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Investigation of Particulate Matter Regional Transport in Beijing Based on Numerical Simulation

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ABSTRACT

The frequent occurrence of regional air pollution makes it challenging to control. Based on source sensitivity research performed with the Chinese Unified Atmospheric Chemistry Environment (CUACE) model and dispersion simulation performed with the Flexible Particle dispersion model (FLEXPART), the regional transport of particulate matter (PM), potential source regions, and transport pathways were investigated for Beijing in summer (July) and winter (December) 2013. The mean near-surface trans-boundary contribution ratio (TBCR) of $PM_{2.5}$ in Beijing was 53.4% and 36.1% in summer and winter 2013, respectively, and 51.8% and 35.1% for PM_{10}. Regional transport in summer was more significant than that in winter. Seasonal difference of meteorological condition combined with the distribution of emission is responsible for seasonal difference of TBCR. The secondary aerosol is mostly contributed by regional transport. The transport of PM is mostly from Hebei province and Tianjin municipality. Based on backward trajectories analysis, the air mass source occurred from different directions in summer, while occurred from northwest in winter. The pollution level and the TBCR were closely related to the transport pathways and distance, especially in summer.

Keywords: Regional transport; Particulate matter; Backward trajectory; CUACE; FLEXPART.

INTRODUCTION

The particle matter (PM) suspending in atmosphere plays an important role in atmospheric visibility (Han *et al.*, 2013), cloud and microphysical properties (Andreae and Rosenfeld, 2008), global climate and the hydrological cycle (Ramanathan and Feng, 2009; Tai *et al.*, 2010). PM, especially fine particulates, is closely related to severe haze (Zhang *et al.*, 2015a) and has become the primary pollutant in most of Chinese cities in recent years (Liang *et al.*, 2014). The mean concentration of $PM_{2.5}$ (particulate matter with aerodynamic diameter less than 2.5 µm) in major Chinese cities was 5 times higher than the mean level of the world (Chai *et al.*, 2014), which has an adverse effect on human health (Tie *et al.*, 2009; Tao *et al.*, 2012). Although $PM_{2.5}$ concentration decreases about 23% during 2003 to 2010 in China (Zhou *et al.*, 2016), population-weighted mean $PM_{2.5}$ concentration in China is the highest value in the world's 10 most populous countries and increases significantly from 1990 to 2010 (Brauer *et al.*, 2016).

Air pollution is closely related to multi-scale meteorological conditions, pollutant emissions, the chemical processes in atmosphere and removal process (He *et al.*, 2016a). In economically developed regions, such as the Beijing-Tianjin-Hebei region, the Yangtze River Delta and the Pearl River Delta region in China, regional air pollution is serious and occurs frequently (Cheng *et al.*, 2013; Liu *et al.*, 2014; Kong *et al.*, 2015; Wang *et al.*, 2015a, b). Regional transport of pollutant plays an import role in regional air pollution, and is influenced by the atmospheric circulation, regional meteorology, terrain and distribution of emission sources (An *et al.*, 2007; Wang *et al.*, 2010). Previous study revealed that both anthropogenic and biogenic secondary organic aerosol in Beijing were dominated by regional transport (Lin *et al.*, 2016). Similar phenomenon was found in other cities in the world, such as Paris, in which the transport of $PM_{2.5}$ reached 87% (Skyllakou *et al.*, 2014). Regional

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transport of pollutant makes air pollution prevention and control challenging. Studying the regional transport of PM can help us to understand the formation of severe regional haze events and effectively implement regional pollution prevention and control.

Beijing, as the capital of China, is one of the most important metropolitan cities in the world. Severe air pollution poses a threat to the sustainable development of Beijing. With special terrain, meteorology and the distribution of emission source, severe regional air pollution occurred frequently in Beijing and surrounding areas. Table 1 lists the trans-boundary contribution ratio (TBCR) of PM in Beijing from previous studies. Air quality numerical modelling was commonly used to investigate regional pollutant transport. Most of the TBCR of PM in Beijing ranged from 30% to 40%. As meteorological conditions vary, the TBCR of $PM_{2.5}$ in Beijing has obvious seasonal variations, with the largest TBCR found in summer, followed by spring, autumn and winter (Lang *et al.*, 2013). Although previous studies on regional transport in Beijing have been conducted, some issues, such as the relation of transport to meteorology and secondary aerosols, are still unclear to some extent. Based on the modelling of Chinese Unified Atmospheric Chemistry Environment (CUACE) model, the TBCR of $PM_{2.5}$ and PM_{10} (particulate matter with aerodynamic diameter less than 10 µm) in Beijing and its characteristics were studied via emission source sensitivity method in this paper. The potential source regions and clustered paths were investigated based on the Flexible Particle dispersion model (FLEXPART).

