

Yan, D., Lei, Y., Shi, Y., Zhu, Q., Li, L. and Zhang, Z. (2018) Evolution of the spatiotemporal pattern of PM2.5 concentrations in China – A case study from the Beijing-Tianjin-Hebei region. *Atmospheric Environment*, 183, pp. 225-233.(doi:10.1016/j.atmosenv.2018.03.041)

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Deposited on: 30 April 2018

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1 2	Evolution of the spatiotemporal pattern of PM2.5 concentrations in China – a case study from the Beijing-Tianjin-Hebei region
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15	
16	Abstract:
17 18	Atmospheric haze pollution has become a global concern because of its severe effects on human health and the environment. The Beijing-Tianjin-Hebei urban
19 20	agglomeration is located in northern China, and its haze is the most serious in China. The high concentration of PM2.5 is the main cause of haze pollution, and thus
21	investigating the temporal and spatial characteristics of PM2.5 is important for
22	understanding the mechanisms underlying PM2.5 pollution and for preventing haze.
23	In this study, the PM2.5 concentration status in 13 cities from the Beijing-Tianjin-
24	Hebei region was statistically analyzed from January 2016 to November 2016, and the
25	spatial variation of PM2.5 was explored via spatial autocorrelation analysis. The
26	research yielded three overall results. (1) The distribution of PM2.5 concentrations in
27	this area varied greatly during the study period. The concentrations increased from
28	late autumn to early winter, and the spatial range expanded from southeast to
29	northwest. In contrast, the PM2.5 concentration decreased rapidly from late winter to
30	early spring, and the spatial range narrowed from northwest to southeast. (2) The

31 spatial dependence degree, by season from high to low, was in the order winter,

32 autumn, spring, summer. Winter (from December to February of the subsequent year)

- and summer (from June to August) were, respectively, the highest and lowest seasons
- with regard to the spatial homogeneity of PM2.5 concentrations. (3) The PM2.5
- 35 concentration in the Beijing-Tianjin-Hebei region has significant spatial spillovers.
- 36 Overall, cities far from Bohai Bay, such as Shijiazhuang and Hengshui, demonstrated
- a high-high concentration of PM2.5 pollution, while coastal cities, such as Chengde
- and Qinhuangdao, showed a low-low concentration.

Keywords: Beijing-Tianjin-Hebei region; Air pollution; PM2.5 concentration; spatial
autocorrelation analysis.

41

42 **1. Introduction**

43 China's serious haze problem has aroused widespread concern among the public. 44 The increase in the concentrations of particulates (including PM10 and PM2.5) in the 45 atmosphere is the main cause of haze production (Xu and Lin, 2016; Hsu et al., 2017; Liao et al., 2017; Liu et al., 2015a). Studies have shown that high levels of PM2.5 and 46 47 PM10 are closely linked to high concentrations of fungi and bacteria in the air, which 48 can cause serious harm to humans (Liu et al., 2017a; Liu et al., 2017b; Liu et al., 2015b). In 2012, China issued newly revised ambient air quality standards; PM2.5, 49 regarded as a routine indicator of atmospheric pollution, was included in the new 50 standard and has become the key focus of atmospheric pollution research. PM2.5 51 52 refers to fine particles with a dynamic diameter of less than 2.5 µm. These particles 53 are composed of a wide variety of complex chemical substances emitted from various natural and anthropogenic sources (Alves et al., 2012; Wang et al., 2012; Zhao et al., 54 55 2014). A high concentration of PM2.5 not only significantly reduces atmospheric visibility but also leads to increased incidences of respiratory and cardiovascular 56 57 diseases (Dockery et al., 1993; Li et al., 2016; Chalbot et al., 2014; Hu et al., 2014). At the beginning of 2013, China suffered from the most serious fog and haze 58 pollution since the start of observational records, and this rare continuous high-59 intensity air pollution swept the middle and eastern parts of China. The most serious 60 pollution occurred in the Beijing-Tianjin-Hebei (BTH) region, where the daily PM2.5 61 concentration reached 500 μ g/m³(Wang et al., 2014). The BTH region is China's 62 political and cultural center and is an important core area of North China's economy. 63 Since the 1980s, the economy, society and culture of the BTH region have developed 64 remarkably. The gross domestic product (GDP) increased approximately seven-fold 65 66 from 593.342 billion yuan in 1980 to 4715.233 billion yuan in 2013(Beijing

67 Statistical Yearbook 2014; Tianjin Statistical Yearbook 2014; Hebei Economic Yearbook 2014). However, at the same time, serious air pollution exists in the 13 68 cities within the BTH region. According to the Ministry of Environmental Protection, 69 70 the worst ten cities in terms of air quality in 2015 were Baoding, Xingtai, Hengshui, Tangshan, Zhengzhou, Jinan, Handan, Shijiazhuang, Langfang and Shenyang. Seven 71 of these cities are within the BTH region. The haze pollution in the BTH region 72 cannot be overlooked, and haze governance has become the top priority for the BTH 73 74 region (Cai et al., 2017). Therefore, studying the temporal and spatial characteristics of PM2.5 in this region has great practical value. Understanding the spatial variation 75 of the PM2.5 concentration will not only add to our knowledge of the mechanism of 76 air pollution but also provide a scientific reference for implementing targeted control 77 measures. 78

The consumption of fossil energy, such as coal and oil, generates a large amount of waste gas, which affects the climate and endangers human health. Therefore, energy-saving emission reduction is crucial (Ma et al., 2017a; Yan et al., 2017). Many scholars have focused on carbon emissions reduction in their research (Shuai et al., 2017; Ma et al., 2017b; Ma et al., 2017c), and now society as a whole is beginning to pay attention to the haze issue in China.

