Characterizations of PM$_{2.5}$ Pollution Pathways and Sources Analysis in Four Large Cities in China

Baolei Lv$^1$, Yu Liu$^2$, Peng Yu$^3$, Bin Zhang$^1$, Yuqi Bai$^1$*

$^1$ Ministry of Education Key Laboratory for Earth System Modeling, Center for Earth System Science, Tsinghua University, Beijing, 100084, China
$^2$ Department of Electronic Engineering, Tsinghua University, Beijing, 100084, China
$^3$ Department of Hydraulic Engineering, Tsinghua University, Beijing, 100084, China

ABSTRACT

Particulate matter with an aerodynamic diameter of 2.5 micrometers or less (PM$_{2.5}$) is a primary pollutant in most cities in China. PM$_{2.5}$ poses a significant human health risk, especially in the most densely populated urban areas. We used observations of PM$_{2.5}$ and backward air mass trajectories modeled by HYSPLIT-4. We characterize how air movement patterns influence pollution levels in four large cities of China. Then we developed a method to evaluate regional and local sources and contributions of PM$_{2.5}$. For Beijing and Shanghai, PM$_{2.5}$ concentrations are sensitive to air moving direction, indicating significant influence of air movement on PM$_{2.5}$ pollution. In Beijing, PM$_{2.5}$ concentrations were higher when the air masses were from the south and the east. In Shanghai, pollution was greater with northerly air mass flows. Regional contributions of PM$_{2.5}$ in Beijing during 2013 were 46, 62, 52, and 39% in spring, summer, autumn and winter, respectively. In Shanghai, regional contributions over four seasons were 36, 39, 45, and 35%. In Guangzhou and Chengdu, PM$_{2.5}$ concentrations were more sensitive to speed rather than direction of air mass movements, indicating weaker pollution pathways. In Guangzhou, regional contributions were smaller over the four seasons: 15, 28, 16, and 22% while in Chengdu, they are 21, 52, 28, and 14%. These results are comparable to previous results obtained using complex atmospheric chemical transport models.

Keywords: PM$_{2.5}$; Large city; China; Pollution pathway; Source analysis.

INTRODUCTION

Particulate matter with an aerodynamic diameter of 2.5 µm (PM$_{2.5}$) is the primary pollutant in most cities in China. These particles can readily penetrate into human lungs and bronchi (Nel, 2005; Pope III et al., 2011). Epidemiological studies show that long-term exposure to PM$_{2.5}$ increases morbidity and mortality (Pope III et al., 2011). Due to its complex composition and sources, PM$_{2.5}$ can have a variety of adverse effects on climate, weather, and human health (Eatough et al., 2006; Zhao et al., 2011; Tao et al., 2014).

Rapid economic development and urbanization has led to large city areas with high population densities. Beijing-Tianjin-Hebei (BTH), Yangtze River Delta (YRD), and Pearl River Delta (PRD) are three major city clusters in China. In 2010 their populations were 104, 35, and 64 million and the total accounted for 15% of the China population in only 4.7% of the area. These regions are economically well-developed and BTH, YRD, and PRD contributed GDPs of 11%, 17%, and 13% respectively to the Chinese economy in 2013. Dense populations and thriving economies have massive power requirements and consume large amounts of goods and services. Social and economic activities consume significant quantities of fossil fuels. Therefore, air in large cities is always polluted by emissions from coal and oil combustion. Huang et al. (2011a) estimated that, in 2007, anthropogenic emissions of pollutants such as NO$_x$, CO, SO$_2$, PM$_{10}$, and PM$_{2.5}$ were 2.29, 6.70, 2.39, 3.12, and 1.51 Tg respectively in the YRD. Air pollution is severe in these regions (Wang et al., 2012; Zhao et al., 2013; Chen et al., 2014) and it can cause severe adverse effects on human health to quite dense populations in large cities.

