Spatial-temporal characteristics of PM_{2.5} in China: A city-level perspective analysis

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Abstract: Haze pollution has become a severe environmental problem in the daily life of the people in China. PM_{2.5} makes a significant contribution to poor air quality. The spatio-temporal features of China's PM_{2.5} concentrations should be investigated. This paper, based on observed data from 945 newly located monitoring sites in 2014 and industrial working population data obtained from International Standard Industrial Classification (ISIC), reveals the spatio-temporal variations of PM_{2.5} concentrations in China and the correlations among different industries. We tested the spatial autocorrelation of PM_{2.5} concentrations in the cities of China with the spatial autocorrelation model. A correlation coefficient to examine the correlativity of PM_{2.5} concentrations and 23 characteristic variables for 190 cities in China in 2014, from which the most important ones were chosen, and then a regression model was built to further reveal the social and economic factors affecting PM_{2.5} concentrations. Results: (1) The Hu Huanyong Line and the Yangtze River were the E-W divide and S-N divide between high and low values of China. (2) The PM_{2.5} concentrations shows great seasonal variation, which is high in autumn and winter but low in spring and summer. The monthly average shows a U-shaped pattern, and daily average presents a periodic and impulse-shaped change. (3) PM_{2.5} concentrations had a distinct characteristic of spatial agglomeration. The North China Plain was the predominant region of agglomeration, and the southeastern coastal area had stable good air quality.

Keywords: PM_{2.5}; China; spatial-temporal characteristics; monitoring data

1 Introduction

Since 2013, haze pollution has become a severe environmental problem in China and has impacted the health and daily life of inhabitants; this has aroused great attention. Of all the components of air pollution, particulate matter with an aerodynamic diameter $\leq 2.5 \mu\text{m}$ (PM_{2.5}) is most significant and life-threatening. On the one hand, the increase of PM_{2.5} concentration can not only reduce atmospheric visibility (Kan *et al.*, 2012), have negative impact on human health (Krewski *et al.*, 2000), but also cause climate change by changing the

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radiation balance (Pan *et al.*, 2014). On the other hand, PM_{2.5} concentration, affected by the intensity of emission source, reliefs and climatic factors, is featured by spatial-temporal variability (Cao *et al.*, 2007). Therefore, understanding the variations of PM_{2.5} concentrations at the national scale is beneficial to us to assess their impact on human health and the environment, and carry out targeted control measures.

In China, studies on PM_{2.5} have been carried out since 2000, and have mainly focused on the urban areas of the Pearl River Delta (Ho *et al.*, 2014), the Yangtze River Delta (Li *et al.*, 2011), and the Beijing-Tianjin-Hebei region (Wang *et al.*, 2016; Zhao *et al.*, 2013); these studies have been similar to research in other countries (Austin *et al.*, 2013; Boldo *et al.*, 2011; DeGaetano and Doherty, 2004). Most previous research has focused on measuring the chemical composition and spatial-temporal variability of PM_{2.5} in China (Hu *et al.*, 2014; Liu *et al.*, 2009).

Currently, most researchers conduct studies on national PM_{2.5} from satellite based data (Lin *et al.*, 2014). This is undoubtedly the most efficient method of retrieving spatial heterogeneity of surface PM_{2.5} concentrations. Previous researchers have shown that surface PM_{2.5} concentrations can be derived from satellite based aerosol optical thickness (AOT) observation through the use of linear regression (Wang and Christopher, 2003). However, the correlation between AOT and surface PM_{2.5} varies for different seasons and locations (Hoff and Christopher, 2009), and AOT hardly reflects the spatial pattern of surface PM_{2.5} (Paciorek and Liu, 2008). What is more, all this research was carried out at PM_{2.5} observations less than 100 µg/m³, because higher PM_{2.5} concentrations lead to biased and inaccurate predictions (Gupta *et al.*, 2006), which is always the case in Beijing. Besides, missing values frequently occur in AOT data, especially when the weather is cloudy or hazy. Thus, near-ground observation data of PM_{2.5} concentrations are more accurate for further research.

