AIR POLLUTION (H ZHANG AND Y SUN, SECTION EDITORS)



Impacts of Regional Transport on Particulate Matter Pollution in China: a Review of Methods and Results

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Published online: 5 August 2017

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Abstract China has been suffering serious particulate matter (PM) pollution in recent decades. Local emission and regional transport both contribute to PM pollution. Determining the contributions of local emissions vs. regional transport to PM concentrations is crucial in making effective PM control policies. This paper reviews the recent research on the contributions of regional transport to PM pollution in four regions in China, i.e., the northern China, eastern China, southern China, and the western China, respectively. The major findings include (1) HYSPLIT is the most popular methods in studies in all regions and often is used in combination with the CA, PSCF, and CWT techniques to investigate the transport pathways and source origins; (2) during the relatively polluted period, transport contributes over 50% of the PM concentrations in Beijing, Shanghai, Hangzhou, Guangzhou, Hong Kong, and Chengdu. Regional transport is important for PM pollution in major cities of China; and (3) regional transport exhibits clear seasonal variations and long term trends. The findings have important implication for emission control programs in these regions.

This article is part of the Topical Collection on Air Pollution

Electronic supplementary material The online version of this article (doi:10.1007/s40726-017-0065-5) contains supplementary material, which is available to authorized users.

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Keywords Particulate matter · Regional transport · Pathways · Source areas · Trajectory · China

Introduction

Particulate matter (PM) has been recognized as a major atmospheric pollutant that can cause various health problems, such as such as respiratory diseases [20, 80], cardiovascular diseases [16, 33, 55], lung cancer [56, 63], low birth weight, and birth defects [34-36]. In addition, enhanced concentrations of PM can absorb and scatter light and causes visibility impairment [11, 12, 25, 47]. China has been experiencing serious PM pollution in recent decades due to rapid economic growth, industrialization, and urbanization. Many observational studies have revealed that annual concentrations of both PM₁₀ and PM₂₅ have been at high levels in major cities in China [1, 21, 25, 58, 77, 104]. Health effect studies have estimated that over one million premature deaths can be attributed to PM_{2.5} pollution in China [37, 49]. Visibility studies have suggested that visibility has been continuously worsening associated with PM pollution during the last few decades [2, 10, 25].

To design effective PM control programs, it is necessary to understand the sources of PM. PM can be directly emitted into the atmosphere (called primary PM, such as black carbon, and dust) and can be formed through atmospheric chemical processes (called secondary PM, such as sulfate, nitrate, and secondary organic aerosols) [60]. PM concentrations measured at receptor sites in a city are a combination of contribution of local emissions and transported PM from outside the city. Determining the contributions of local emissions vs. transport to PM concentrations is crucial in making effective PM control policies.



Recognizing the important role of transport in PM pollution, recent studies have been conducted in various cities in China using several different methods. However, systematic understanding the impact of transport on PM pollution in various regions in China is lacking. The objective of this review is to summarize recent research (mostly after 2010) on the contributions of regional transport to PM pollution in China. We focus on publications about regional transport of PM in China in international peer-reviewed journals. In total, we review 77 publications, as shown in Table S1 in the supplemental materials. We first review the methods that have been used in these studies, and then we summarize the major findings in each of the four regions. We summarize the results into four regions in China, i.e., the eastern (including Shanghai, Jiangsu, Zhejiang, Fujian, Shandong, Anhui, Jiangxi, Hubei), southern (including Guangdong, Guangxi, Hunan, Hainan, Hong Kong), western (including Chongqing, Sichuan, Guizhou, Yunnan, Tibet, Xinjiang, Qinghai, Gansu, Ningxia, Shaanxi), and northern China (including Beijing, Tianjin, Hebei, Henan, Shanxi, Inner Mongolia, Heilongjiang, Liaoning, Jilin). We find 22, 16, 10, and 21 studies in cities/subregions in the eastern[13, 14, 17–19, 24, 30, 39, 40, 59, 61, 64, 66–69, 71, 84, 86, 95, 97, 99], southern [3, 8, 9, 15, 26, 27, 44, 47, 50–52, 54, 62, 74, 81, 90], western [6, 7, 41, 46, 48, 75, 83, 93, 94, 100], and northern China [4, 5, 23, 29, 31, 32, 38, 42, 43, 45, 57, 65, 70, 76, 82, 89, 92, 96, 98, 102, 103], respectively, and we find 8 studies that cover either the entire China or multiple regions of China [22, 28, 53, 73, 79, 85, 91, 101].