85 Existing research on the haze problem mainly focuses on two aspects. Initially, 86 most scholars analyzed the composition of PM2.5 from the physical and chemical perspectives (Bates and Sizto, 1987; Hussain et al., 2013; Jansen et al., 2014) and 87 concluded that PM2.5 was mainly composed of industrial waste gas, automobile and 88 machine exhaust, cooking oil smoke and dust. These pollutants are closely related to 89 90 socioeconomic factors such as GDP, population, energy consumption and industrial infrastructure. In light of these findings, a large number of studies have focused on the 91 92 economic and social drivers of the PM2.5 concentration. For example, based on the 93 data on the PM2.5 concentration and the air quality index (AQI) in 73 Chinese cities in 2013, Hao and Liu (2016) analyzed the influencing factors of the PM2.5 94 95 concentration in China's cities and discussed how economic and social development could affect air quality. Their results show that the relationship between the PM2.5 96 97 concentration and GDP per capita exhibits an inverted U shape and that car ownership 98 and secondary industry have significant effects on the PM2.5 concentration. Luo et al. 99 (2017) explored the driving factors of the PM2.5 concentration in China using a 100 geographical regression weighting model, which confirmed the existence of the inverted U-shaped environmental Kuznets curve (EKC) for air quality and that the 101 potential influencing factors of each significant area were different. By using the 102

103 input-output framework and structural decomposition analysis, Guan et al. (2014) studied the socioeconomic drivers of China's primary PM2.5 emissions and found 104 that export is the only final demand category that led to emission growth between 105 106 1997 and 2010. The embodied PM2.5 emissions from Chinese exports are mainly driven by consumption in OECD countries. The second studied aspect is the spillover 107 effects of interregional air pollution. SO₂, NO_x and soot are often used as proxy 108 variables for air pollution (Civan et al., 2015; Moroń et al., 2015; Zhao et al., 109 110 2017). Spatial variability analysis in geography is an important basis for simulating the spatial distribution of variables and revealing the spatial effects of variables (Liu et al., 111 2016; Zuo et al., 2015). Furthermore, spatial autocorrelation analysis is an important 112 technical method that has recently been applied to the field of environmental pollution, 113 particularly air pollution. For example, Zhang et al. (2016) examined the spatial 114 clustering types of CO₂ emission efficiency in 30 provinces in China and confirmed 115 that there is indeed a spatial spillover effect among Chinese provinces. To gain insight 116 into the characteristics of air pollution, Wu et al. (2017) explored the characteristics 117 and determinants of PM2.5 pollution in China using spatial econometrics. Yan et al. 118 (2017) used Moran's index to examine the spatial effects of the power industry in 119 120 various regions of China. The empirical results showed that there is indeed a significant spatial agglomeration effect between carbon emission efficiencies, mainly 121 for high-high and low-low agglomeration types. 122

123 The concentration of air pollutants is affected by the intensity of emission sources and by terrain and meteorological conditions; furthermore, it has remarkable 124 125 temporal and spatial variability. In China, PM2.5 started to attract attention in 2012. The majority of the literature is devoted to the analysis of the formation of fog and 126 127 haze and to the chemical composition of PM2.5 or its influencing factors at the 128 national scale, whereas studies on the distribution pattern of PM2.5 concentrations on the urban scale are relatively rare. Compared with previous research, this study has 129 two main contributions. Thematically, through the PM2.5 pollution index, we analyze 130 the spillover effect of haze between different cities in the BTH urban agglomeration. 131 132 The results will allow the general public to clearly understand the spatiotemporal rules of haze pollution in this urban agglomeration and provide new evidence for haze 133 management. Methodologically speaking, abandoning the geospatial homogeneity 134 hypothesis in spatial econometrics would better fit the real situation of the PM2.5 135 concentration and facilitate the examination of the propagation path of haze pollution. 136 Specifically, we collected data on daily PM2.5 concentrations in 2016 in 13 cities 137 located within the BTH region and studied the spatial autocorrelations and 138

aggregation patterns of PM2.5 concentrations during different seasons. The results
were intended to provide a basis for simulating PM2.5 concentrations in the region,
elucidating the underlying factors contributing to this pollution, and devising an
effective monitoring point layout.

The remaining parts of this paper are organized as follows. The second section describes the data source and model description. In the third section, we present the temporal and spatial variation of PM2.5 concentrations in the BTH region. The fourth section describes and discusses the results of spatial autocorrelation, which is

147 followed by the final conclusions and policy implications in the fifth section.