There were 945 sites in 190 cities since the beginning of 2014, which are mainly located in the southeastern coastal, central, and northeastern regions. The increase in the number and extent of monitoring sites provided a foundation for research on the spatial-temporal characteristics of PM_{2.5}. We collected PM_{2.5} data from 945 automatic air quality monitoring sites across China in 2014 to examine the spatial-temporal variations using several methods.

2 Experimental section

2.1 Data collection

This paper takes the mainland of China as the study area, with PM_{2.5} concentration data derived from urban air quality monitoring data of China's National Environmental Monitoring Centre. The observation includes hourly PM_{2.5} concentrations at 945 monitoring sites in 190 cities in 2014 (Figure 1). Of all the stations, 130 cities are distributed in provinces of Shandong (30), Jiangsu (24), Guangdong (21), Zhejiang (15), Liaoning (11), and Hebei (11), together with Beijing (12), Shanghai (10), and Guangzhou (11), which corresponded to three major developed regions, namely, the Bohai Rim, the Yangtze River Delta, and the Pearl River Delta, respectively. A Thermo Fisher 1405F monitor was used to measure PM_{2.5} mass concentration. The principle is that air containing PM_{2.5} particles were inhaled into the equipment (device) at a constant flow rate, with the aid of a sampling cutter, combined with

a filter dynamics measurement system (FDMS) and tapered element oscillating microbalance (TEOM) (Wang *et al.*, 2015b).

Figure 1 Spatial distribution of air quality monitoring sites in megalopolitan regions of China in 2014 (a. Bohai Rim; b. Yangtze River Delta; c. Pearl River Delta)

2.2 Data preparation

According to Ambient Air Quality Standards (on trial) (AAQS) of China GB3095-2012 requirements for the validity of air pollutants concentration data, we conducted PM_{2.5} data quality control. First, we excluded the hourly value less than 0 or greater then 9999 and the missing values of PM_{2.5} concentrations in the raw data; secondly, in calculating the daily mean, if missing data of the current day covers more than four hours, the data on the same day is invalid, and thus was deleted; thirdly, in calculating monthly mean, if the lack of monitoring data of the current month covers less than 27 days, then the data in the same month is invalid, and thus was deleted; fourthly, in calculating annual mean, if the lack of monitoring data of the current year covers less than 324 days, then the data in the same year is invalid, and thus was deleted; finally, we removed some abnormal values of >900 μg/m³ in an hour. Data at all the monitoring sites is in a variety. In this study, the monitored 24-h averaged PM_{2.5} concentrations at all the stations in a city represented the polluted level of the city. Thus, we obtained 354 valid days in 2014 for all the stations across the country (data is missing or invalid on March 29–31, May 25–26, July 14, September 27–29, December 27–28 and was removed). According to the definition of GB3095-2012, herein “daily average” means the arithmetic mean of a natural daily 24-h average concentrations, “monthly average” means the arithmetic average of the mean concentrations of each day in a

calendar month, “seasonal average” means the arithmetic average of the mean concentrations of each day in a calendar quarter, “annual (yearly) average” means the arithmetic average of the mean concentrations of each day in a calendar year; spring refers to March to May, summer covers June to August, autumn refers to September to November, and winter covers January, February and December.

2.3 The spatial autocorrelation model

Tobler (1970) put forward the First Law of Geography that any object is related to other objects with special consideration of distance, therefore, the closer objects are located, the stronger the correlation that exists between them. This is called spatial autocorrelation, which can be measured by Moran’s I (Moran, 1948; Geary, 1954). PM_{2.5} concentrations follow the same rule (Wu *et al.*, 2015). When the observation values are similar within a certain distance (d), the Moran’s I is significantly positive (p value < 0.1), otherwise it is negative. If the observation values are arranged randomly, the Moran’s I is zero. Moran’s I can be classified into Global Moran’s I (GMI) and Local Moran’s I (LMI) (Anselin, 1995). GMI is used to judge the spatial agglomeration degree of China’s PM_{2.5}, and LMI is used to examine the spatial distribution of the “hot spots” and “cold spots.” Due to the possible local spatial autocorrelation observations existing in the overall spatial random sample distribution, GMI and LMI were both used in this paper to analyze the agglomeration features of PM_{2.5} concentrations.