METHODS

Developed by the China Meteorological Administration, The CUACE model is a unified chemical weather numerical forecasting system. Considering the mixing scheme, clear-sky processes, dry deposition, below-cloud scavenging and in-cloud processes, the aerosol module in CUACE includes sulfates, soil dust, black carbon, organic carbon, sea salts, nitrates and ammonium, which were divided into 12 bins with a diameter ranging from 0.01 to 40.96 µm (He *et al.*, 2016b). It is used to evaluate the contribution of regional transport to ambient pollutant concentrations in Beijing through emission source sensitivity test. A more detailed description of CUACE can be found in Gong and Zhang (2008). And it has been evaluated and applied systematically in previous studies (Wang *et al.*, 2010; Wang *et al.*, 2015b; He *et al.*, 2016b). The FLEXPART model, developed by the Norwegian Institute of Air Research, is a Lagrangian transport and dispersion model that is suitable for the simulation of atmospheric transport processes. The potential source regions and clustered transport paths were identified by a FLEXPART backward run. The meteorological fields for CUACE and FLEXPART were supplied by the fifth-generation Penn State/NCAR mesoscale model (MM5).

Three nested domains were used for MM5-CUACE to reduce spurious inner domain boundary effects with a horizontal resolution of 27 km, 9 km, and 3 km (Fig. 1). In the vertical direction, there are a total of 35 full eta levels extending to the model top at 10 hPa, with 16 levels below 2 km. The simulation periods of July (summer) and December (winter) of 2013 were selected to investigate the seasonal impacts of the TBCR. Fig. 2 shows the annual average PM emission in D02. Many emission sources are distributed in the region around Beijing, acting as a significant source of regional pollution over North China. The comparison of the CUACE emission inventory to other inventories, and the details of the integration scheme,

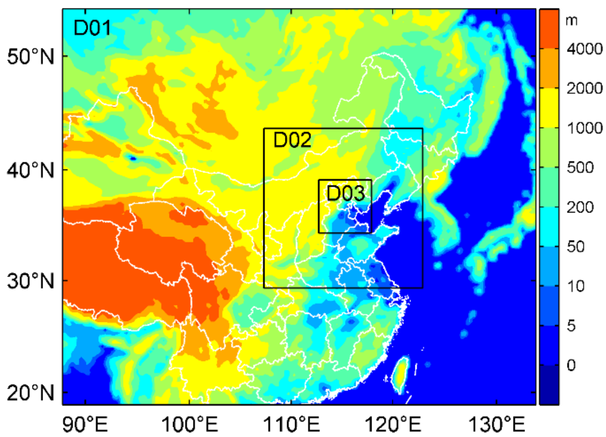

Fig. 1. The three nested domains for simulation with horizontal resolutions of 27 km, 9 km and 3 km. The colour bar represents altitude, the white line represents the administrative boundaries of province.

Table 1. The trans-boundary contribution ratio of PM in Beijing from previous studies.

Source	Period	Method	TBCR
An *et al.* (2007)	Heavy pollution episode in Beijing during 3–7 Apr. 2005	CMAQ simulation using emission switch on/off method	$PM_{2.5}$: 39%
Chen *et al.* (2007)	Four typical months of 2002	CMAQ simulation using emission switch on/off method	PM_{10}: 34.7%
Streets *et al.* (2007)	July 2001	CMAQ simulation using emission switch on/off method	$PM_{2.5}$: 34%
Wu *et al.* (2011)	Aug. 2006	NAQPMS simulation using tagged method	PM_{10}: 25%
Lang *et al.* (2013)	Four typical months of 2010	CMAQ simulation using emission switch on/off method	$PM_{2.5}$: 42.2%
Wang *et al.* (2015c)	2005–2010	HYSPLIT simulation based on PSFC method	$PM_{2.5}$: 35.5%
Zhang *et al.* (2015b)	2013–2015	GEOS-Chem adjoint method	$PM_{2.5}$: 47.2%