Transport pathway analysis of particulate matter is often done using backward trajectory clustering and analysis (Broge et al., 2007; Ji et al., 2012). Trajectory analysis is also commonly used to analyze pollution episodes (Ji et al., 2012; Ji et al., 2014) and to trace their potential sources using a potential source concentration function (PSCF; Pongkiatkul and Kim, 2007; Zhang et al., 2013a). In China, pathway analysis using long-term particulate matter data has
been carried out in Beijing (Wang et al., 2004; Zhu et al., 2011) and Shanghai (Li et al., 2012). However, these analyses used PM$_{10}$ rather than PM$_{2.5}$ data. Given the large difference in their size, potential sources of PM$_{2.5}$ also require consideration. In addition, particulate matter transport analysis typically focuses on analyzing the difference of PM$_{2.5}$ pollution levels among different trajectory clusters (Zhu et al., 2011; Wang et al., 2015) while intensive, exploratory and quantitative studies are generally lacking.

Quantitative assessment of trans-boundary transport of PM$_{2.5}$ is important for implementing targeted emission control measures. To achieve this, atmospheric chemistry transport models provide important tools for calculating local pollutant contributions by turning off emissions in neighboring regions (Chen et al., 2007; Cheng et al., 2007; Streets et al., 2007; Wang et al., 2008; Cheng et al., 2013; Lang et al., 2013). This method is effective only in the context of accurate emission inventories, reliable weather simulations, and comprehensive description of chemical reactions. However, there are still many uncertainties in these aspects. Hourly PM$_{2.5}$ concentrations are routinely monitored in nationwide monitoring stations. But these data are typically underexploited and only used to describe and understand real-time pollution levels. Normally these data are not used for source analysis of ambient particulate matter. Based on transport pathway identification and analysis, we developed a novel method for using these data to conduct source analysis in large cities.

In this study, we investigated sensitivities of PM$_{2.5}$ pollution levels to air pathways. Based on a newly proposed method, we quantified the trans-boundary contributions to PM$_{2.5}$ in four large Chinese cities.

**DATA AND METHODS**

**Data Source**

Hourly concentrations of PM$_{2.5}$ in Beijing (BJ), Shanghai (SH), Guangzhou (GZ) and Chengdu (CD) (black stars in Fig. 1) were recorded by the US Embassy (consulates) in 2013. The January data in BJ and CD are missing. The PM$_{2.5}$ monitors are included in the AirNow-International (AirNow-I) system (www.airnow.gov/index.cfm?action=ani.main), which is same to the U.S. Environmental Protection Agency’s (EPA) real-time air quality data management and display system known as AirNow. AirNow-I includes a suite of software for data processing, quality control methods. Furthermore, the data set shows good agreement with data from the China National Environmental Monitoring Center that uses the TEOM method with guaranteed data calibration and quality control. The US Embassy data has been used in published studies (Zhang et al. (2013b), Wang et al. (2013a)). Unlike previous studies, we used hourly PM$_{2.5}$ observations rather than the daily mean. In Chinese cities, diurnal variations are often significant (Ji et al., 2014), so
hourly PM$_{2.5}$ concentrations are more representative and informative.

**Backward Trajectory Modeling and Analysis**

Air parcel trajectories are paths of infinitesimally small particles of air mass (Wang et al., 2010). Air parcels can carry particulate and gaseous pollutants to remote regions via weather systems. Air mass trajectories are a convenient and effective method for evaluating pollutant transportation pathways. In this study, a 24-hour air mass backward trajectory analysis was calculated using the National Oceanic and Atmospheric Administration (NOAA) Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT-4) model (http://www.arl.noaa.gov/ready/open/hysplit4.html). This model is used to calculate dispersion and air mass trajectories (Wang et al., 2004; Zhang et al., 2013a). The archived meteorological data was obtained from the NCEP’s Global Data Assimilation System (GDAS). Trajectory clustering was used to group similar trajectories in three dimensions and to find those which behaved similarly (Lee et al., 2007). Trajectory calculating, clustering, and statistics were conducted using TrajStat software. TrajStat software uses the K-means method to cluster trajectories (Wang et al., 2009). The goal of clustering is to generate results that are distinctive and representative. In our study, we tested various numbers in each city and the proper number of clusters was determined by evaluating changes of mean PM$_{2.5}$ concentrations in the clusters (Wang et al., 2004). The experience-based visual judgement method was also used to help determine the optimal number of clusters (Wang et al., 2010).

