}$ net fluxes in seven cities of the two regions during the four representative months.
being larger than that in SJZ. Furthermore, the overall characteristics of the net flux in July displayed a similar pattern, that is, three capital cities served as “convergence” cities below 500 m and acted as a “source” up to 1000 m. This phenomenon is related to the wind speed; as the altitude increases, the wind rotating clockwise becomes stronger, according to the Ekman spiral (Holton and Hakim, 1973). As for October, except for the elevation of the net flux peak in contrast with that in July, the trend is essentially the same with that of BJ. For the coastal cities, it is noted that in SH, affected by its geographical area, the outflow is the strongest among the four cities. It is followed by HF, which is affected by coal burning for heating in the NCP in winter. The outflow in HZ and HF are third and fourth in intensity, as influenced by its geographical features.

2.4. Identification of PM$_{2.5}$ transport pathways

Considering the influence of inflow and outflow fluxes, we determined the various PM$_{2.5}$ transport pathways in the
two regions during the four representative months. Fig. 5 shows the transport flux through each boundary segment of the target cities. For BJ, there are two key transport pathways: the southwest-northeast direction (SW-NE, Baoding → BJ → Chengde) and the northwest-southeast direction (NW-SE, Zhangjiakou → BJ → Langfang), which were the transport pathways common in April. Indeed, the in-NW-SE transport pathway, which is ascribed to the winter monsoon in the North China Plain, under the Siberia, Eurasia, and Mongolia, high pressure quickly increases the frequency of the northwest wind. In July, the pivotal PM$_{2.5}$ transport pathways were in the southeast-northwest direction (SE-NW, Langfang → BJ → Chengde), and the SW-NE direction (Baoding → BJ → Zhangjiakou). The SW-NE pathway is caused by the summer monsoon which takes place in both lower and upper layers. When vertical mixing becomes stronger, it can remarkably affect transport and concentration, which further explains the phenomena that net flux peaks occurred in the lower and upper layers in July. In October, the principal pathways continue in the SE-NW and SW-NE directions in BJ. Comparing all the pathways on the BTH region, it is clear that, with the exception of opposite transport pathways during the monsoon in different seasons, the SW-NE pathway is the intrinsic transport pathway of the flux in the BTH region, regardless of the season. Moreover, for the representative coastal cities such as SH, there are also two main transport pathways: that in the SW-NE direction (Suzhou, Jiaxing → SH → Donghai Sea) and in the NW-SE direction (Nantong → SH → Zhanzhou) in January. It was verified that the dominant northwest wind in winter improves the transport pollution in the surrounding cities as opposed to the down the wind cities. For April and July, the chief pathway was in the SW-NE direction (Suzhou, Jiaxing → SH → Donghai Sea). In October, the pathway was changed to the northeast-southwest direction (NE-SW, Nantong → SH → Jiaxing). This implies that the cooperative control of transport in both inland and coastal cities should be taken into account for air quality improvement. The descriptions of the other regions are found in the Supplementary material.

To determine the relationship between wind and PM$_{2.5}$ concentrations, the frequency of wind direction, corresponding wind speed, and PM$_{2.5}$ concentrations were calculated and showed as “wind rose” plots. The plots for TJ were regarded as an example in Fig. 6, and the description of the other cities SH (for YRD) is found in the Supplementary Appendix D.

In January, the dominant winds at the lower layer were from the northwest, with the highest wind speed and frequency also occurring in this direction (Fig. 6a, b). Due to the large emission of pollutants in the BTH region and the convergence under the Mountain Taihang and Yanshan, PM$_{2.5}$ concentrations transported through the northwest wind were relatively high. Moreover, the wind directions and the corresponding concentrations were still dominant in the northwest wind at the higher altitudes (Fig. 6c, d). The speed of southwestern winds is much higher in the upper layers than in the lower layers, but PM$_{2.5}$ concentrations have the opposite characteristic. Therefore, the dominant NW-SE pathway was ascribed to the northwestern winds at both the lower and upper layers. In April, the most frequent wind became the southwest wind at lower and higher altitudes, and it had the highest corresponding PM$_{2.5}$ concentration (Fig. 6e, f). Emissions from Cangzhou could be transported into TJ via the southwest winds. Wind direction was chiefly from the south and its frequency also high, contributing to a strong output flux from TJ to Tangshan.

For July, the prevalent wind directions at the lower altitudes ranged from southwest to east. The east wind has the highest frequency and corresponding wind speed, which results in the summer monsoon and increase in the wind speed and frequency in coastal cities (Fig. 6i, j). During the summer monsoon, Bohai Sea and Tangshan changed as the main inflow flux. In addition, the dominant winds at the higher altitudes from the northeast were stronger than that at the lower altitudes, resulting in the Tangshan inflow flux being higher than that of Bohai Sea (Fig. 6k, l). In October, the frequency of the southwest and east winds were the highest (Fig. 6m, n). Northeast winds have the highest wind speed, while southwest winds have the highest PM$_{2.5}$ concentrations. The PM$_{2.5}$ concentrations carried via the stronger and more frequent southwest wind inflows from Cangzhou to TJ. On the other hand, wind and the corresponding PM$_{2.5}$ concentrations differed for the higher layer. The main winds shifted to the east and northeast directions, while the highest wind speed occurred in southwest (Fig. 6o, p). This better explain why Cangzhou contributed significantly to the inflows of TJ at both lower and upper layers.