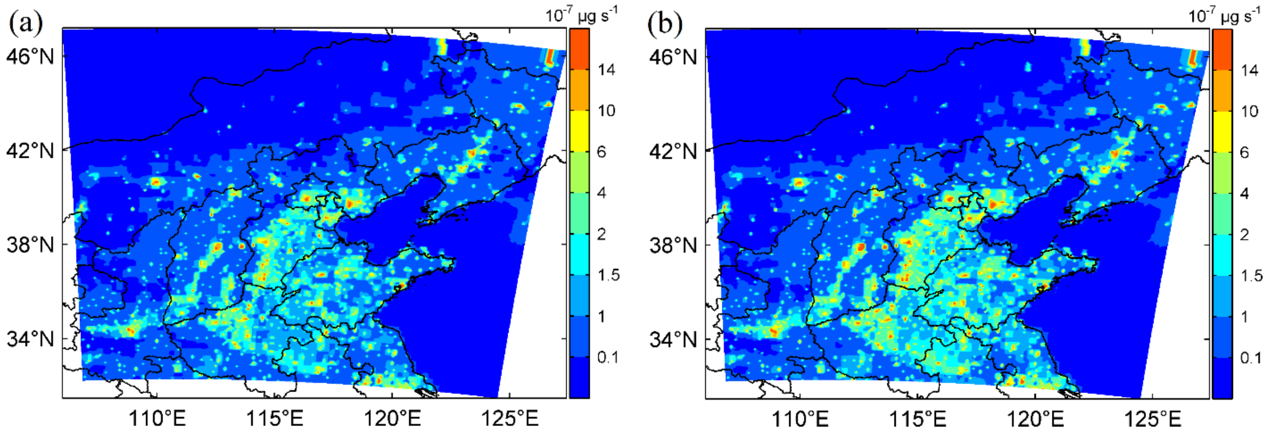

Fig. 2. The annual average $PM_{2.5}$ (a) and PM_{10} (b) emission in D02 (see Fig. 1).

initial and boundary conditions were presented in He *et al.* (2016b). The base run (hereafter refer to as BR) is simulated based on the CUACE default emission inventory with the replacement of vehicle emissions by high temporal-spatial resolution vehicle emissions (Jing *et al.*, 2016). Because of the nonlinear characteristics of air pollution, four sensitivity runs, i.e., switching of local emissions in Beijing (SR1), switching of the emissions outside Beijing (SR2), switching of the emissions of gas (SR3), and switching of the emissions of particle matter (SR4) are conducted for different purposes. As the primary and secondary concentrations of PM cannot be acquired directly in CUACE simulation, the concentrations of PM from SR3 and SR4 represent primary and secondary concentrations of PM. The impact of initial condition on chemical process of aerosol diminishes after relative long spin up (He *et al.*, 2016b). Compared with the BR, SR1 and SR2 are used to calculated the TBCR, SR3 and SR4 are used to calculated the contribution rate of secondary aerosol to PM concentration (CR_S), as shown in Eqs. (1) and (2):

$$TBCR = \frac{C_{SR1} + C_{BR} - C_{SR2}}{2C_{BR}} \times 100\% \quad (1)$$

$$CR_S = \frac{C_{SR4} + C_{BR} - C_{SR3}}{2C_{BR}} \times 100\% \quad (2)$$

where C represents the average concentrations of PM in Beijing.

Particle transport and dispersion can be well captured by FLEXPART in complex terrain with the input of hourly meteorological fields (Brioude *et al.*, 2012). Hourly meteorological fields of D02 from MM5 were used to run FLEXPART. The time step of FLEXPART is 180 s. Particle locations were outputted hourly for backward trajectories and footprints analysis with a residence time of 24 h. Twenty-four hour backward trajectories were selected because they sufficiently determined the probable locations of regional emission sources and explained the regional transport pathways for Beijing (Wang *et al.*, 2010). Meanwhile, turbulence parameterization was switched on for footprints and off for trajectories analysis. The receptor point is located in the centre of the urban region at an altitude of 2 m above the ground. The transport pathways were identified based on a cluster analysis of 24-h three-dimensional backward trajectories. The TBCRs of seven clustered backward trajectories were analysed. As most of the anthropogenic emission sources are located below 100 m, the contribution of air mass sources below 100 m (CR_{air}), combined with the pollutants emission inventory (E_{PM}), was used to calculate the potential source regions of PM (PS_{PM}), as shown in Eq. (3):

$$PS_{PM} = CR_{air} \times E_{PM} \times 100\% \quad (3)$$

With the same model set as this paper, the performance of MM5 and CUACE has been evaluated in previous study (He *et al.*, 2016b). In generally, MM5 and CUACE model can well capture the temporal and spatial distribution of meteorological fields and pollutant concentration respectively. And the evaluation is not shown again in this paper.