To investigate pollution transport pathways in the four cities, we modeled 24-hour backward trajectories of the air parcels arriving at BJ (39.95°N, 116.47°E), SH (31.21°N, 121.44°E), GZ (23.12°N, 113.32°E) and CD (30.63°N, 104.07°E) in 2013. The arrival time was hourly from 00:00 (midnight) to 23:00 (11:00 pm). The arrival height was 200 m above the ground level (A.G.L.). The height of 200 m was used by previous studies and no significant differences were found among the modeled trajectories of different receptor heights below 1000 m in Beijing and Shanghai (Zhu et al., 2011; Li et al., 2012). To better understand pollution transport pathways in each season, both annual and seasonal trajectories were clustered, as illustrated in Fig. 2.

**A Novel Method for Evaluating Regional and Local Contributions**

Urban particulate pollution is usually attributed to both regional and local sources. Pollution levels are closely related to transport pathways used by the air masses. When air masses come from clean pathways, the pollution levels tend to be lower and vice versa. It is reasonable to conclude that particulate pollution will be at a minimum level when incoming air masses move along the cleanest pathways. The lowest PM$_{2.5}$ concentrations among different pathways can be regarded as background pollution levels created only by local sources. The corresponding average PM$_{2.5}$ concentrations therefore represent background pollution levels (Man et al., 2001; Wang et al., 2015). Differences between average PM$_{2.5}$ concentrations of each cluster and the lowest concentrations result from regionally transported pollutant contributions. This conclusion is based on the assumption that local meteorological conditions (relative humidity, wind speed, etc.) are similar under different pathways. To minimize the bias caused by discrepancies to this assumption, we employed a procedure of pathway identification rather than using the original clusters. Based on the resulting analysis we developed a method to quickly calculate regional and local contributions. The method can be implemented as follows:

(1) **Pathway Identification**

Correct characterization of pathway direction needs to be made to investigate variations of pollutant transport among the different pathways (Wang et al., 2015). This was achieved by merging the trajectories of different clusters if these clusters were directionally close. To merge the trajectories, corresponding PM$_{2.5}$ concentrations were the weighted average of the numbers of the trajectories in each cluster, as shown in Eq. (1). For example, trajectory merging was used in BJ and GZ. In BJ, northwesterly clusters were merged to represent the entire northwest pathway. In GZ, the clusters that coincided in a similar direction were also merged (Table 2).

**Weighted average PM$_{2.5}$ concentrations (LP)**

\[
LP = \frac{\sum (N_i \times C_i)}{\sum N_i}
\]  

where $N_i$ are the number of trajectories in cluster $i$ and $C_i$ is the corresponding average PM$_{2.5}$ concentration in cluster $i$.

(2) **Local Contribution Calculation**

\[
Local \ contribution \ (LC) = \frac{Local \ Concentration \ (LC)}{C_{avg}}
\]  

where $C_{avg}$ is the annual or seasonal average PM$_{2.5}$ concentration. $C_{min}$ is the lowest concentration among all the pathways in a season. $LC$ refers to local induced part of PM$_{2.5}$ concentration ($\mu$g m$^{-3}$) and.

(3) **Regional Contribution Calculation**

\[
Regional \ contribution \ (P_i) = \frac{N_i \times (C_i - C_{min})}{\sum_{i=1}^{m} [N_i \times (C_i - C_{min})]} \times (1 - LP)
\]

where $m$ and $N_i$ are the number of pathways and number of trajectories in pathway $i$ and $C_i$ is the concentration of pathway $i$. LP (\%) is the percentage that $LC$ constitutes. $P_i$ is the fraction of regional transport through pathway $i$. 

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RESULTS AND DISCUSSION

Transport Pathways and Potential Sources

Mean PM$_{2.5}$ mass concentrations recorded by the US embassy in 2013 were 92.9, 59.7, 56.1, and 87.2 µg m$^{-3}$ in BJ, SH, GZ, and CD respectively. Data from the air quality monitoring system gave readings of 89.5, 62, 53, and 86.3 µg m$^{-3}$. The results of two data sets show excellent agreement.