148

149 2. Data and methodology

150 2.1 Overview of the BTH region and data sources

In 2013, China established a total of 612 PM2.5 concentration monitoring sites in 151 74 cities, and the number of monitoring sites in each city was different. By 2015, the 152 153 monitoring sites increased to 1436 (Wang et al., 2015). Presently, the sites that 154 monitor the PM2.5 concentration in China are mainly concentrated in the Pearl River Delta, Yangtze River Delta and BTH region. Among these areas, the urban 155 agglomeration of the BTH region suffers from relatively more severe haze pollution 156 157 than other cities, and some studies have suggested that this was because several coal-158 based industries, such as coal-fired power plants and steel manufacturing, are stationed in the region (Zhao et al., 2012). This area is adjacent to Bohai Bay and is 159 composed of 13 cities, namely, Beijing, Tianjin, Shijiazhuang, Xingtai, Handan, 160 Hengshui, Baoding, Cangzhou, Langfang, Tangshan, Zhangjiakou, Chengde and 161 162 Qinhuangdao. The land area of this region is 218,000 square kilometers, and the 163 resident population is approximately 110 million people. Industrialization has greatly bolstered the BTH region's economic development, and the main industrial types are 164 steelmaking, petroleum processing and coking, nuclear fuel processing, building 165 materials manufacturing and chemical manufacturing. The common feature of these 166 167 industries is high energy consumption; thus, a large amount of atmospheric emissions is produced. In 2015, the total industrial emissions from this region was 4.138 million 168 tons, including dust, sulfur dioxide and nitrogen dioxide. These chemical substances 169 are precursors of sulfate and nitrate of PM2.5. 170

In this study, the hourly PM2.5 concentration data of the urban air qualitymonitoring sites in 13 cities in 2016 were obtained from the China Environmental

5

173 Monitoring Station as raw data. According to the arithmetic average method, these data from cities or monitoring sites can be calculated as daily averages, monthly 174 averages, quarterly averages and as an annual average within 2016. In accordance 175 with the requirements of the Ambient Air Quality Standard for the effectiveness of air 176 pollutant concentration data, the PM2.5 data were pretreated. First, we removed 177 missing values. Second, when the monthly mean was calculated, the monitoring site 178 was eliminated if the number of daily PM2.5 concentration measurements was less 179 180 than 27 days in a month. Finally, a small number of abnormal monitoring points were eliminated (such as hourly PM2.5 concentrations greater than 900 µg.m⁻³ or hourly 181

182 PM2.5 concentrations less than 0).

183 2.2 Spatial interpolation method

The spatial interpolation method is often used to convert point scale data into a 184 continuous surface scale. Additionally, this method helps us better understand the full 185 spatial distribution of variables in the region. The accuracy of the spatial interpolation 186 187 method is greater than that of remote sensing inversion (Lee S J et al., 2012). Many specific methods are commonly used for spatial interpolation, such as the Inverse 188 Distance Weighted (IDW) method and the Ordinary Kriging method (OK). The 189 accuracy of the estimates using the former method is influenced by the distance to a 190 191 known point, and the requirements for the dispersion and uniformity of the 192 interpolation points are relatively higher. The latter method will generate the best estimation algorithm for the output surface and will create a comprehensive 193 calculation of the spatial behavior of the interpolation point attribute prior to the 194 generation of the algorithm; thus, the results from the second method are preferable 195 196 (Zhang et al., 2013). Generally, the OK method can reflect the spatial distribution of PM2.5 more scientifically and the result has second-order stationarity. 197

198 2.3 Spatial clustering analysis of the PM2.5 concentration based on Moran 's Index

According to the first law of geography, entities with geographical attributes are 199 related with each other. Clustering, random and regular distribution exist, and 200 correlation decreases with distance (Tober, 1970). This phenomenon is called spatial 201 202 autocorrelation. Spatial autocorrelation statistics can describe the potential 203 interdependence or compactness of variables within the same area. This approach is often used to analyze the spatial agglomeration and trend of geographical elements to 204 provide evidence for exploring spatial and temporal clustering and evolution. The 205 206 spatial correlation of atmospheric activity may reveal that the values of PM2.5 207 concentrations in similar areas are statistically close (Wu et al., 2015). Spatial

autocorrelation analysis includes global spatial autocorrelation and local spatialautocorrelation, in which the global Moran Index is calculated as follows:

210
$$I = \frac{n \sum_{i=1}^{n} \sum_{j=1}^{n} w_{ij} Z_i Z_j}{S_o \sum_{i=1}^{n} Z_i^2}$$
(1)

where
$$S_o = \sum_{i=1}^{n} \sum_{j=1}^{n} w_{ij}$$
, and w_{ij} is the spatial weight matrix; in this paper, the
adjacent unit is 1, and the remaining units are zero. The Moran Index is in the range [-
1, 1]. A value less than 0, equal to 0, or greater than 0 indicates negative correlation,
no correlation or positive correlation, respectively. For the Moran Index, the
standardized statistic *Z* can be used to test the existence of spatial autocorrelation. The
formula for the standardized statistic *Z* is as follows:

217
$$Z_i = \frac{I - E[I]}{\sqrt{V[I]}}$$
(2)

218
$$E[I] = -1/(n-1)$$
 (3)

219
$$V[I] = E[I^2] - E[I]^2$$
 (4)

Among these formulas, at the 0.05 significance level, Z(I)>1.96 indicates positive spatial autocorrelation between PM2.5 spatial units, and -1.96<Z(I)<1.96 indicates that the spatial correlation of PM2.5 concentrations is not obvious. If Z(I)<-1.96, then there is a negative correlation between PM2.5 spatial units, and the attribute value tends to be distributed. Local spatial autocorrelation is used to determine the specific location of spatial agglomeration, and the local Moran Index is calculated as follows:

226
$$I_i = \frac{X_i - \overline{X}}{S_i^2} \sum_{j=1, j \neq i}^n w_{ij} (X_i - \overline{X})$$
(5)