RESULTS AND DISCUSSION

The Characteristics of TBCR

The hourly near-surface TBCR to regional average $PM_{2.5}$ concentration in Beijing is from 13.3% to 82.7%, and 11.4% to 70.4%, with the mean values of 53.4% and 36.1% in summer (July) and winter (December) 2013, respectively. For PM_{10}, the mean TBCR is 51.8% and 35.1%, with a maximum of 80.3% and 67.5% and a minimum of 14.4% and 11.7%, in summer and winter 2013, respectively. The mean TBCR of PM_{10} is slightly lower than the TBCR of $PM_{2.5}$ due to easy long-range transport for fine particulate with relative long lifetime in atmosphere. Based on the observation from China National Environmental Monitoring Centre (http://106.37.208.233: 20035/), the ratio of $PM_{2.5}$ to PM_{10} is 0.84 and 0.78 in July and December 2013, while the simulated ratio is 0.89 and 0.85 respectively. The high ratio of $PM_{2.5}$ to PM_{10} is the reason for relative small difference of TBCR between $PM_{2.5}$ and PM_{10}, while the overestimation of the ratio of $PM_{2.5}$ to PM_{10}, especially in winter, might result in the underestimation of the difference of TBCR between $PM_{2.5}$ and PM_{10}. Compared with previous studies of analysis period before 2010 (Table 1), the TBCR is slightly higher, relating to great efforts of air pollution control measures in recently years which decrease the local emission contribution significantly. With similar analysis period, the TBCR in this paper is the same as Zhang *et al* (2015b) basically. It indicates that the joint prevention and control of pollutant emission are very important for continuous improvement of air quality in Beijing.

Based on one-way analysis of variance, the TBCRs are significant different between summer and winter. The TBCR in summer is significant larger than winter, which is consistent with the findings by Lang *et al* (2013). Seasonal difference of meteorological condition combined with the distribution of emission is responsible for seasonal difference of TBCRs. Fig. 3 shows the wind dependence map of the near-surface TBCRs of $PM_{2.5}$ and PM_{10}. Closely related to the wind direction, regional transport is significant when southerly wind is prevailing near the ground because lots of anthropogenic emission are located in the south of Beijing (Fig. 2). In summer, Beijing is dominated frequently by southerly wind, which is beneficial for pollutant transport from Tianjin and Hebei and results in a large TBCR. Atmospheric ventilation capacity was enhanced with the increase of local wind speed and beneficial for pollutant dispersion (He *et al.*, 2013). However, the transport of pollutant from the outside is significant at the same time in regional pollution regions. As we can see from Fig. 3, wind speed is another factor in determining the TBCR, and a high wind speed is often associated with a large TBCR. Influenced by the poor vegetation cover in Inner Mongolia and surrounding areas, the TBCR of PM_{10} is slightly larger than that of $PM_{2.5}$ in northwest wind. Compared with the TBCR of PM_{10}, a large number of fine particulate emissions from industry and vehicles in the provinces south of Beijing results in a large TBCR of $PM_{2.5}$ in southerly wind. Long-range transport for fine particulate is another reason for the large TBCR of $PM_{2.5}$.

Turbulence mixing in planetary boundary layer affects the TBCR significantly. Using multiple linear regression, the relation between hourly planetary boundary layer height (PBLH) and the near-surface TBCR to PM was analysed. The near-surface TBCRs of $PM_{2.5}$ and PM_{10} are positively correlated with PBLH, with a positive effect of 6 % km^{-1} and 16 % km^{-1} in summer and winter, respectively. Strong turbulence enhances the local contribution in the upper atmosphere and results in a large near-surface TBCR. The TBCR exhibits significant change in vertical direction (Wu *et al.*, 2011). With increasing altitude, the TBCR increases rapidly from 10% to 90% below 3 km, implying that local contributions and regional transport are more significant at the near-surface and upper atmosphere, respectively. The development of the planetary boundary layer and turbulence mixing in daytime restrains the effect of the transport of pollutants at the upper atmosphere, and local contributions