At the average-linking cluster stage, we investigated the linkage of four to nine clusters. By visual inspection, the number of clusters chosen best represented the classifications of air mass trajectories in 2013, as shown in Fig. 2 and Table 1. PM$_{2.5}$ concentrations are very sensitive to air mass movements, which were revealed by the trajectories in different clusters. In BJ, the highest PM$_{2.5}$ concentrations are usually found in the southeastern and southwestern clusters, namely #5, 3, 5, and 4 in clusters of spring, summer, autumn, and winter of 2013 as shown in Fig. 2. Anthropogenic emissions are significant in the area to these directions (Zhao et al., 2012) and strong regional transport from Hebei, Shandong, and other locations which are documented in previous studies (Pu et al., 2015). The lowest PM$_{2.5}$ concentrations were from the northwestern clusters, due to rapidly moving clean air masses from that direction. In this scheme, air flows travel from the north and over forests and grasslands, as the land cover types show in the Fig. 1. The ratio of the highest PM$_{2.5}$ concentrations to the lowest (H/L) in spring, summer, autumn and winter were respectively 3.1, 3.88, 8.05, and 9.03. The higher ratios indicate stronger regional pollutant transport during autumn and winter.

In Fig. 2(A), the trajectories within clusters 3 and 5 have

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$^{[1]}$ The index of each cluster as shown in Fig. 2; $^{[2]}$ Number of trajectories in a cluster; $^{[3]}$ Average concentration of PM$_{2.5}$ in a cluster.

Fig. 2. Annual (A) and seasonal ((B): spring, (C): summer, (D): autumn, (E): winter) every-hour 24-hour backward trajectories (yellow lines) and their clusters (blue lines) in four megacities in 2013.
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Lv et al., Aerosol and Air Quality Research, x: 1–8, xxxx 5

similar lengths but different directions. Higher average PM$_{2.5}$ mass concentrations are found in cluster 3, which is more prone to western influence. This is also the case for clusters 2 and 4 in Fig. 2(D) and clusters 1 and 3 in Fig. 2(E). This indicates contributions from long-distance transport of terrestrial particles and industrial aerosols (Zhang et al., 2012), from the Gobi desert and intensive coal industries located in the western part of Inner Mongolia (Wang et al., 2004; Zhang et al., 2010). The trajectories within cluster 2 and 5 (Fig. 2(A)) are from similar directions but differ in length. Higher PM$_{2.5}$ pollution levels exist in the cluster with the longer trajectories. This is true for clusters 3 and 4 in autumn (Fig. 2(D)) and clusters 2 and 3 in winter (Fig. 2(E)). Faster clean air flow from the northeast can effectively accelerate the diffusion of local emissions of PM$_{2.5}$.

In SH, high PM$_{2.5}$ concentrations were found in northerly inland clusters, namely #3, 2, 3, and 1 over the four seasons. The highest PM$_{2.5}$ concentrations were closely associated with strong emissions from the North China Plain, where there is intensive industry and heavily populated cities (Li et al., 2011). In this instance, the mean PM$_{2.5}$ concentration was 145.8µg m$^{-3}$ in winter. The lowest concentrations usually corresponded to clusters from the East China Sea, which are #5, 4, 2, and 4 in four seasons (Table 1). Marine air masses are relatively clean and can carry moisture leading to precipitation. The H/L ratios over the four seasons were 2.52, 2.14, 3.69, and 2.24 and the ratios indicate that regional transport was not as great as that in BJ.

In GZ, the lowest PM$_{2.5}$ concentrations were associated with the clusters with longer air mass trajectories. These clusters were #2, 4, and 4 over four seasons (Fig. 2). In the other aspect, the higher PM$_{2.5}$ concentrations are found in clusters with faster air flow moving speeds. In GZ, marine air masses are relatively clean and they are associate with the lowest mean PM$_{2.5}$ concentration. The ratios of H/L in spring, summer autumn and winter were 1.31, 2.12, 1.53, and 1.88 and these small ratios indicate that local emissions within PRD play a major role for PM$_{2.5}$ pollutions in GZ.

In CD, higher PM$_{2.5}$ concentrations were found in clusters of shorter trajectories as in Fig. 2(A). As in GZ, PM$_{2.5}$ concentrations exhibited less variation among different clusters. The ratios of H/L were 1.42, 2.13, 1.56, and 1.84 and they are much smaller than in the other cities. This indicates minor contributions from regional transport to PM$_{2.5}$ pollution levels.