227 where X_i is the attribute value of i, \overline{X} is the average value, and w_{ij} is the spatial 228 weight matrix; then,

229 $S_i^2 = \frac{\sum_{j=1, j \neq i}^n w_{ij}}{n-1} - \overline{X}^2$ (6)

230 The standardized statistic of local Moran Index test is Z[I]:

231
$$Z_{Ii} = \frac{I_i - E[I_i]}{\sqrt{V[I_i]}}$$
(7)

232
$$E[I_i] = -1 / (n-1)$$
 (8)

233
$$V[I_i] = E[I_i^2] - E[I_i]^2$$
(9)

At the 0.05 significance level, Z>1.96 indicates that cities with high PM2.5 concentrations are surrounded by other cities with high PM2.5 concentrations and that cities with low PM2.5 concentrations are surrounded by other cities with low PM2.5 concentrations. Z<-1.96 indicates that cities with high PM2.5 concentrations are surrounded by cities with low PM2.5 concentrations and that cities with low PM2.5 concentrations are surrounded by cities with high PM2.5 concentrations. When Z=0, the observations demonstrate an independent random distribution.

241

3. The variation and regularity of PM2.5 concentrations in the BTH region

243 3.1 The monthly variation characteristics of PM2.5 concentrations

244 An investigation of the PM2.5 concentration changes from the cities in the BTH region over the 12 months in 2016 (see Figure 1) reveals that the median monthly 245 PM2.5 concentrations in the 13 cities demonstrate U-shaped oscillations over the 246 entire time period. Specifically, the PM2.5 concentration demonstrated a downward 247 trend from January to May, was overall stable from June to August, and finally 248 249 increased from October to December. Among the latter months, the highest peak occurred in December and was 147.34 μ g.m⁻³; meanwhile, the lowest peak appeared 250 in August and was 38.95 µg.m⁻³. From May to September, the median PM2.5 251 concentration was under 60 μ g.m⁻³; thus, this time represents the highest air quality 252 period in the BTH region within the entire year. In general, PM2.5 concentrations 253 showed significant differences by month. In the winter, PM2.5 pollution is the most 254 severe, but during spring, it begins to decrease and eventually maintains a stable state 255 256 in the late spring. By the summer, PM2.5 pollution decreases to the lowest level and then again begins to increase during late autumn. It can be reasonably speculated that 257 the PM2.5 concentration value varies with the seasonal weather and forms a cyclical 258 259 change pattern.

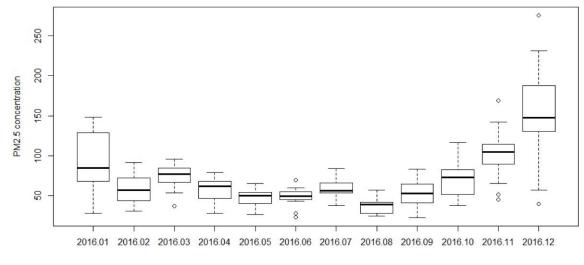






Fig. 1 The monthly average PM2.5 concentrations of cities in the BTH region

263 3.2 The seasonal variation in characteristics of PM2.5 concentrations

264 Hourly data of PM2.5 concentrations were collected during spring (from March 265 to May), summer (from June to August), autumn (from September to November) and winter (from December to February), and the quarterly averages and annual averages 266 of PM2.5 concentrations in each city were obtained (see Table 1). The average annual 267 PM2.5 concentration in the BTH region was 69.38 μ g·m⁻³, which is far above the 268 secondary standard limit (35 µg.m⁻³) of the Ambient Air Quality Standard. In the 269 same period, the average annual PM2.5 concentration in 338 cities was 47 μ g.m⁻³. 270 Furthermore, in the Yangtze River Delta and Pearl River Delta, the average annual 271 PM2.5 concentrations were 46 μ g.m⁻³ and 32 μ g.m⁻³, respectively, indicating that the 272 PM2.5 pollution in the BTH region is still the most serious among all the studied 273 274 areas. From the perspective of seasonal changes, the seasonal differences in ambient air pollution in the BTH region were obvious. Specifically, the PM2.5 concentration 275 in winter was approximately twice that in summer, and heavy pollution weather 276 appeared primarily in winter, especially during the heating period. Among the winter 277 months, the PM2.5 concentration in the BTH region was $135 \ \mu g.m^{-3}$ during the 278 heating period from November 15th to December 31st, 2016, which was 2.4 times 279 that of the values during the non-heating period. Five large-scale air pollution 280 processes occurred only in December. From the perspective of the city, PM2.5 281 concentrations in all cities in the BTH region, except Zhangjiakou, were above the 282 283 national secondary standards. The annual average PM2.5 concentration in Baoding

was the highest at 93.91 μ g.m⁻³, which exceeds the national secondary standard limit by 168%.