$$I = \frac{n}{S_0} \frac{\sum_{i=1}^n \sum_{j=1}^n w_{ij} (x_i - \bar{x})(x_j - \bar{x})}{\sum_{i=1}^n (x_i - \bar{x})^2} \quad (1)$$

where n is the total number of the sample, x_i and x_j are respectively the PM_{2.5} concentrations in places of i and j . $\bar{x}$ is the average value, w_{ij} is spatial weight matrix ($n \times n$), and S_0 is the sum of all its elements.

$$I_i = \sum w_{i,j} Z_i Z_j \quad (2)$$

where Z_i and Z_j are the standardized values of PM_{2.5} concentrations. When I_i and Z_i are both significantly positive (p value < 0.05), it means that the PM_{2.5} concentrations in place i and the units nearby are high, which is termed a High Concentration Area (HH); by contrast, when I_i and Z_i are negative, which shows that the PM_{2.5} concentrations in place i and its neighboring units are low, this is called a Low Concentration Area (LL). If I_i is positive and Z_i is negative, it means that the PM_{2.5} concentration in place i is higher than those of the neighboring units; this is referred to as a High Low Concentration Area (HL); while if I_i is negative and Z_i is positive, the PM_{2.5} concentrations of place i are lower than those of the units nearby, which are then termed Low High Concentration Areas (LHs).

Moran’s I is a weighted correlation coefficient used to detect departure from spatial randomness; it is used to determine whether the neighboring areas are more similar than expected under the null hypothesis. Moran’s I index ranges from -1 to 1 ; if the locations are not related, the value is expected to be close to 0 ; when I is a negative value, it generally denotes negative autocorrelation; when I is a positive value, it indicates positive autocorre-

lation. Spatial autocorrelation mainly depends on Z value. When Z(I) is positive and significant, it indicates that there is a positive autocorrelation in an area, that is, High-High or Low-Low agglomeration; when Z(I) is negative and significant, there is negative spatial autocorrelation, namely High-Low agglomeration or Low-High agglomeration; when the Z(I) value is 0, the observation values were distributed in an independent random manner. In general, Z is significant when it is greater than 1.96 and it is extremely significant when it is greater than 2.58.

We tested the spatial autocorrelation of PM_{2.5} concentrations in the cities of China with the spatial autocorrelation model. Results showed that, Moran's I was greater than 0 for annual, seasonal, and monthly values in 2014, with April and May close to 0, indicating there was no significant spatial autocorrelation. The remaining months ranged from 0.16 to 0.31, indicating there was spatial autocorrelation (Table 1). The average Z(I) of 12 months was 19.86, which exceeded 2.58, suggesting that there was significant positive spatial autocorrelation of PM_{2.5} concentrations in the cities of China (High-High or Low-Low agglomeration). In the analysis of the Spatial Autocorrelation Model based on ArcGIS10.2, Gi_Bin was used to identify statistically significant hot spots and cold spots. Confidence intervals of +3 to −3 elements reflected a confidence level of 99% statistical significance, a confidence interval of +2 to −2 elements reflected a confidence level of 95% statistical significance, confidence intervals of −1 to +1 elements reflected a confidence level of 90% statistical significance; and a confidence interval of 0 clustering was not statistically significant.