Analysis of Local and Regional Source Contributions

One advantage of our method is that contributions of regional transport through specific transport pathways could also be examined as shown in Table 2. In BJ, PM$_{2.5}$ concentrations caused by local emissions were higher in winter and this is reasonable considering the intensive coal combustion in winter. But for the contributions from local emissions, they were higher in spring and summer. Considering the wide spread biomass and coal burning in Hebei and Shandong in autumn and winter, stronger regional transport would be concluded in the two season. Generally, regional transport of pollutants is significant in BJ (Xu et al., 2011; Zhang et al., 2012; Zhao et al., 2013) and our results

Table 1. Estimated local and regional contributions to PM$_{2.5}$ in four mega-cities in 2013. The numbers in the brackets are the cluster ids included in the corresponding pathways.

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Analysis of Local and Regional Source Contributions

One advantage of our method is that contributions of regional transport through specific transport pathways could also be examined as shown in Table 2. In BJ, PM$_{2.5}$ concentrations caused by local emissions were higher in winter and this is reasonable considering the intensive coal combustion in winter. But for the contributions from local emissions, they were higher in spring and summer. Considering the wide spread biomass and coal burning in Hebei and Shandong in autumn and winter, stronger regional transport would be concluded in the two season. Generally, regional transport of pollutants is significant in BJ (Xu et al., 2011; Zhang et al., 2012; Zhao et al., 2013) and our results
show that they are 46, 62, 52, and 39% in four seasons, and comparable to other studies (Table 3). Regional contributions to PM$_{2.5}$ pollution in BJ are strongest from the east and south directions and emissions from these directions contribute 36, 38, 46, and 27% to the observed PM$_{2.5}$ concentrations respectively in the four seasons. In SH, local contributions remain constant at approximately 60% in the four seasons and regional contributions are most significant from inland areas, especially North China by inspecting the starting points of inland trajectories. In GZ, significant local contributions, approximately 80% year round, indicate that air contamination is mainly caused by emissions within PRD. Studies have shown that in GZ, most carbon-based matter in PM$_{2.5}$ (Huang et al., 2012), secondary products (Zhang et al., 2008), and particulate matter (Lai et al., 2007; Huang et al., 2011b) are of local origin. For example, our estimate of PM$_{2.5}$ at 84% in autumn is similar to the 82% value (PM$_{10}$, 2006) of Cheng et al. (2013). Locally derived PM$_{2.5}$ pollution dominates in CD except during summer and this is probably because there is more rain in summer which would weaken PM$_{2.5}$ local formation. Li et al. (2013) concluded that local emissions constituted about 77% of PM$_{2.5}$ pollution in October of 2012 a result similar to the 72% autumn estimate in our study.

**CONCLUSIONS**

For the first time, hourly PM$_{2.5}$ concentration data in four large Chinese cities over a complete year (2013) were used to conduct backward trajectories using a HYSPLIT-4 Model to locate air pollution transport pathways. We investigated variations in pollution levels among trajectory clusters. We also employed a novel method using observation data to calculate local and regional contributions. The method was easy to use and proved to be effective. In BJ, high PM$_{2.5}$ concentrations are found in the southwest and southeast pathways, directions from which heavy industrial cities are located. In SH, inland transport pathways caused higher PM$_{2.5}$ pollution levels and they reached levels as high as 145.8 µg m$^{-3}$ in winter, due to a prevailing north wind and intensive coal combustion in North China. In GZ and CD, there are no significant regional pollution pathways, indicating that pollution is largely due to local emissions. We also found that the H/L ratio was an indicator of the intensity of regional transport.

Both regional and local PM$_{2.5}$ contributions are significant in BJ and SH, and local contributions dominate PM$_{2.5}$ pollution in GZ and CD. In BJ, regional contributions are 46, 62, 52, and 39% in the four seasons, and are comparable to previous studies. Regional transport is mainly through the southern and eastern transport pathways in BJ. In SH, regional contributions are 36%, 39%, 45%, and 35% in the four seasons and they are mostly from inland pathways. In GZ and CD, regional transport is minor and contributes 15, 28, 16, and 22% seasonally in GZ. Regional contributions are 21, 52, 28, and 14% in CD which is consistent with results of other studies.

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**REFERENCE**


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