City		Р	(PM2.5)/(µg.m ⁻	3)	
	Spring	Summer	Autumn	Winter	Annual mean
Beijing	71.35	58.16	79.55	87.72	74.2
Tianjin	64.95	49.01	73.68	83.16	67.7
Shijiazhuang	65.88	45.27	122.82	121.56	88.88
Qinhuangdao	45.89	36.63	46.99	50.02	44.88
Xingtai	68.31	52.99	91.25	134.67	86.81
Handan	65.17	48.24	71.69	118.13	75.81
Baoding	70.13	54.42	102.58	148.5	93.91
Chengde	40.32	30.07	40.11	52.86	40.84
Langfang	53.08	49.45	61.87	101.68	66.52
Zhangjiakou	30.02	28.88	34.82	30.81	31.13
Hengshui	74.4	66.09	78.93	143.67	90.77
Cangzhou	56.41	48.06	73.15	92.83	67.61
Tangshan	65.4	53.03	81.78	91.13	72.84
BTH	59.33	47.72	73.79	96.67	69.38

286 Seasonal and annual means of the PM2.5 concentration in cities of the BTH region during287 2016

288

289 3.3 The spatial distribution characteristics of PM2.5 concentration

By using ArcGIS software and the Kriging interpolation method, we evaluated
the spatial interpolation of PM2.5 concentrations in the BTH region by month in 2016.
The results are shown in Figure 2. The spatial interpolation of the PM2.5
concentration was characterized by severe haze in the southern region, relatively light
haze in the northern region and a slightly prominent pattern in some areas.
Furthermore, the difference between north and south was large. Southern cities,

296 including Baoding, Shijiazhuang, Xingtai and Handan, within Hebei Province

- 297 exhibited the highest concentrations. Through the combination of temporal and spatial
- analysis, we demonstrate that the PM2.5 concentration in the BTH region began to
- 299 increase from December to February in the following year; haze first appeared in the
- southern region but expanded to cover the whole area by February. From March to
- 301 May, the scope of fog and haze narrowed from northwest to the southeast and then
- 302 remained stable until September. The haze pollution suddenly increased in October
- and demonstrated a growing trend.

Fig. 2 Spatial distribution of PM2.5 concentrations of cities in the BTH region in 2016

306

305

307 4. Results and discussion

- 308 4.1 Spatial autocorrelation test of the PM2.5 concentration
- 309 The global spatial autocorrelation analysis can be used to compare the spatial
- spillover effect of PM2.5 concentrations within different months. As shown in Table 2,
- the global Moran Index in terms of the PM2.5 concentration in the BTH region during

each of the twelve months from January to December was 2.971, 3.036, 0.732, 1.325,

313 0.797, 0.181, 2.186, 0.746, 1.190, 1.380, 1.380, 2.482, and 2.372, respectively. These

data were obtained using GeoDa software. The Z(I) values of the PM2.5

315 concentrations were greater than 1.96 in January, February, July, November and

316 December, and the significance test indicated that the PM2.5 concentrations in these

317 months were spatially homogeneous. As a function of season, winter (from December

to February of the following year) and summer (from June to August) respectively

represent the highest and lowest spatial autocorrelation seasons of the PM2.5

320 concentration within one year. In other words, the spatial spillover effect is higher in

321 these two seasons than in the other seasons, and PM2.5 pollution is more homogenous

322 during these two seasons

Table 2 Spatial autocorrelation index of PM2.5 concentrations in the BTH region of China in
 2016

Month	Global Moran's Index	Std-err	P-value	Z-stat
1	0.493	0.193	0.001	2.971
2	0.495	0.194	0.001	3.036
3	0.039	0.176	0.213	0.732
4	0.144	0.179	0.103	1.325
5	0.056	0.181	0.208	0.797
6	0.117	0.184	0.305	0.181
7	0.321	0.183	0.014	2.186
8	0.049	0.182	0.218	0.746
9	0.126	0.186	0.126	1.190
10	0.155	0.177	0.084	1.380
11	0.322	0.167	0.013	2.482
12	0.354	0.186	0.012	2.372

325

4.2 The spatial pattern evolution characteristics of PM2.5 concentrations in the BTH

327 region

328 Local spatial autocorrelation analysis was used to analyze the spatial pattern evolution characteristics of PM2.5 pollution and included Moran's Index scatter plot 329 and local indicators of spatial association (LISA) agglomeration analysis. Figure 3 330 shows the global Moran Index scatter plot for each season of the BTH region in 2016. 331 332 The abscissa represents standardized PM2.5 concentration in cities, and the ordinate is the neighboring PM2.5 concentration value as determined by the spatial weight matrix 333 based on the Euclidean distance, also known as the space lag vector. The four 334 335 quadrants of the Moran Index scatter plot represent different agglomeration types. The first quadrant indicates the high-high (HH) agglomeration zone, which means that the 336 PM2.5 concentrations in a city and in its surrounding cities are high. The second 337 quadrant indicates the low-high (LH) aggregation zone, which means that the PM2.5 338 concentration in a city is low, but values in the surrounding cities are high. The third 339 and fourth quadrants indicate low-low (LL) and high-low (HL) aggregation zones, 340 respectively. The HH and LL agglomeration zones reflect the homogeneity of PM2.5 341 pollution, which indicates positive spatial autocorrelation. The HL and LH 342 aggregation zones reflect the heterogeneity of PM2.5 pollution, which indicates 343 negative spatial autocorrelation. The spatial correlation of PM2.5 concentration in the 344 345 BTH region varies with seasons, and the spatial spillover effect in winter is the most significant. The cities with the highest PM2.5 concentration are clustered near the 346 origin point in spring and show strong spatial heterogeneity. 347

349

Fig. 3 Global Moran Index scatter plot of PM2.5 concentrations in different seasons