Table 1 Spatial autocorrelation index of PM_{2.5} concentrations in the major cities of China in 2014

Month	Jan.	Feb.	Mar.	Apr.	May	Jun.	Jul.	Aug.	Sept.
Moran's I	0.16	0.30	0.24	0.09	0.05	0.20	0.31	0.26	0.16
Z(I)	15.94	29.85	24.03	10.78	5.59	20.66	31.10	26.11	16.18
Number of HH cities	119	110	123	36	15	126	120	123	70
Month	Oct.	Nov.	Dec.	Spring	Summer	Autumn	Winter	Annual	
Moran's I	0.16	0.26	0.16	0.16	0.27	0.21	0.27	0.28	
Z(I)	16.16	25.94	15.92	16.65	27.25	20.91	27.22	28.32	
Number of HH cities	63	121	124	121	131	118	121	127	

Note: Moran's I was greater than 0 for annual, seasonal, and monthly values in 2014, with April and May close to 0, indicating there was no significant spatial autocorrelation. The remaining months ranged from 0.16 to 0.31, indicating there was spatial autocorrelation. The average Z(I) of 12 months was 19.86, which exceeded 2.58, suggesting that there was significant positive spatial autocorrelation of PM_{2.5} concentrations in the cities of China.

3 Results

3.1 Temporal change

The PM_{2.5} concentrations had a U-shaped pattern of “high in autumn and winter but low in spring and summer” in the major cities of China. The annual average PM_{2.5} concentrations were 61 µg/m³ in the 190 cities monitored in China in 2014. Among the top ten cities in terms of the annual average value, seven were located in Hebei Province and three in Shandong Province. Xingtai, Baoding, Shijiazhuang, Handan, and Hengshui ranked first to fifth, with PM_{2.5} concentrations over 100 µg/m³. Meanwhile, according to AAQS (GB 3095-2012),

there were only 18 cities (9.47%) meeting the annual standard of China ($<35 \mu\text{g}/\text{m}^3$), 136 cities (71.58%) meeting 24-h Air Quality Standard ($<75 \mu\text{g}/\text{m}^3$), and the other 36 cities (18.95%) were below the two grades.

$\text{PM}_{2.5}$ in China presented distinct seasonal variations. Coal combustion in winter led to the seasonal maximum value of $\text{PM}_{2.5}$ concentrations in northern China, with an average of $84 \mu\text{g}/\text{m}^3$. The highest and lowest values were in Shijiazhuang and Lhasa with values of 202 and $29 \mu\text{g}/\text{m}^3$, respectively. Spring ranked second with an average of $59 \mu\text{g}/\text{m}^3$ with the highest value in Korla at $134 \mu\text{g}/\text{m}^3$ and Xingtai at $106 \mu\text{g}/\text{m}^3$. The former was caused by frequent sand storms in spring; the latter was closely related to coal combustion for heating. The lowest value was observed at $6 \mu\text{g}/\text{m}^3$ in cities such as Beihai and Qiqihar. Autumn ranked third; the mean was $58 \mu\text{g}/\text{m}^3$, with the highest and lowest values in Baoding and Sanya at 138 and $20 \mu\text{g}/\text{m}^3$, respectively. Summer ranked last with an average $\text{PM}_{2.5}$ concentration of $45 \mu\text{g}/\text{m}^3$, with the highest value in Shijiazhuang at $85 \mu\text{g}/\text{m}^3$ and the lowest in Sanya at $10 \mu\text{g}/\text{m}^3$. None of the average seasonal values reached the National Average Standard (Figure 2).

Figure 2 Monthly changes in $\text{PM}_{2.5}$ concentrations in the major cities of China in 2014

Monthly average $\text{PM}_{2.5}$ concentrations in Chinese cities showed a U-shaped pattern (Figure 2). It saw a downward trend from January to May, basically stable but a slightly decreasing trend from June to September and an increasing trend from October to December. The average values of January and February did not reach the Grade II standard. The highest monthly average value was observed in January, at $108 \mu\text{g}/\text{m}^3$, with the maximum being in Daqing ($280 \mu\text{g}/\text{m}^3$) and the minimum in Lhasa ($14.97 \mu\text{g}/\text{m}^3$). The lowest value was found in September ($33 \mu\text{g}/\text{m}^3$), reaching the air quality of the annual standard of China, while the values in all other months achieved the 24-h standard level. In March, October, November, and December, the average concentration had a range of $60\text{--}70 \mu\text{g}/\text{m}^3$, while from April to August, the value was $39\text{--}52 \mu\text{g}/\text{m}^3$.