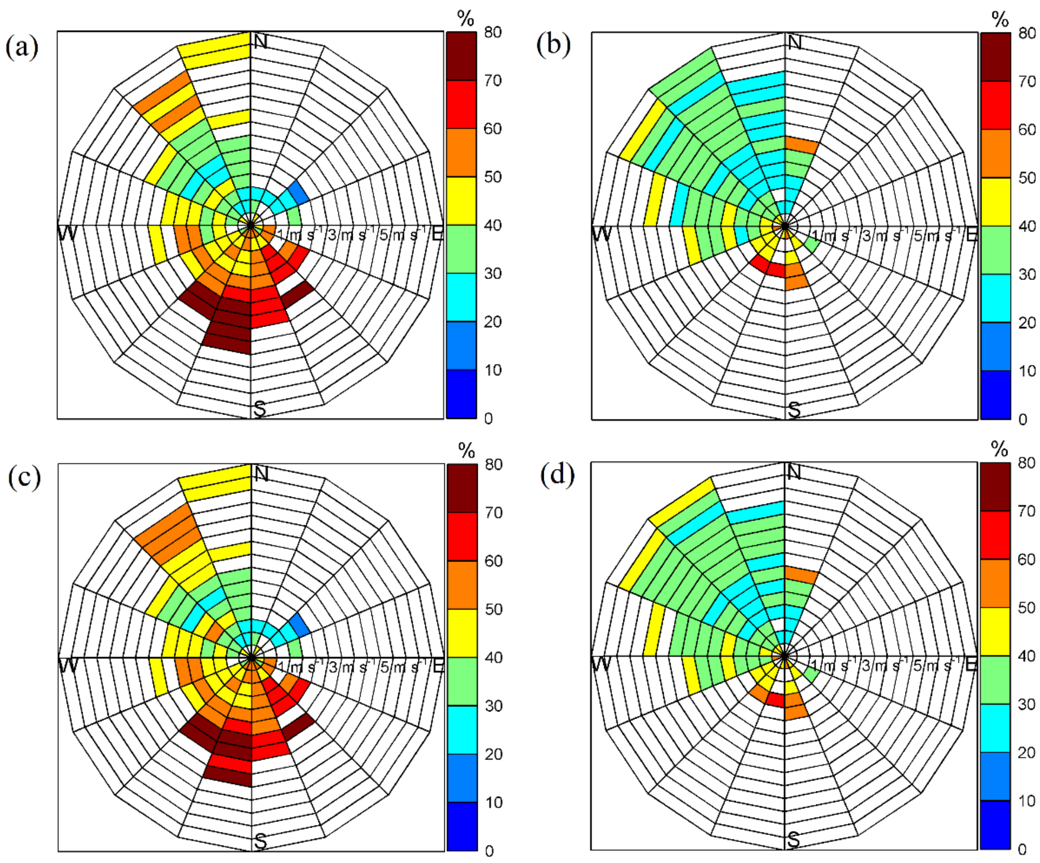

Fig. 3. The wind dependence map of the near-surface TBCR of $PM_{2.5}$ and PM_{10} in July (a, c) and December (b, d) 2013, respectively. The colour bar represents the near-surface TBCR.

can reach higher altitudes. At night, local contributions are limited in the lower atmosphere.

The relation between the TBCR and the CR_S is an important scientific issues. Huang *et al* (2014) investigated secondary aerosol to particulate pollution in four cities in China based on source apportionment method and found the ratio of secondary aerosol in Xi'an (local pollution) is smaller than Beijing, Shanghai, and Guangzhou (regional pollution), which indicates the important of regional transport on secondary aerosol formation. The hourly near-surface CR_S to regional average $PM_{2.5}$ concentration in Beijing is from 9.7% to 65.9%, and 10.0% to 56.5%, with the mean values of 34.8% and 21.5% in summer and winter 2013, respectively. The CR_S of PM_{10} is almost the same as $PM_{2.5}$. The results from CUACE are comparable with previous source apportionment based on observation (Huang *et al.*, 2014). The CR_S in summer is significantly larger significantly than winter. The CR_S is positive correlated with the TBCR significantly. Based on multiple linear regression, CR_S increases 0.5% and 0.27% with the increase of 1% to TBCR in summer and winter 2013, respectively. This indicates that, with large TBCR, aerosol exists a long time and occurs chemical conversion in atmosphere, resulting in large rate of secondary aerosol formation. The secondary aerosol is mostly contributed by regional transport.

Back Trajectories and Potential Source Regions

Possible source regions were frequently investigated by backward trajectory modelling (Zhang *et al.*, 2012; Liu *et al.*, 2013). In this study, the potential source regions of PM to Beijing was identified based on the contribution of air mass sources from the FLEXPART dispersion model and the PM emission inventory of CUACE. Fig. 4 shows the contribution of air mass sources below 100 m and potential source regions of $PM_{2.5}$ in summer and winter 2013. The contribution of air mass sources to receptor is mostly located in the south and north of Beijing in summer and winter respectively. As seasonal difference of atmospheric circulation, potential source regions to receptor point in Beijing have broader source regions in summer, with the south boundary reaching the northern part of Jiangsu and Anhui provinces, the west boundary reaching the eastern part of Shanxi provinces, and the north boundary reaching the southeastern part of Inner Mongolia. The potential source regions are much smaller in winter, with the south boundary reaching the southern part of Hebei province and north boundary reaching the centre of Inner Mongolia. This pattern implies more significant pollutant transport in summer, consistent with the TBCRs for different seasons mentioned in 3.1 and it is similar with Zhang *et al.* (2015). Hebei province is the most important potential source regions to Beijing, followed by Tianjin municipality and Shandong province, which indicates the joint prevention and control of atmospheric pollution is very important for further air quality improvement. A similar pattern exists for the potential source regions of PM_{10}, the characteristics of which are not depicted in this paper.