350 Figure 4 shows that overall, Shijiazhuang, Hengshui and other cities that are located far from the Bohai Bay are HH agglomeration centers. This finding may be 351 due to favorable conditions for air diffusion in coastal cities and to the fact that 352 atmospheric pollutants are easily spread. Meanwhile, Chengde, Qinhuangdao and 353 354 other coastal cities showed LL agglomeration characteristics, which may be attributed 355 to the low and flat terrain of the southern BTH region, which is not conducive to PM2.5 diffusion. Furthermore, the spatial dependence of the PM2.5 concentration in 356 the urban agglomeration of the BTH region shows a periodic change. During the 357 period from November to February of the following year, the Z-value index was the 358 highest, which indicated that the agglomeration was obvious. This phenomenon is 359 mainly caused by the initiation of coal-fired heating in the northern parts, leading to 360 the spread of PM2.5 from Shijiazhuang and Hengshui to neighboring cities and 361

triggering an increasing area of fog and haze. From March to June, the Z-value index
was reduced to the lowest level in the whole year. Because no feature points
dominated and the spatial homogeneity was weakened, the range of PM2.5 pollution
tended to disappear. From July to October, the scope of the HH agglomeration zone
expanded again, and the fine air quality of the northern city tended to be stable. This
stability may be caused by the increase in rainfall in summer, which would enhance
the purification effect on PM2.5.

Interestingly, a regional haze pollution community has been formed, as the 369 PM2.5 concentration in the BTH region has shown obvious convergence 370 characteristics. This phenomenon may exist because the BTH region is the most 371 concentrated area of China's steel industry, and this industry consumes a great deal of 372 373 fossil fuel and produces a large amount of industrial emissions. During the autumn and winter of the heating period in the BTH region, coal smoke pollutants caused by 374 industrial boilers and heating boilers increased significantly. Although the atmosphere 375 is stable, the frequency and intensity of inversion is high and thus prone to the 376 377 agglomeration of pollutants. However, the weather is dry, windy and rainy in the spring, and when summer arrives, the atmospheric stability decreases with 378 379 concentrated rainfall. Under these conditions, these seasons are not conducive to the formation of concentrated pollutants. Air is a public good without property rights, and 380 despite the serious pollution of fog and haze, regional governments have introduced a 381 382 large number of high energy-consuming industries in order to develop the economy. Thus, the internal costs have been externalized. There is a linkage between the haze 383 pollution in the cities of the BTH region. If governance measures are implemented 384 only for a single city, the elevated PM2.5 concentration in the surrounding area will 385 still cause the local haze concentration to increase. Therefore, regional joint 386 387 governance is needed.

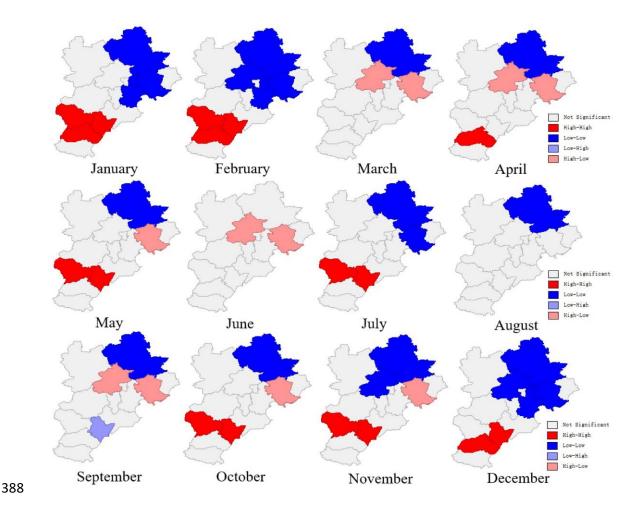


Fig. 4 Monthly spatial agglomeration diagram of PM2.5 concentrations in the BTH region in
 2016

392 5. Conclusions and policy implications

The urban agglomeration in the BTH region is a typical haze-prone area, and 393 394 studying the spatial pattern evolution characteristics of PM2.5 concentrations in the BTH region is important for understanding the mechanism of PM2.5 pollution and the 395 prevention of haze phenomenon. Based on the PM2.5 data released by the China 396 Environmental Monitoring Station, this research analyzed the spatial autocorrelation 397 398 degree and spatial clustering pattern of PM2.5 concentrations in different seasons of 399 the BTH region based on spatial dependence theory. The primary conclusions are follows. 400

401 (1) The distribution of PM2.5 concentrations in this area varied greatly in 2016.402 On one hand, it increased from late autumn to early winter, and the spatial range

403 expanded from southeast to northwest. On the other hand, the PM2.5 concentration
404 decreased rapidly from late winter to early spring, and the spatial range was narrowed
405 from northwest to southeast.

406 (2) The degree of spatial dependence by season was in the following order from
407 highest to lowest: winter, autumn, spring, summer. Winter (from December to
408 February in the following year) and summer (from June to August) were, respectively,
409 the highest and lowest seasons with regard to the spatial homogeneity of PM2.5
410 concentrations.

(3) The agglomeration pattern of PM2.5 concentrations in the BTH region is
significant. Generally, cities such as Shijiazhuang and Hengshui, which are located far
from Bohai Bay, exhibited a high-high concentration of PM2.5 pollution, whereas
coastal cities, such as Chengde and Qinhuangdao, demonstrated a low-low
concentration.