Daily average $\text{PM}_{2.5}$ concentrations presented a periodic and impulse-shaped change in the major cities of China in 2014. The cycle between the peaks of spring and winter was relatively short, about seven days on average, while that of summer and autumn was longer, about 15 days. Overall, the $\text{PM}_{2.5}$ daily average concentration had a pattern of “high in spring and winter but low in summer and autumn” (Figure 3). The daily maximum value happened on January 17th at $148 \mu\text{g}/\text{m}^3$, while the minimum was on September 3rd at 27

Figure 3 Daily change of PM_{2.5} concentrations in the major cities of China in 2014

In terms of seasonal change and grades, PM_{2.5} concentrations indicate “there are more cities with high grades in winter and autumn but less in spring and summer”. According to the 24-h pollution level in China defined in the technical requirements based on the Ambient Air Quality Index (on trial) (AAQI), the PM_{2.5} concentrations were divided into six grades: excellent (0–35 µg/m³), good (35–75 µg/m³), slightly polluted (75–115 µg/m³), moderately polluted (115–150 µg/m³), heavily polluted (150–250 µg/m³), and seriously polluted (250–500 µg/m³). We calculated the daily PM_{2.5} concentrations in all 190 cities, and summed the number of days for each grade in each city. In 2014, the number of days with excellent air quality was 274, and only 17 cities reached this level. Based on the average PM_{2.5} concentrations in 2014, there were more cities and more days with excellent quality in summer, followed by autumn, and spring, while the fewest took place in winter. In summer, 86.84% of cities or 79 days reached the standard, followed by autumn (76.65% or 67 days), spring (75.57% or 66 days), and winter (55.51% or 49 days). Based on monthly data, 80% reached the standard from June to August, with August at the highest level (90.32%), followed by April, May, and October at 60%–70%; February, March, November, and December at 50%–60%; and January, the lowest, at 41.94%. There were more pollution days in spring and winter and fewer in summer and autumn. January, February, March, November, and December witnessed the largest proportion of pollution days (>30%), with the first month of the year at 58.06%; and heavily and seriously polluted air quality in spring and winter, with the latter occurring in January and February.

3.2 Spatial change

In terms of spatial distribution in 2014, the Hu Huanyong Line and Yangtze River were the respective E-W and S-N divides between high and low values. Kriging interpolation based on the semivariogram theory of variable values within a limited region is an optimal method for unbiased estimation that can explain the spatial distribution of PM_{2.5} concentrations (Wang *et al.*, 2015a). Cities with high PM_{2.5} concentrations were mainly located in the urban agglomerations in some regions east of the Hu Huanyong Line and north of the Yangtze River such as the Bohai Rim, Central Plains, Yangtze River Delta, middle Yangtze River, and Harbin-Changchun (Figure 4a). Cities with 72 µg/m³ of PM_{2.5} were all found within these regions, while regions south of the Yangtze River and west of the Hu Huanyong Line

(excluding western Xinjiang) had relatively low pollution levels.

According to the threshold of $35 \mu\text{g}/\text{m}^3$ (excellent air quality) in AAQS (GB 3095-2012), 17 cities reached the standard, and 90% of cities exceeded the value. In terms of the temporal distribution of exceeded values, the average ratio was 70% or 246 days. A total of 44 cities had excessive values for more than 300 days ($>85\%$); these were mainly distributed in the urban agglomerations in the regions of Beijing-Tianjin-Hebei, Yangtze River Delta, Central Plains, Yangtze-Huaihe river basin, and middle Yangtze River. A total of 105 cities had excessive values for 200–300 days ($57\%–85\%$), mainly distributed in the urban agglomerations of the regions including the Yangtze River Delta, Shandong Peninsula, East Liaoning Peninsula, Chengdu-Chongqing, Harbin-Changchun, Central Shanxi, Guanzhong-Tianshui, and northern slope of the Tianshan Mountains. Only 41 cities had excessive values for less than 200 days, mainly distributed in southeast coastal China such as the Pearl River Delta, as well as in Yunnan, Tibet, and Inner Mongolia (Figure 4b).

