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

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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 human 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

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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 nation-wide 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

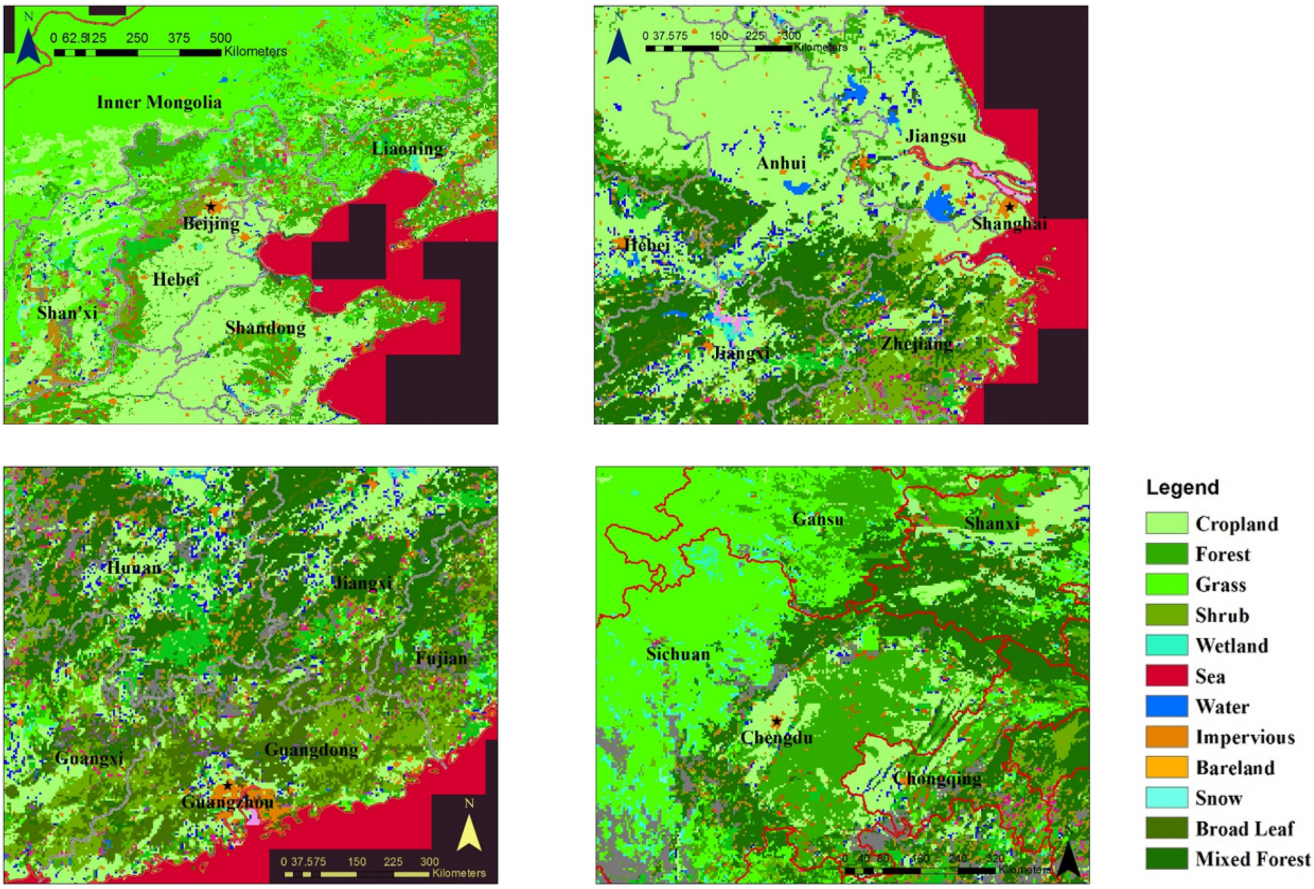

Fig. 1. Locations of four megacities (black stars) and the land covers in the surrounding area.

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).

$$Weighted\ average\ PM_{2.5}\ concentrations\ (LP) = \frac{\sum (N_i \times C_i)}{\sum N_i} \quad (1)$$

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

$$Local\ contribution\ (LP) = \frac{Local\ Concentration\ (LC)}{C_{avg}} \quad (2)$$

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 (μg m^{-3}) and.

(3) Regional Contribution Calculation

$$Regional\ contribution\ (P_i) = \frac{N_i \times (C_i - C_{\min})}{\sum_{i=1}^{m}\left[N_i \times (C_i - C_{\min})\right]} \times (1 - LP) = \frac{N_i \times (C_i - C_{\min})}{(C_{avg} - C_{\min}) \times \sum_{i=1}^{m} N_i} \times (1 - LP) \quad (3)$$

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*.

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

Table 1. Average $PM_{2.5}$ concentration in different backward trajectory analysis (µg m^{-3}).