In 2013, the State Council issued the Air Pollution Prevention and Control 416 Action Plan. By 2017, the concentration of fine particles in the BTH region, the 417 Yangtze River Delta and the Pearl River Delta decreased by 25%, 20% and 15% 418 respectively. Among these areas, the BTH region had the most stringent reduction 419 420 targets because of its severe air pollution. The PM2.5 concentration is the result of the atmospheric reaction of air pollutants, which is affected by atmospheric diffusion 421 conditions. Reducing coal consumption, adjusting energy structure and implementing 422 other initiatives can reduce pollutant emissions, but it is difficult to determine the 423 424 specific air quality objectives that can be achieved. There are uncertainties in the 425 policy effects of air pollution control programs. Based on the above empirical results, the following suggestions are put forward for haze control in the BTH urban 426 agglomeration. Haze pollution exhibits a linkage relationship in the BTH region. If 427 only a single city is controlled, the high PM2.5 concentration in the surrounding cities 428 429 will increase the PM2.5 concentration in this city. Therefore, a regional joint governance approach should be adopted. For example, an air pollution joint defense 430 mechanism could be set up for early warning of PM2.5 pollution and rational 431 distribution of pollution control costs among cities in the BTH region through an 432 ecological compensation mechanism. In addition, the haze pollution in the BTH 433 434 region has two main features. First is the significant spatial difference. The haze pollution in the southern region is relatively more serious, while that in the northern 435 region is relatively light. Second is the significant seasonal difference. The haze 436 during the heating season is serious, while that during the non-heating season is 437

438 relatively light. Therefore, the policy focus for haze pollution control should be shifted to the control of pollutant discharge in the southern part of the BTH region and 439 should reduce the frequency of severe fog and haze occurrences during the heating 440 441 season. Finally, monitoring points in the southern region of the BTH region should be increased, and cities far from Bohai Bay, such as Shijiazhuang and Hengshui, should 442 be listed as key cities for prevention and control. Generally, regional joint 443 management is of great value in controlling pollutant emissions and improving the 444 quality of the atmospheric environment. It is necessary to improve the BTH urban 445 agglomeration's collaborative facilities and to promote the integration of the BTH 446 region in order to minimize governance cost. 447 448 Presently, existing studies on the PM2.5 concentration in China still concentrate on PM2.5 components, source and space-time phenomena. The factors 449 450 influencing PM2.5 and its interaction with the urbanization rate based on long-term data are 451 important topics for future research. 452 453 454 455 Acknowledgments 456 The authors express their sincere thanks for the support from the National 457 Natural Science Foundation of China under Grant No. 71173200, the Development

and Research Center of China Geological Survey under Grant No. 12120114056601
and No. 12120113093200, the National Science and Technology Major Project under

Grant No. 2016ZX05016005-003 and the Fundamental Research Funds for the

461 Central Universities under Grant No. 53200859633.

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464 Appendix A

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Table A1 A Nomenclature List

Full name

Abbreviation

Data Envelopment Analysis	DEA
Beijing-Tianjin-Hebei region	BTH region
Environmental Kuznets Curve	EKC
Air Quality Index	AQI
Gross Domestic Product	GDP
Inverse Distance Weighted method	IDW method
Ordinary Kriging method	OK method
High-High agglomeration	HH agglomeration
Low-High aggregation	LH agglomeration
Low-Low aggregation	LL agglomeration
High-Low aggregation	HL agglomeration
Local Indicators of Spatial Association	LISA

469		Table	e A2 PM2	2.5 concer	ntrations in	n 13 cities	in the BT	TH region	by month	1		
City	2016.01	2016.02	2016.03	2016.04	2016.05	2016.06	2016.07	2016.08	2016.09	2016.10	2016.11	2016.12
Beijing	68.15	43.48	92.67	67.54	53.85	59.28	68.6	46.6	54.63	84.73	99.3	130.55
Tianjin	73.52	50.29	80.84	63.35	50.65	53.32	52.97	40.75	52.47	64.07	104.49	134.9
Shijiazhuang	129.63	71.61	83.16	60.33	54.15	44.91	65.72	25.18	83.02	116.4	169.03	276.06
Qinhuangdao	40.2	38.09	63.57	39.36	34.73	42.53	39.75	27.62	36.49	39.65	64.83	84.3
Xingtai	129.09	81.34	88.2	66.1	50.64	54.17	65.86	38.95	67.35	82.23	124.17	188.04
Handan	108.58	72.3	75.09	78.67	41.76	48.75	54.73	41.23	56.12	50.87	108.07	231.65
Baoding	148.28	83.37	76.78	68.23	65.37	54.46	70.95	37.85	69.35	96.25	142.13	189.99
Chengde	44.79	40.86	53.25	37.63	30.07	27.56	37.13	25.52	27.59	41.66	51.07	56.73
Langfang	88.45	50.74	73.08	46.09	40.07	48.9	56.38	43.08	41.07	55.29	89.25	151.21
Zhangjiakou	27.52	30.14	36.49	27.45	26.11	22.87	39.78	24	22.08	37.59	44.8	39.25
Hengshui	133.78	91.38	95.28	72.41	55.52	58.11	83.57	56.59	47.27	75.19	114.34	159.27
Cangzhou	84.17	56.59	66.63	57.25	45.36	46.79	56.25	41.13	43.93	72.87	102.64	142.25

Tangshan	78.32	58.92	84.6	61.53	50.07	69.52	55.2	34.38	64.04	72.96	108.33	147.34
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471 **References**

Alves C, Vicente A, Pio C, et al. Organic compounds in aerosols from selected
European sites–Biogenic versus, anthropogenic sources[J]. Atmospheric Environment,
2012, 59(59):243-255.