This review is organized as follows: in "Methods for PM transport studies in China," we review the methods that have been used for PM transport studies in China; in "Results of PM transport studies in China," we summarize the major findings in the 77 studies; we finally conclude our review and discuss the future research in "Conclusions and implications."

Methods for PM Transport Studies in China

Atmospheric dispersion models are widely used to study the transport and diffusion of pollutants in the atmosphere. There are two general methods to develop the models: Lagrangian method and Eulerian method. Lagrangian method tracks the position and properties of an air parcel according to the mean wind field data along the path that air parcel travels (called "trajectory"). The air parcels may be followed either forward (called "forward trajectories") or backward (called "backward trajectories") in time [88]. In contrary, Eulerian method solves the atmospheric properties (such as concentration of pollutants) over time based on the conservation of mass in "fixed" points (not following the trajectory) [87]. Lagrangian models are ideal for single point sources and they are computationally efficient, but they are not efficient for multiple sources and do not consider complex chemistry processes. Eulerian models solve over the

entire domain and are ideal for multiple sources and also easily handle complex atmospheric chemistry (i.e., Eulerian chemical transport models), but Eulerian models require more computational sources which sometimes have problems with artificial numerical diffusion. In addition, uncertainties associated with the emission estimates would cause bias in the model predictions. In the total 77 studies, 52 studies used the Lagrangian trajectory model, the Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) model, three studies used the Lagrangian dispersion models FLEXPART, and 25 studies used the Eulerian chemical transport models (there are three studies used both HYSPLT and Eulerian chemical transport models).

Lagrangian Trajectory Model

HYSPLIT model was developed by the Air Resources Laboratory (ARL) of the United States National Oceanic and Atmospheric Administration (NOAA). Backward trajectories are commonly calculated to investigate the air mass origins and pathways that reach the receptor sites at a given time using National Centers for Environmental Prediction (NCEP) reanalysis data. Seventy-two- or 48-h backward trajectories are typically used in most of the studies, although 24-[23] and 120-h [45] backward trajectory analysis is also used in a few studies. The altitude of the receptor site is set in the range of 50 to 1000 m above ground level in the studies. Two hundred meters is more often used to consider the ground level PM concentrations. Zhu et al. [102] investigate the effects of the receptor heights on the results and found no significant difference in the results with 100, 200, 300, 500, and 1000 m as the receptor height. After the backward trajectories being calculated, several techniques are often used to further determine the transport pathways and source origins:

(1) Cluster analysis (CA)

CA is a multivariate statistical analysis technique to divide the trajectory data into distinct transport groups or clusters. The GIS-based software TrajStat [78] is commonly used for CA. There are two clustering methods in TrajStat: angle distance and Euclidean distance. Angle distance is based on the direction from which the air masses that reach the site are originated while Euclidean distance is based on the transport positions along the pathways.

(2) Potential source contribution function (PSCF) method

The PSCF method is to identify the possible source areas of the pollutants reaching the receptor site. The entire geographic region covered by the trajectories is divided into an array of grids (defined by the cell indices *i* and *j*). The PSCF value of each grid cell is calculated using the following equation:



$$PSCF_{i,j} = \frac{S_{i,j}}{T_{i,j}} \tag{1}$$

where $S_{i,j}$ represents the number of trajectory endpoints for which the measured pollutant concentration exceeds a threshold value selected for this pollutant in the (i,j) grid cell, and $T_{i,j}$ represents the total number of trajectory endpoints that fall in the grid cell. Therefore, grids with higher PSCF values indicate that they are more likely to be the sources of the pollutant concentrations measured at the receptor sites.