Based on the K-means clustering algorithm, backward

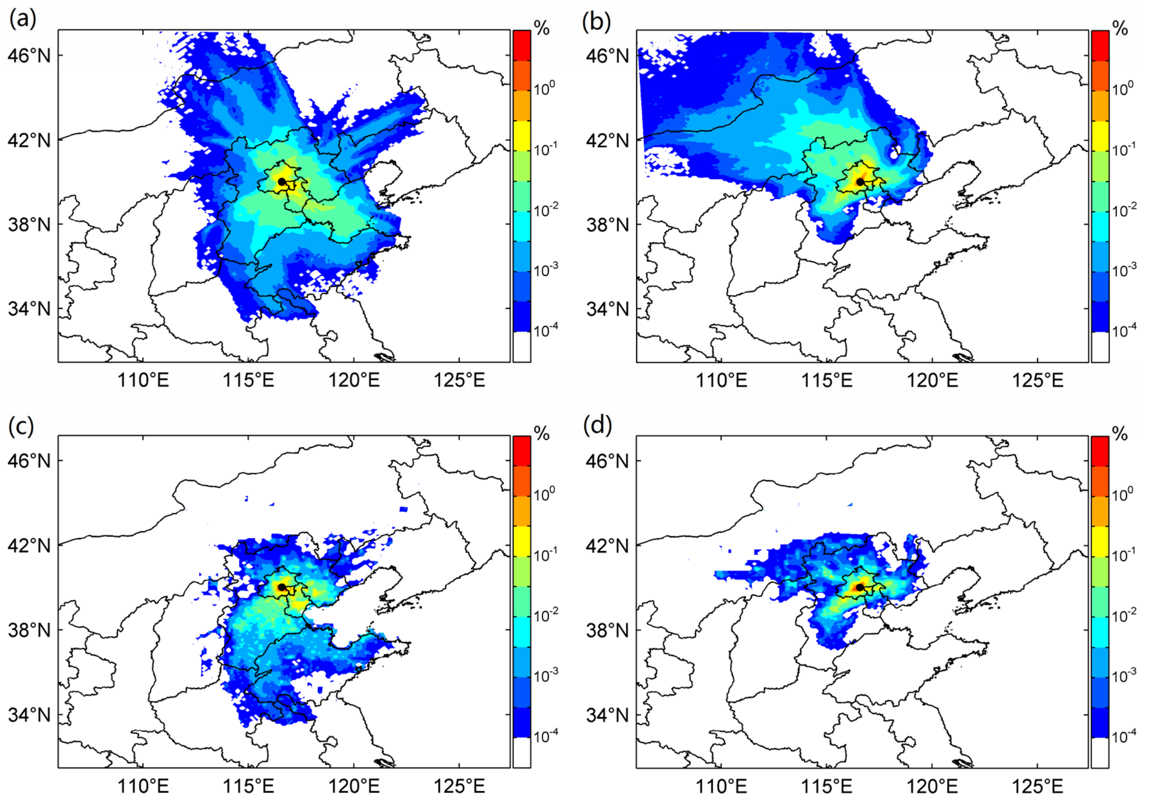

Fig. 4. Contribution of air mass sources below 100 m and potential source regions of $PM_{2.5}$ in July (a, c) and December (b, d) 2013.

trajectories were divided into seven cluster-mean trajectories to identify the relationship between atmospheric transport patterns and hourly PM levels and the TBCR. The number of clusters was dependent on the criterion function (Liu and Gao, 2011). We found that seven clusters can represent the characteristics of transport patterns. Fig. 5 shows the cluster-mean backward trajectories in horizontal and vertical directions. There are significant seasonal differences of transport pathways due to circulation variations. In summer, southeast transport pathways (T1 and T5) are the main transport pathways from the lower atmosphere, with a combined frequency of 38.8% (Table 2). As many emission sources are distributed in southeast areas, severe pollution and significant regional transport occur in the conditions of southeast transport (Fig. 6). The secondary transport pathway is the northeast transport pathway (T7), with a frequency of 17.5% (Table 2). Because less emission sources are distributed in northeast areas (Fig. 2) and because of the short transport distance and high altitude of trajectories, the TBCR is the smallest with relative low pollution levels. The third transport pathways are northwest and northerly transport pathways (T2 and T3), with a combined frequency of 17.2% and the highest altitude of trajectories. Under these conditions, Beijing has the lowest pollution level, with relatively low regional transport. The southerly transport pathway T6, with a frequency of 13.3% and low altitude of trajectories, frequently results in significant regional transport and relatively serious air pollution. The T4 pathway has an obvious directional change from west to southeast, which is not conducive to the long transport and dispersion of pollutants, and it often accompanies relatively serious pollution. In winter, the pathways are mostly from the northwest and north (T1, T3, T4, T5 and T6), with a combined frequency of 70.6%. However, these pathways have significant differences in vertical direction and transport distance. These differences are the main reason for the difference of pollution level and TBCR. For example, T5 is mainly from the upper atmosphere and results in low pollution levels and TBCR at the receptor point, while T1 is from the lower atmosphere and results in a relatively high pollution level and TBCR, although it is very closely to T5 in the horizontal layer. The transport pathway T7 is from west of Beijing with a change of direction. As observed in Fig. 2, there are no significant emission sources in