### 2.5. PM$_{2.5}$ transport flux based on fixed-point lidar inversion observation

In order to compare the flux variations in the different vertical layers, fixed-point lidar observations were carried out for verification. Fig. 7 displays the total hourly simulated and observed net fluxes across the fixed-point station on January 14 to 17, from which we conducted the comparative analysis of the PM$_{2.5}$ flux to verify the accuracy of the flux modeling calculation. Different from the equation (1), the PM$_{2.5}$ flux for the fixed-point is calculated as follows:

$$\text{Flux}_{\text{NS}} = C_{\text{PM2.5}} \cdot V_{\text{NS}}$$

where $\text{Flux}_{\text{NS}}$ refers to the PM$_{2.5}$ flux intensity across the fixed-point in the north and south direction, $C_{\text{PM2.5}}$ refers to the PM$_{2.5}$ concentration, and $V_{\text{NS}}$ is the simulated wind vector of the north and south. The inflow flux was positive, indicating that PM$_{2.5}$ flowed across the station from south to north; the outflow flux was negative, showing that PM$_{2.5}$ flowed out of the station from north to south. As shown in Table 3, the PM$_{2.5}$ net flux simulations across the fixed-point station corresponded well with observations, with the CORs of 0.4–0.9, 0.7–0.9, 0.7–0.9, and 0.5–0.9, respectively, in different observation data. Moreover, the obtained NMB and NME varied between 1.3%–84.5%, 25.0%–66.3%, 21.7%–84.3%, -3.3%–89.8%, and 2.6%–80.5%, 16.2%–69.6%, 44.5%–84.9%, and 39.8%–89.8%, respectively. It is noteworthy that both the simulations and observations have similar daily variation trends, with the
Fig. 5 – Transport flux through each boundary segment of the two regions in January. The size of the arrows represents the magnitude of the net flux, and white and black arrows denote the net flux at the lower (layer 1–6 in the model) and upper (layer 7–12 in the model) layer, respectively.
highest outflow intensity occurring in January 15 (-1062.6 and -1381.7 μg/(m²·sec) and inflow intensity in January 16 (185.7 and 79.8 μg/(m²·sec)).

In-depth insights into the inflow and outflow across the fixed-point can improve our understanding of the variations in the PM$_{2.5}$ interactions between the station and its surroundings. As shown in Fig. 7a to d, it is evident that the most of the transport flux comes from the southern region, which is the main reason for heavy haze periods. Taking the hourly variation as an example, the intensities of the observed positive inflow fluxes was 1.84–102.31, 0.29–30.34, 7.85–185.87, and 2.43–158.84 (μg/m²·sec), respectively, while the magnitudes of the negative outflow fluxes were -0.87 to -32.66, -0.56 to -1381.7, -7.24 to -325.36, and 0, respectively. From the vertical distribution of the PM$_{2.5}$ transport flux, it can be seen that the flux intensity was generally stronger at lower and higher altitudes at medium altitudes, both for the positive inflow flux and the negative outflow flux during the observation. Especially in January 14 and 15, the transport flux tended to decrease gradually with altitude. In the former period, lower wind speeds caused the accumulation of pollutants, while in the latter one, higher wind speed contributed to the diffusion of pollutants and cleaning the air. In contrast to the aforementioned periods, the transport flux in January 16 showed a fluctuating positive and negative evolution trend, at the range of 7.85–185.87 μg/m²·sec in lower altitudes and around -325.36 μg/m²·sec at higher altitudes, which may be the principal reasons for the severe pollution levels in present day.
Fig. 7 – Simulated and observed PM$_{2.5}$ flux in the fixed-point in January 14 to 17.

Table 3 – Comparison and verification of transmission flux observation and simulation on the fixed-point.

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3. Conclusions

In this study, we investigated the PM$_{2.5}$ inflow, outflow, and net flux through seven cities, categorized into inland and coastal cities, in January, April, July, and October 2016 via the WRF and CAMx models. The comparison of the observed and simulated flux intensity and vertical distribution of PM$_{2.5}$ in inland and coastal cities demonstrated the accuracy of the model and its ability to simulate the characteristics of flux across cities.

Based on the results of the modeling of inland and coastal cities, it was shown that the intensity of the average inflows and outflows was the strongest in April for the capital cities in the BTH region and in January for the capital cities in the YRD region. Some representatives of the inland cities (BJ, SJZ) were regarded as “convergence” cities, characterized by more
intense inflows than outflows, while coastal cities (TJ, SH, HZ) mainly acted as "source" cities. Nevertheless, in April and July, the roles of "convergence" and "source" cities changed due to the decline in PM$_{2.5}$ concentrations because of the frequent occurrence of favorable weather conditions.

As for the PM$_{2.5}$ net flux, the value varied with the altitude, in that, the magnitudes were generally greater at the middle or upper layers for the seven cities in the BTH and YRD regions, implying that PM$_{2.5}$ transport does not only occur near ground but also affects air quality at a long range. In contrast, the net flux in inland cities (BJ, SJZ) showed stronger intensities at the low altitudes in January and October, suggesting that coordinated inter-regional prevention and control is essential for the BTH regions in autumn and winter. In coastal cities (SH, HZ, TJ and so on) higher magnitudes of net flux were observed at both lower and upper layers, indicating that joint prevention and control should not only be carried out for adjacent cities but also for cities that follow the downwind. According to the net fluxes and their vertical distribution, the vital transport pathways were identified in January, NW-SE in April, and SE-NW in July and October in the BTH capital cities. Therefore, there must be a greater focus on the intensity and path of high pressure when setting the range and time of joint emergency controls.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jes.2020.10.017.
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