(3) Concentration weighted trajectory (CWT) method

The CWT method is to assign each grid cell a residencetime weighted concentration by averaging the measured concentrations with their associated trajectories that cross the grid cell. The CWT value is calculated using the following equation:

$$CWT_{i,j} = \frac{\sum_{k=1}^{m} C_k \tau_{i,j,k}}{\sum_{k=1}^{m} \tau_{i,j,k}}$$
 (2)

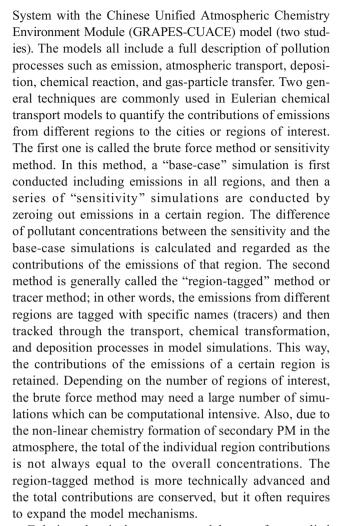
where $CWT_{i,j}$ is the CWT value in the (i,j) grid cell, k is the index of the trajectory, m is the total number of trajectories, C_k is the pollutant concentration observed on arrival of trajectory k, and $\tau_{i,j,k}$ is the time spent in the (i,j)grid by trajectory k. Grids with higher CWT values imply that they are more associated with high concentration measured at the receptor.

Lagrangian Dispersion Models

Three studies utilized the three-dimensional Lagrangian dispersion model, FLEXPART [4, 92], to determine the origin and transport pathways of the air mass arriving in Beijing. FLEXPART simulates the transport and dispersion of tracers by calculating the trajectories of a large amount of particles (air parcels), which are termed plume (cloud) trajectories. Both studies use the Weather Research and Forecasting model (WRF) to provide the meteorological fields to drive the FLEXPART model calculation. Using the calculated backward trajectories, the source of the air mass arriving at the receptor sites can be directly estimated from the locations of their parcels.

Eulerian Chemical Transport Models

Three-dimensional Eulerian chemical transport models were used in 25 studies to investigate the regional transport of PM in China. The models include the Community Multi-scale Air Quality (CMAQ) model (11 studies), the Comprehensive Air Quality Model with Extension (CAMx) (eight studies), the Weather Research and Forecasting model coupled with Chemistry (WRF/Chem) (two studies), the Nested Air Quality Predicting Modeling System (NAQPMS) (two studies), and the Global/Regional Assimilation and PrEdiction



Eulerian chemical transport models are often applied with 3- to 12-km horizontal grid resolutions in a regional scale, although Jiang et al. [29] used 25-km resolution in their study over the north China, and Hu et al. [22] and Ying et al. [85] used 36-km resolution in their studies that covered the entire China. The Fifth Generation Penn State/NCAR Mesoscale Model (MM5) or WRF model simulations are conducted to provide the meteorological fields that are required by the Eulerian chemical transport models.

Figure 1 shows the statistics of the methods used in the studies in different regions. HYSPLIT trajectory model is the most common methods used in the transport studies in all the four regions of China. FLEXPART Lagrangian dispersion model is only applied in the studies in northern China. Eulerian chemical transport model is the second most used method, especially in southern China, but in general, it still only accounts for a small fraction of the studies. Figure 2 shows the techniques used in the HSPLIT studies. CA, PSCF, or CWT methods have been largely used in the HYSPLIT studies to investigate the transport pathways and source origins.