Figure 4 Spatial distribution of PM_{2.5} concentrations in the major cities of China in 2014

In terms of spatial distribution in seasonal change, cities with heavy pollution were mainly located in Beijing-Tianjin-Hebei and northwest Shandong Province. According to the Technical Regulation on Ambient Air Quality Index (on trial) HJ633-2012 of China, only a 24-h pollution level of PM_{2.5} in China is defined; annual and monthly levels have not yet been defined. Referring to GB 3096-2012 and HJ633-2012, we identified five grades of monthly average of PM_{2.5} pollution, namely, excellent and good ($0–35 \mu\text{g}/\text{m}^3$), slightly polluted ($35–75 \mu\text{g}/\text{m}^3$), moderately polluted ($75–115 \mu\text{g}/\text{m}^3$), heavily polluted ($115–150 \mu\text{g}/\text{m}^3$), and seriously polluted ($>150 \mu\text{g}/\text{m}^3$). In the aspect of seasonal change, cities with moderate pollution in spring (Figure 5a) were mainly observed in Northwest China, such as Xinjiang, those with slight pollution were mainly found in Shandong and Hebei. Most of the 20 cities with excellent air quality were distributed in coastal areas and northern China, and the remaining 154 cities had good air quality. Summer witnessed better air quality over a wider range (Figure 5b). Cities with slight pollution were mainly in Hebei. Sixty cities in coastal, southwestern, and northern China had excellent air quality, and the remaining 122 reached good quality. The situation was worse in autumn (Figure 5c) than in summer and spring. Beijing, Hebei, Shandong, and Henan had 28 cities with slight pollution and above. Winter was the worst season for air quality. In this season (Figure 5d), 15 heavily and 25 moderately

polluted cities were observed over a wider range compared with other seasons, including Beijing, Tianjin, Hebei, Shandong, Henan, Shaanxi, Hubei, Sichuan, and Heilongjiang. Meanwhile, the number of slightly polluted cities increased to 21 provincial units including Xinjiang.

Figure 5 Seasonal changes of air pollution in the major cities of China in 2014

As for monthly change, January, February, November, and December saw more polluted cities, which were distributed in 29 provincial units. Observation of polluted cities were 132, 99, 61, and 61 in January, February, November, and December, accounting for 69.47%, 52.11%, 32.11%, and 32.11% of the monitoring sites, respectively. Among them 73, 51, 14, and 5 cities with moderate pollution and above were found in January, February, November, and December, accounting for 38.42%, 26.84%, 7.37%, and 2.63% of the sites, respectively. As can be seen, the air quality was better in early winter, which was closely related to the environmental protection measures taken jointly by Beijing, Tianjin, Hebei, Shanxi, Inner Mongolia, and Shandong during the APEC Summit in November, including imposing vehicle limits and the closing of petrochemical and other polluting enterprises (Liu *et al.*, 2015); December continued this trend. Since March, the number of polluted cities had decreased, and was now dominated by slight pollution, mainly distributed in Beijing, Tianjin, Hebei, Shandong, and Henan. From April to September, there was an average of fewer than 17 polluted cities by month, among which 5 were in May and 4 in September. Beginning in October, the polluted areas expanded, with 40 cities mainly in Beijing, Tianjin, Hebei, Shanxi,

Henan, Liaoning, and Hubei. The monthly differences of urban $PM_{2.5}$ concentrations were a sharp contrast to the stable monthly urban economy in 2014. This evidenced the viewpoint that $PM_{2.5}$ was negatively associated with wind speed (Hu *et al.*, 2014).