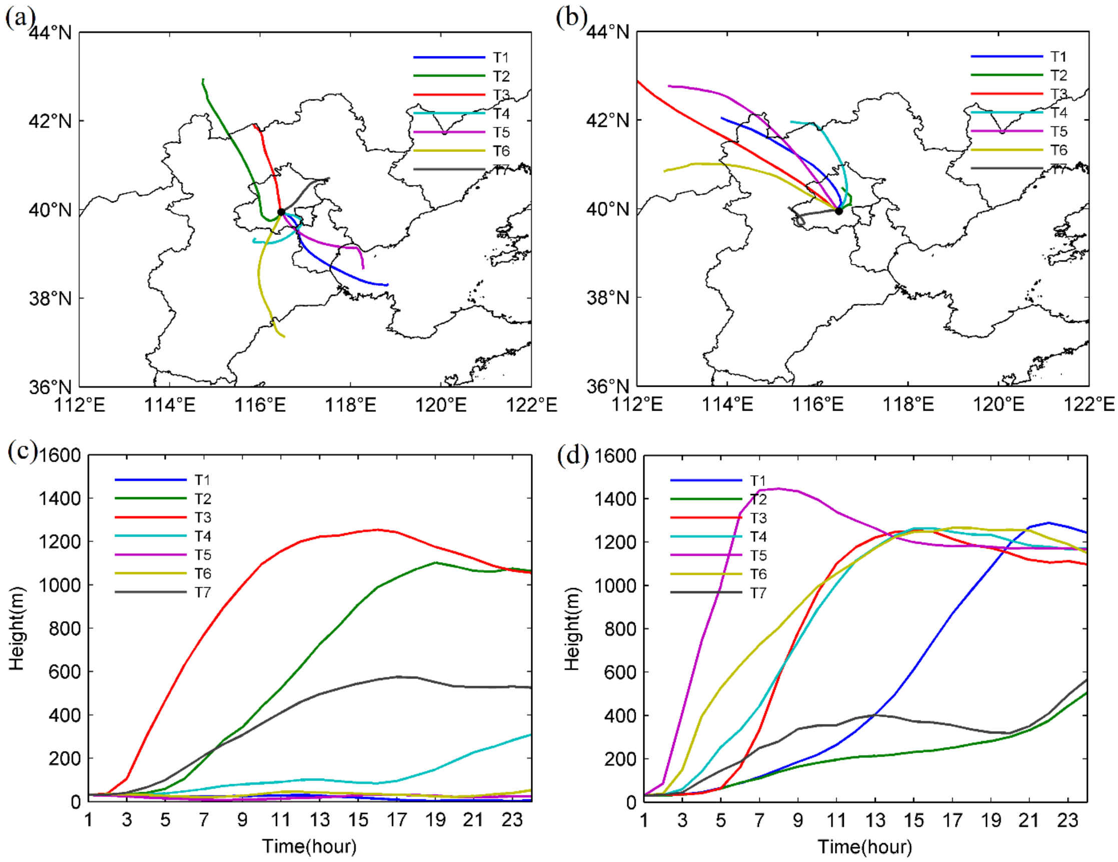

Fig. 5. Cluster-mean backward trajectories in the horizontal and vertical directions in July (a, c) and December (b, d) 2013.

Table 2. The frequency of seven cluster backward trajectories (Unit: %).

	T1	T2	T3	T4	T5	T6	T7
July	19.7	7.0	10.2	13.2	19.1	13.3	17.5
December	16.0	18.6	7.2	22.5	11.9	13.0	10.8