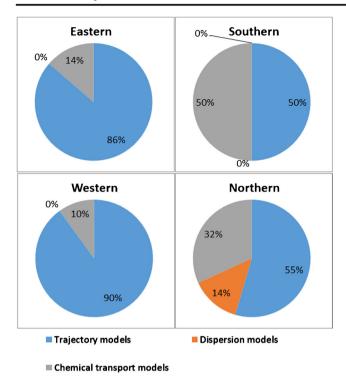


Fig. 1 Relative fractions of different methods used in transport studies in different regions of China

Results of PM Transport Studies in China

Northern China

Most of the studies in the northern China are in the North China Plain (NCP, including Beijing, Tianjin, and Hebei province). Studies indicate that northwest winds in spring have contributed to the high spring PM_{10} concentrations in Beijing [102]. Northerly winds in this region in spring would have a diluting effect on pollutants in winter, while southerly winds would bring in pollutants that have accumulated during transport. During the polluted period, surface $PM_{2.5}$ mainly

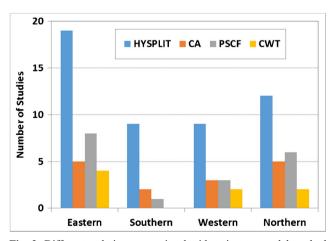


Fig. 2 Different techniques associated with trajectory model method used in the transport studies in different regions of China

originated from sources inside this region (48–72%). In the entire NCP area, southern Hebei represented the largest internal contribution (33%), while the main external contributions came from Shandong (SD) (10%) and Henan (HN) (4%) [5].

Beijing is the core area of the NCP region, and most of the studies conducted for the NCP region focused on Beijing. Studies with the CMAO model reveal that the contribution of surrounding emissions accounted for 67.3 and 61.0% for the first and second PM_{2.5} pollution peaks, respectively [89]. Studies with trajectory analysis reveal that southerly transport is associated with high PM pollution in Beijing [4, 29, 102]. The source areas leading to high PM in Beijing are in Hebei and Shandong, located in south to Beijing [22, 29, 45]. The transport from the near Beijing areas such as Baoding and Hengshui becomes more important during heavy PM pollution events [38]. The northerly transport is often associated with strong winds and leads to better air quality in Beijing, except during the dust storm events. In terms of the regional source contributions, studies show that transport of non-Beijing emissions are very important for PM concentrations in Beijing [82]. The transport contributes over about 40% on an annual basis [32] and contributes over 60% under the southerly and westerly air flows [70]. The contribution of transport during high PM pollution episodes is even larger. The contribution of southerly transport is also confirmed by Wang et al. [76]. In their study, they found that after the control measures were implemented, PM, sulfates, and nitrates were significantly reduced when the northerly air masses prevailed. When the polluted southerly air masses prevailed, the local source control measures in Beijing did not effectively reduce the ambient sulfate concentration due to the enormous regional contribution from the North China Plain.

The transport contributions to PM in Beijing in different seasons were determined in these studies, and seasonal variations in the transport contributions can be concluded. Beijing is affected by trajectories from the south and southeast in summer and autumn. In winter and spring, Beijing was affected not only by the trajectories from the south and southeast but also by trajectories from the north and northwest [38]. The regional transport contributes over 60% of PM_{2.5} in Beijing in summer [82] and 55% in winter during extreme haze pollution events [29]. From the seasonal analysis of PSCF calculation, high PSCF values are found over Hebei province in spring and autumn and over Jiangsu province and Yellow Sea in summer. In winter, local emissions become more important [70]. The results of trajectory clustering and the PSCF method demonstrated that regional sources could be crucial contributors to PM pollution in Beijing. The predominant clusters are N (30%) and S (44%) in spring, S (73%) in summer, NW (50%) in autumn, and NW (88%) in winter [70]. Changes in the regional transport over years are also observed. The main source areas of PM₁₀ have changed from the northwest to the south and southeast of Beijing during 2009–2012. The trend



of transported contribution percentages increased from 2005 to 2010 with a linear rate of 1.2/year [70]. Regional contributions of $PM_{2.5}$ in Beijing during 2013 were 46, 62, 52, and 39% in spring, summer, autumn, and winter, respectively [17].