CITY	ID[1]	ANNUAL		SPRING		SUMMER		AUTUMN		WINTER	
		A[2]	B[3]	A	B	A	B	A	B	A	B
BEIJING	1	3673	127.8	390	47.24	160	31.12	577	115.28	397	80.81
	2	1192	74.51	385	108.66	429	66.79	368	55.14	370	110.08
	3	669	63.6	453	80.92	603	112.66	308	61.85	254	24.96
	4	1156	103.54	292	53.63	183	29	302	19.55	509	225.3
	5	1102	28.89	518	146.63	681	79.37	612	157.49		
	All	7792	96.55	2038	92.51	2056	78.27	2167	96.05	1530	126.69
SHANGHAI	1	1922	48.78	283	39.96	610	44.95	805	36.59	758	145.81
	2	2483	63.96	345	62.25	382	50.61	489	26.48	507	75.1
	3	1009	49.11	390	95.63	556	21.45	397	97.63	449	71.35
	4	1475	35.45	471	57.26	433	20.96	388	49.47	263	65.54
	5	1110	115	455	37.76						
	All	7999	60.26	1944	58.76	1981	34.2	2079	48.27	1977	100.09
GUANGZHOU	1	1677	52.87	480	63.99	538	40.01	586	62.63	793	66.26
	2	2782	57.88	729	51.56	303	22.19	922	55.75	431	107.19
	3	1455	75.37	651	67.47	732	24.34	89	69.83	288	68.56
	4	1037	46.98			170	46.98	134	48.04	399	57.93
	5	558	25.3					240	52.32		
	All	7509	56.22	1860	60.33	1743	31.01	1971	57.49	1911	74.1
CHENGDU	1	1930	92.21	596	95.66	830	63.04	445	63.06	381	135.86
	2	1878	83.34	204	67.63	668	57.7	497	97.94	354	143.25
	3	1481	64.71	342	88.85	275	33.03	362	91.44	293	114.73
	4	1436	105.16	501	79.32	281	57.4	408	97.81	271	137.35
	All	6725	86.44	1643	85.78	2054	56.52	1712	87.47	1299	133.42

[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.

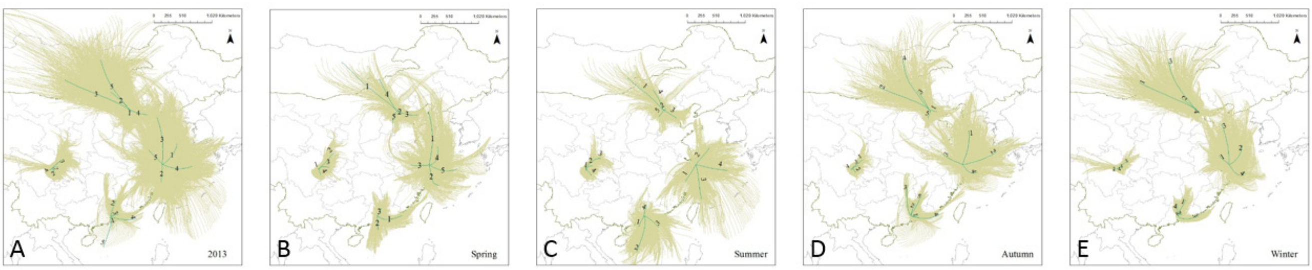

Fig. 2. Annual (A) and seasonal ((B): spring, (C): summer, (D): autumn, (E): winter) every-hour 24-hour backward trajectories (yellow lines) and their clusters (blue lines) in four megacities in 2013.

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.

City	ID	An.	Sp.	Su.	Au.	Wi.	City	An.	Sp.	Su.	Au.	Wi.
Beijing	P_1	0.36(1)	0(1,4)	0(1,4)	0.19(1)	0(1,2,3)	Shanghai	0.05(1)	0.01(1)	0.22(1)	0.08(1)	0.31(1)
	P_2	0(2,4,5)	0.12(3)	0.10(2)	0(2,3,4)	0.39(4)		0.15(2)	0.07(2)	0.17(2)	0(2)	0.02(2)
	P_3	0.07(3)	0.07(4)	0.31(3)	0.33(5)			0.03(3)	0.20(3)	0.00(3)	0.28(3)	0.01(3)
	P_4		0.27(5)	0.21(5)				0(4)	0.08(4)	0(4)	0.09(4)	0(4)
	P_5							0.18(5)	0(5)			
	LP	0.57	0.54	0.38	0.48	0.61		0.59	0.64	0.61	0.55	0.65
	LC	55.08	49.98	29.99	46.26	77.52		35.45	37.76	20.96	26.48	65.54
Guangzhou	P_1	0.054(1)	0.05(1)	0.18(1)	0.08(1)	0.05(1)	Chengdu	0.09(1)	0.12(1)	0.21(1)	0(1)	0.05(1)
	P_2	0.12(2)	0(2)	0(2)	0.07(2,5)	0.15(2)		0.06(2)	0(2)	0.14(2)	0.12(2)	0.06(2)
	P_3	0.12(3)	0.09(3)	0.03(3)	0.02(3)	0.02(3)		0(3)	0.05(3)	0(3)	0.07(3)	0(3)
	P_4	0(4,5)		0.08(4)	0(4)	0(4)		0.10(4)	0.04(4)	0.06(4)	0.09(4)	0.18(4)
	LP	0.7	0.85	0.72	0.83	0.78		0.75	0.79	0.58	0.72	0.86
	LC	39.4	51.56	22.19	48.04	57.93		64.71	67.63	33.03	63.06	114.73

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, 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 3. Comparison of regional contributions of $PM_{2.5}$ in Beijing in different studies.

Season	Our study	Lang *et al.*, 2013	Chen *et al.*, 2007	Streets *et al.*, 2007	Wang *et al.*, 2008	Wang *et al.*, 2013b	Han *et al.*, 2005
	2013	2010	2008	2008	2008	2007	2003
Spring	46%	34.4%	37.9%				59%
Summer	62%	75.2%	40.0%	34%	44%		
Autumn	52%	33.6%	37.4%				17%
Winter	39%	24.9%	23.4%		21%	50%	
Annual	43%	42.2%					

show that they are 46, 62, 52, and 39% in four seasons, and comparable to other studies (Table 3). Regional contributions to $PM_{2.5}$ pollutions 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 $PM_{2.5}$ pollutions 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 n GZ. Regional contributions are 21, 52, 28, and 14% in CD which is consistent with results of other studies.

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