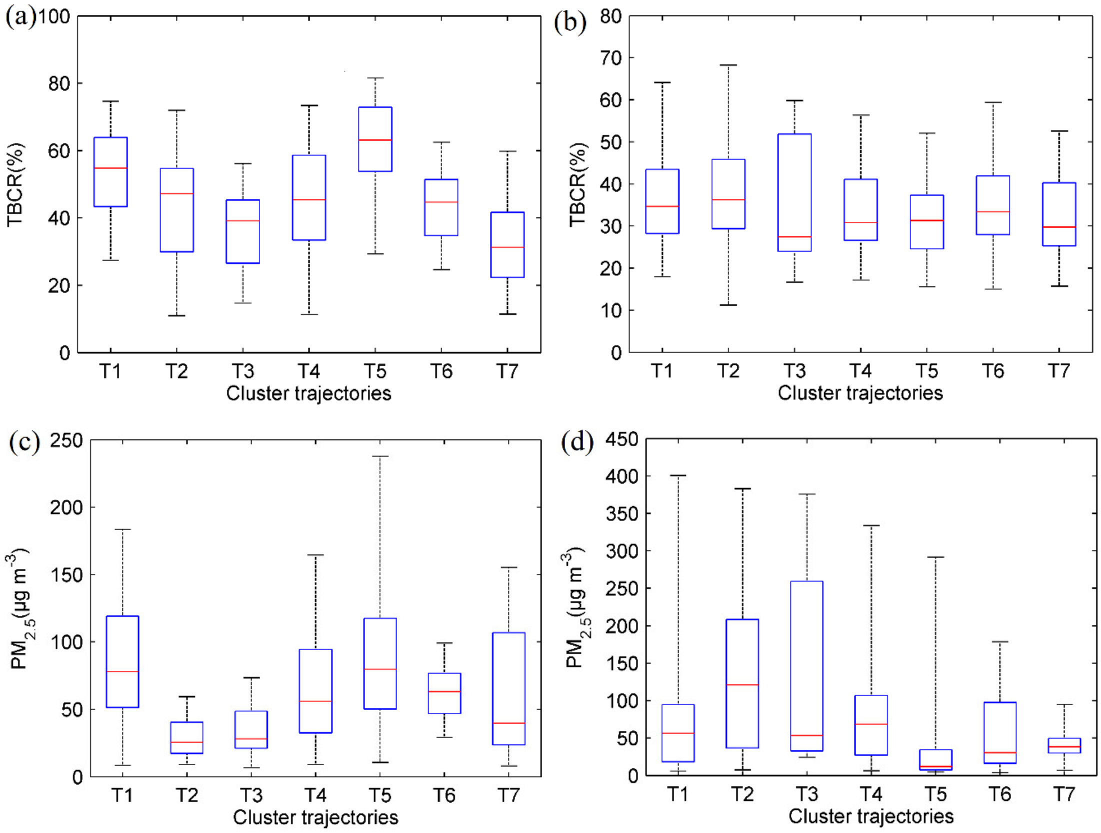

Fig. 6. Box graph of the TBCR and concentration of $PM_{2.5}$ for seven cluster backward trajectories in July (a, c) and December (b, d) 2013. The central mark (red line) is the median, the edges of the box are the 25^{th} and 75^{th} percentiles, and the whiskers extend to the most extreme data**.**

pathway T7, indicating the air mass reaching Beijing is clean air. Meanwhile, low pollution levels and TBCR are detected in this pathway. The transport pathway T2 has the shortest transport distance, suggesting that the weather has static stability. In this situation, severe air pollution occurs in Beijing and the surrounding areas due to the accumulation of pollutants. Despite slow advection, severe pollution in the surrounding areas brings on a large TBCR. Based on the above analysis, there is a significant difference between seven cluster-mean trajectories in summer, while is unapparent in winter. The emission control in Tianjin and Hebei can substantial improve the air pollution in Beijing, especially for summer.

CONCLUSIONS

Regional air pollution frequently occurs in China, indicating the importance of pollutant regional transport effects on pollution levels. Previous studies show that air quality in Beijing is affected by local emissions as well as regional transport. In this study, the TBCR of PM in Beijing, transport pathways and potential source regions of PM were investigated based on simulation results from the CUACE model and FLEXPART backward trajectory model. According to sensitivity analysis conducted by switching on/off emission source, the mean near-surface TBCRs of $PM_{2.5}$ in Beijing were 53.4% and 36.1% in summer and winter 2013 and 51.8% and 35.1% for PM_{10}, respectively. As $PM_{2.5}$ can stay in the atmosphere for a longer period of time and facilitates long distance transport, the TBCR of $PM_{2.5}$ is slightly larger than that of PM_{10}. The high ratio of $PM_{2.5}$ to PM_{10} is the reason for small difference of TBCR between $PM_{2.5}$ and PM_{10}. Regional transport in summer was more significant than that in winter. Further analysis revealed that the turbulence mixing (PBLH) and local atmospheric circulation had significant effects on the TBCR. Closely related to the wind direction and wind speed, regional transport was significant when southerly wind or a high wind speed were prevailing near the ground. The TBCR increases 6% and 16% with the increase of 1 km to PBLH in summer and winter 2013, respectively. The CR_S is closely related to the TBCR. Large TBCR is benefit for chemical conversion and formation of aerosol, resulting in large CR_S. The potential source regions of PM depended on circulation, which is more widespread in summer than winter. Hebei province is the most important potential source regions to Beijing, followed by Tianjin municipality and Shandong province. The air mass source occurred from different directions with a significant difference in cluster-mean trajectories in summer, and occurred from northwest with unapparent differences in winter. The pollution level and regional transport were closely related to the transport pathways and distance, especially in summer.

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