Eastern China

The Yangtze River Delta (YRD) region is the main focus of the studies in the eastern China. When particle pollution increases, the effects of long-range regional transport significantly increase. The inner regional effects from Jiangsu and the outer regional effects from the upwind adjacent province, Shandong, are crucial for the YRD region. [18]. Emissions from the YRD contributed to over 70% of the O₃ precursor CO, with a majority from the YRD and the North China Plain which are the main contributors to PM_{2.5} pollution in this region, especially for the burning episode days [13]. During dust storm events, this dust storm which broke out in Xinjiang and Mongolia arrived in the YRD region after 2–4 days and the transported dust particles contributed to the mean surface layer concentrations of PM₁₀ in the YRD region 78.9% [19].

Most of the studies for YRD region have been taken place in the city of Shanghai. More than 60% of PM pollution events in Shanghai can attribute to region transport [84]. The air quality in Shanghai was mostly influenced by the air masses from north, east, and west directions. The air masses from north directions accounted for 44.8% of all the air masses, followed by those from east directions (30.4%) and those from west directions (24.8%) [68]. Two studies revealed that long-range transport of dust can cause serious PM pollution in spring, and the transport pathways pass though the areas such as Hebei and Shandong with high anthropogenic emissions also contributed to high PM levels in Shanghai [17, 64]. The major possible source areas for Shanghai include Hebei, Shandong, Anhui, and Jiangsu to Shanghai. With the northerly air flow, regional transports can lead to high concentrations of PM in Shanghai, and long-range transport from the northwestern China including the Inner Mongol had effects in winter and autumn [18].

In Shanghai, regional contributions over four seasons were 36, 39, 45, and 35% in spring, summer, autumn, and winter, respectively [17]. The northwest and the north paths were the major air mass transport paths in December, among which 79.6% of the total trajectories came from mainland while 20.4% reached Shanghai through the ocean [17]. The most potential source areas are located in YRD and the Circum-Bohai-Sea Region near Shanghai in winter, Yellow sandstorm and southwestern regions in spring, the southwestern coastal regions in Zhejiang, Fujian and PRD region in summer, and northern regions in China in autumn.

Regional transport is also important for other cities in the eastern of China, such as Nanjing and Hangzhou. In Nanjing, the possible source regions are mainly located in areas to the

northeast of the city. High aerosol pollution is mainly contributed by regional transport associated with northeastern air masses [61]. The high biomass-burning pollutant concentrations are linked to the air masses from the western (summer harvest) and southern (autumn harvest) areas [61]. The transport of pollutants from Hebei, Henan, and Hubei probably contributed to the haze pollution in Jiangsu. The sources affecting the extremely high PM_{2.5} in Hangzhou are mainly located in southeastern coast of Zhejiang and Fujian provinces, north part of Jiangxi, and central part of Jiangsu province [86].

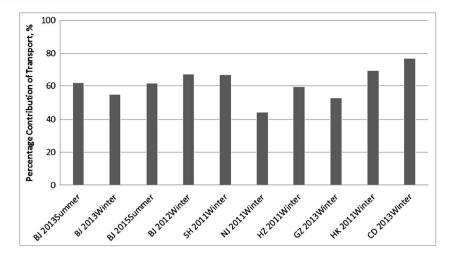
Southern of China

The key region of research in the southern China is the Pearl River Deal (PRD) region. Super-regional transport was an important contributor for both sulfates and nitrates in all ten cities in PRD region in both winter and summer. The average super-regional contribution of sulfate and nitrate reached up to 80 and 56%, respectively, in winter [50]. The source apportionment result suggests that point source and super regional source are the ones that contribute the pollutants most in the rain water over PRD region [50]. The Hong Kong/PRD region's super-regional transport and mobile vehicles are the two major fine particle sources, contributing 62 and 21% of the total figure in December and 42 and 28% in April [15].