3.3 Spatial agglomeration

The average $Z(I)$ of twelve months exceeded 2.58, suggesting that there was significant positive spatial autocorrelation of $PM_{2.5}$ concentrations in China. Analysis of annual and seasonal agglomerations indicated that hot spots were mainly in east coastal areas and the Bohai Rim, the Yangtze River Delta, Central Plains, and the northern slope of the Tianshan Mountains. The $Z(I)$ index values of $PM_{2.5}$ concentrations for annual (Figure 6a), spring (Figure 6b), summer (Figure 6c), autumn (Figure 6d), and winter (Figure 6e) are 28.32, 16.65, 27.25, 20.91, and 27.22, respectively, showing a similar spatial agglomeration. Hot spots were mainly in the Bohai Rim and the Yangtze River Delta region in east coastal China as well as Central Plains, Guanzhong-Tianshui and Inner Mongolia. Furthermore, three monitoring cities in Xinjiang had a high agglomeration of $PM_{2.5}$ concentrations, which was related to frequent sand storms in spring. Cold spots were mostly in the Pearl River Delta region in south coastal China as well as Yunnan and Tibet. No significant spots were found in the Yangtze-Huaihe river basin, middle Yangtze River and Chengdu-Chongqing regions or in Northwest and Northeast China. Figure 6 demonstrates that there were stable and contiguous polluted areas in the Bohai Rim, the Yangtze River Delta region, and the Central Plains, where there were more urban agglomerations. Results showed that urban agglomerations in northern China was the region with high $PM_{2.5}$ concentrations, while southeast coastal China was a stable and continuous area with excellent and good air quality.

Figure 6 Spatial agglomeration of annual and seasonal $PM_{2.5}$ concentrations in the major cities of China in 2014

Analysis of monthly variations indicated that $PM_{2.5}$ concentrations presented a cycle with two advances and two retreats. Local Moran's I correlation exhibited periodic monthly changes of $PM_{2.5}$ concentrations in the cities of China. In January (Figure 7a), February

(Figure 7b), and March (Figure 7c), hot spots were widely distributed in central and south-east coastal China and their ranges retreated gradually north of the Yangtze River. January was dominated by hot spots and no significant spots. The Yangtze River was a distinct divide between hot spots and cold spots in February and March, suggesting that the air quality, along with the end of coal combustion for heating, improved when the polluted range retreated from southeast to north of the Yangtze during January–March. The Z(I) index tended to be the lowest in April (Figure 7d) and May (Figure 7e) during the year. During these two months, no significant spots were dominant and this extended from southeast coastal to inland areas, indicating that as spring arrives, winds tended to be strong in northern China. Thus, the polluted range tended to disappear to western regions. The range of hot spots

Figure 7 Spatial agglomeration of monthly PM_{2.5} concentrations in the major cities of China in 2014

extended again in June (Figure 7f) and July (Figure 7g). Meanwhile that of cold spots extended in the areas south of the Yangtze River, suggesting that the increased precipitation from the monsoon climate purified the air of PM_{2.5}. As a result, PM_{2.5} concentrations decreased in each city. However, there was high agglomeration in the Yangtze River Delta region and North China, indicating that industrial and population agglomerations played a leading role in the increase of PM_{2.5} concentrations. Subsequently, the range of hot spots and cold spots retreated to the west and north. No significant spots changed from the Yangtze River Delta region and the Bohai Rim to the inland areas in August (Figure 7h), September (Figure 7i), and October (Figure 7j), suggesting that PM_{2.5} pollution spread to inland areas, and that it tended to be stable in the North China Plain in October. Along with the successive start of coal combustion for heating, hot spots dominated by high PM_{2.5} concentrations were densely distributed in areas north of the Yangtze River in November and December (Figures 7k and 7l). Meanwhile, the range of cold spots dominated by excellent and good air quality tended to be stable. In summary, PM_{2.5} concentrations throughout the year were cyclical, namely, a wide distribution in eastern and central regions – a retreat to northern China – a wide distribution in eastern and central regions. A small number of the cities with high agglomeration were observed only in April, May, September, and October, which was closely associated with the monsoon climate (Hu *et al.*, 2014).