The studies in the PRD region are mostly in Guangzhou and Hong Kong. Regional transport also plays an important role in the two cities. In wet season, Guangzhou was mainly affected by the emissions from the south, including PRD region and the ocean areas. Regional transport contributes 11.6%. But in dry season, the pollutants transported from the areas north of PRD region both played important roles contributing 52.9% [9]. The trans-boundary PM₁₀ transport contributes nearly half of PM₁₀ on average [8]. Three surrounding cities (Foshan, Dongguan, and Huizhou) had a total emission contribution of nearly 30% to Guangzhou's PM₁₀ concentration. The emissions from these three cities also accounted for 70 to 94% of the total trans-boundary contributions from Guangzhou's nine surrounding cities. In Hong Kong, an overall increasing trend is observed in ambient PM₁₀ sulfate and nitrate concentration. The local sources and ship emissions in the South China Sea are not the cause of observed overall increase trends. Evident increases in PM₁₀ sulfate and nitrate levels are observed in air masses originating from eastern China [54]. Source analysis showed that the increased mass concentrations on high PM days were mainly caused by air pollutant transport from the outside-Hong Kong regions [54]. Monthly averaged regional contribution to Hong Kong is 69.4% in December and 53.8% in April [54]. Monthly averaged super-regional contribution to Hong Kong is 69.4% in December and 53.8% in April, indicating the high correlation between air quality in Hong Kong and the emissions in Mainland China [81].



Fig. 3 The percentage contribution of transport to PM_{2.5} concentrations in various cities/ episodes. The cities are *BJ* Beijing, *SH* Shanghai, *NJ* Nanjing, *HZ* Hangzhou, *GZ* Guangzhou, *HK* Hong Kong, *CD* Chengdu



Western China

Studies in the western China are relatively less, compared to the three regions discussed above. There are some studies in the Sichuan Basin (SCB) and the Tibetan Plateau. The transport pathways and potential sources of PM_{2.5} have been investigated during the extreme haze episode in winter 2013 in SCB. This study revealed that over 77% of the air mass approaching Chengdu during winter of 2013 came from inside the SCB. The main potential sources of PM_{2.5} were located in southeast cities and the western margin of the SCB [46]. The dust events in Chengdu were mostly influenced by desert regions in western and northern China, different from dust transport in northern and eastern China. The Qinghai-Tibet Plateau prevented the direct entrainment of dust from the west, and a similar northeastward turning of dust air was observed before arriving in Chengdu [46]. Xin [83] suggested that in Tibetan Plateau uplift area, in spring and winter, eastern Xinjiang, border areas between Gansu and Inner Mongolia, and southern Tibet were the dominant potential sources. The study also revealed that Tibet in China was one of the potential sources of PM₁₀ in Xining [83]. During the monsoon period, pollutants from SCB and eastern Yunnan province can be transported to Lijiang (southeast to Tibetan Plateau) with high loading of sulfate, ammonium, and some heavy metal elements [93]. During the dust events, the model simulations showed that the Tibetan dust aerosols appeared at approximately 7-8 km above sea level, and the plumes originated from the nearby Taklimakan Desert and accumulated over the northern slope often Tibet Plateau during the summer [72].

Transport Contributions in Major Cities

Figure 3 shows percentage contributions of transport to PM concentrations in Beijing, Shanghai, Nanjing, Hangzhou, Guangzhou, Hong Kong, and Chengdu. Multiple studies have been conducted in Beijing in different years and seasons. The

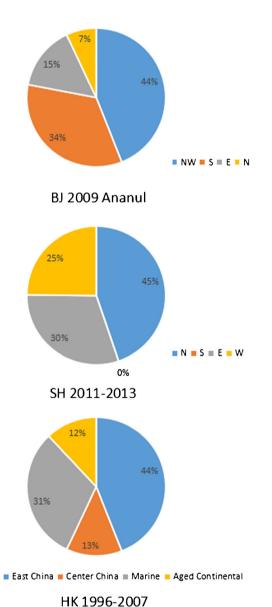


Fig. 4 The transport directions in Beijing (BJ), Shanghai (SH), and Hong Kong (HK)



results indicate that transport contributes 43 (Nanjing) to 78% (Chengdu). Transport contributes 55 (2013 winter) to 65% (2012 winter) to the PM concentrations in Beijing. The contributions of transport are also over 50% in Shanghai, Hangzhou, Guangzhou, and Hong Kong. Contributions over 60% are found mainly in winter episodes when PM pollution is more serious.

Figure 4 shows the transport trajectories from different directions or regions summarized from the studies for Beijing, Shanghai, and Hong Kong for over 1 year or longer time. The prevailing transport directions are northwest (44%) and south (34%) in Beijing and north (45%) and east (30%) in Shanghai. In Hong Kong, 44% transport is from east China, and 31% is from the ocean. The prevailing transport direction, combined with high pollutant emissions in the upwind regions (such as Hebei and Shandong located in the south to Beijing, Shandong and Jiangsu located in the north to Shanghai), causes high PM pollutions in these cities.

Conclusions and Implications

A few conclusions can be drawn from the review of the recent research on the regional transport of PM in four regions of China. Also, the results have important implications for emission control in these regions.

- (1) Both Lagrangian and Eulerian models have been used in studies in China. HYSPLIT is the most popular methods in studies in all regions and often is used in combination with the CA, PSCF, and CWT techniques to investigate the transport pathways and source origins
- (2) Studies indicate that transport contributes over 50% of the PM concentrations in Beijing, Shanghai, Hangzhou, Guangzhou, Hong Kong, and Chengdu. Regional transport is important for PM pollution in major cities of China. Therefore, to effectively control the PM pollutions in these regions, regional or inter-regional emission control plans are recommended.
- (3) Regional transport pathways are quite different among different regions. The prevailing transport directions are northwest and southern Beijing and north and eastern Shanghai. Southerly transport from Hebei and Shandong leads to high PM pollution in Beijing, northerly and northwesterly transport from YRD, central and northern China is important for Shanghai, and northerly transport from eastern China is import for PRD, while most of the PM in Chengdu is transported from inside the SCB during haze pollution events. This implies that lowering the emissions in the northern and central China not only will benefit the PM pollution there but also will improve air quality in eastern and southern China.

To better understand the role of regional transport in the formation of severe PM pollution and to design effective emission control programs in China, we suggest that more studies should be considered in the future.

- (1) Although HYSPLIT analysis provides valuable information about the transport pathways and source origins, quantitative contributions of transport to the concentrations at the receptor cannot be determined. More studies using the Lagrangian dispersion models or Eulerian chemical transport models should be conducted. Some PM components, such as sulfate, nitrate, ammonium, and secondary organic aerosols, account for a large fraction of total PM mass and they are formed through chemical processes during the transport pathways. Chemical transport models are needed to simulate the processes.
- (2) Most of the current studies focus on a few hotspot regions/cities, such as Beijing, Shanghai, Guangzhou, and Hong Kong. Studies in other regions are limited, especially in SCB and the Guanzhong Plain, which also experience severe PM pollution.
- (3) Studies in Beijing have revealed that transport changed over years with the change of climate and the change of emissions in upwind areas. Considering the fast development of economy and society in other regions of China, the similar change is expected. Therefore, longterm studies should be conducted in other cities/regions to investigate the seasonal variations and long-term trends of the regional transport.

Acknowledgements The authors would like to thank the support from the National Natural Science Foundation of China under contract no. 41675125, Natural Science Foundation of Jiangsu Province under contract no. BK20150904, and the Priority Academic Program Development of Jiangsu Higher Education Institutions (PAPD), Jiangsu Key Laboratory of Atmospheric Environment Monitoring and Pollution Control of Nanjing University of Information Science and Technology, and Jiangsu Province Innovation Platform for Superiority Subject of Environmental Science and Engineering (no. KHK1201).

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