4 Conclusions and discussion

(1) The PM_{2.5} concentrations in Chinese cities had an apparent U-shaped seasonal variation and an impulse-shaped daily variation. The annual average PM_{2.5} concentrations of 190 monitoring cities in 2014 were 61 µg/m³, reaching the national Grade II standard. The PM_{2.5} concentrations in winter were the highest, followed by that of autumn and spring, both reaching the national Grade II standard, whereas the concentrations in summer were the lowest, at the national Grade I standard. The monthly average PM_{2.5} concentration curve showed a U-shaped pattern, reaching a maximum in January and a minimum in September. During 2014, September was the only month that reached the Grade I standard, while the remainder belonged to the Grade II standard. The daily average PM_{2.5} concentrations throughout the year also presented a U-shaped curve of “high in spring and winter, but low in summer and autumn.” The daily peak in the year happened on January 18th (138 µg/m³), while the minimum value appeared on September 3rd (26 µg/m³). According to the national annual statistical standard for the assessment of air pollution, there were 9.47% of the cities with excellent air quality, 71.58% with good, 17.37% slightly polluted, 1.58% moderately polluted and no heavily or seriously polluted cities. In terms of the monthly change, cities above the moderately polluted level were mainly observed in winter.

(2) The PM_{2.5} concentrations in Chinese cities showed distinct spatial distribution. The Hu Huanyong Line and Yangtze River were the respective E-W and S-N divides between the high and low values of China. Cities with high PM_{2.5} concentrations were mainly located in the urban agglomerations in regions east of the Hu Huanyong Line and north of the Yangtze River, such as the Bohai Rim, Central Plains, Yangtze River Delta, Middle Yangtze River, and Harbin-Changchun (Figure 5a). Cities with 72 µg/m³ of PM_{2.5} were all found within this area, while regions south of the Yangtze River and west of the Hu Huanyong Line (exclud-

ing western Xinjiang) had relatively low pollution levels. The PM_{2.5} pollution varied from season to season. The largest polluted extent was observed in winter, including 21 provincial units, while the smallest one was found in summer, which was primarily in Hebei Province. From summer to winter, the polluted extent expanded from the North China Plain to other regions, especially to the southwest. The PM_{2.5} pollution also presented inter-monthly variations, with the greatest polluted extent in January, February, November, and December, covering eastern and central China. The smallest polluted extent was reported in May and September. Affected by climate, the polluted extent started to retreat in March but was enlarged in October.

(3) The PM_{2.5} concentrations had spatial agglomeration, with the urban agglomerations of northern China as the core areas. The North China Plain and the Yangtze River Delta regions were the main PM_{2.5} agglomerated areas in the study year, while the southeast coastal areas had stable excellent or good air quality. The PM_{2.5} concentrations had intra-annual periodicity by month. The PM_{2.5} agglomerated areas covered eastern and central China from January to March and the polluted extent started to retreat to the north of the Yangtze River from the south. The extent tended to disappear in western China in April and May. The polluted area expanded again to the North China Plain in the following June and July. In August and September, the polluted extent spread to the inland area and remained stable in the North China Plain in October. Subsequently, the polluted extent continued to expand to northern China, as it reached the end of the cycle. The extent with high values shrank in April, May, September, and October, which coincided with the prevailing monsoon.

(4) This paper contains some limitations to research on PM_{2.5} concentrations. Due to the short monitored history in China (since the beginning of 2013), only data from 2014 is included in this paper, which therefore does not reveal variations in PM_{2.5} concentrations over a longer period. The working population in different industries could only show the relationship between influencing factors and PM_{2.5} concentrations. Further studies are needed with regard to the contribution that specific industries and production processes can make to PM_{2.5} concentrations.

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