- Bates D V, Sizto R. Air pollution and hospital admissions in Southern Ontario:
 the acid summer haze effect[J]. Environmental research, 1987, 43(2): 317-331.
- 477 Cai S, Wang Y, Zhao B, et al. The impact of the "Air Pollution Prevention and
 478 Control Action Plan" on PM2.5 concentrations in Jing-Jin-Ji region during 2012479 2020[J]. Science of the Total Environment, 2017, 580:197-209.
- 480 Chalbot M C G, Jones T A, Kavouras I G. Trends of non-accidental,
 481 cardiovascular, stroke and lung cancer mortality in Arkansas are associated with
 482 ambient PM2.5 reductions[J].
- 483 Civan M Y, Elbir T, Seyfioglu R, et al. Spatial and temporal variations in
 484 atmospheric VOCs, NO2, SO2, and O3 concentrations at a heavily industrialized
 485 region in Western Turkey, and assessment of the carcinogenic risk levels of
 486 benzene[J]. Atmospheric Environment, 2015, 103: 102-113.
- 487 Dockery D W, Pope C A, Xu X, et al. An association between air pollution and
 488 mortality in six US cities[J]. New England journal of medicine, 1993, 329(24): 1753489 1759.
- Guan D, Su X, Zhang Q, et al. The socioeconomic drivers of China's primary
 PM2.5 emissions[J]. Environmental Research Letters, 2014, 9(2): 024010.
- Hao Y, Liu Y M. The influential factors of urban PM 2.5 concentrations in China:
 a spatial econometric analysis[J]. Journal of Cleaner Production, 2016, 112: 14431453.
- Hsu C Y, Chiang H C, Chen M J, et al. Ambient PM2.5 in the residential area
 near industrial complexes: Spatiotemporal variation, source apportionment, and health
 impact.[J]. Science of the Total Environment, 2017.
- Hu W, Downward G S, Reiss B, et al. Personal and indoor PM2.5 exposure from
 burning solid fuels in vented and unvented stoves in a rural region of China with a

500 high incidence of lung cancer[J]. Environmental science & technology, 2014, 48(15): 8456-8464. 501 Hussain S Q, Ahn S H, Park H, et al. Light trapping scheme of ICP-RIE glass 502 texturing by SF 6/Ar plasma for high haze ratio[J]. Vacuum, 2013, 94: 87-91. 503 504 Jansen R C, Shi Y, Chen J, et al. Using hourly measurements to explore the role of secondary inorganic aerosol in PM2.5 during haze and fog in Hangzhou, China[J]. 505 Advances in Atmospheric Sciences, 2014, 31(6): 1427-1434. 506 507 Lee S J, Serre M L, van Donkelaar A, et al. Comparison of geostatistical 508 interpolation and remote sensing techniques for estimating long-term exposure to ambient PM2.5 concentrations across the continental United States.[J]. Environmental 509 health perspectives, 2012, 120(12): 1727. 510 511 Li L, Yang J, Song Y F, et al. The burden of COPD mortality due to ambient air pollution in Guangzhou, China[J]. Scientific reports, 2016, 6. 512 513 Liao T, Wang S, Ai J, et al. Heavy pollution episodes, transport pathways and potential sources of PM2.5 during the winter of 2013 in Chengdu (China).[J]. Science 514 515 of the Total Environment, 2017, 584-585:1056. Liu C, Henderson B H, Wang D, et al. A land use regression application into 516 517 assessing spatial variation of intra-urban fine particulate matter (PM 2.5) and nitrogen dioxide (NO2) concentrations in City of Shanghai, China[J]. Science of the Total 518 Environment, 2016, 565:607-615. 519 520 Liu Q, Wang Q. Sources and flows of China's virtual SO2, emission transfers embodied in interprovincial trade: A multiregional input-output analysis[J]. Journal 521 of Cleaner Production, 2017, 161. 522 523 Liu Z, Cheng K, Li H, et al. Exploring the potential relationship between indoor air quality and the concentration of airborne culturable fungi: a combined 524 experimental and neural network modeling study[J]. Environ Sci Pollut Res Int, 525 2017b(6):1-8. 526 Liu Z, Hu B, Wang L, et al. Seasonal and diurnal variation in particulate matter 527 (PM 10, and PM 2.5) at an urban site of Beijing: analyses from a 9-year study[J]. 528 Environmental Science and Pollution Research, 2015a, 22(1):627-642. 529

530	Liu Z, Li H, Cao G. Quick estimation model for the concentration of indoor
531	airborne culturable bacteria: an application of machine learning[J]. International
532	Journal of Environmental Research and Public Health, 2017a, 14(8): 857.
533	Liu Z, Zhu Z, Zhu Y, et al. Investigation of dust loading and culturable
534	microorganisms of HVAC systems in 24 office buildings in Beijing[J]. Energy and
535	Buildings, 2015b, 103: 166-174.
536	Luo J, Du P, Samat A, et al. Spatiotemporal Pattern of PM2.5 Concentrations in
537	Mainland China and Analysis of Its Influencing Factors using Geographically
538	Weighted Regression[J]. Scientific Reports, 2017, 7.
539	Ma M, Shen L, Ren H, et al. How to Measure Carbon Emission Reduction in
540	China's Public Building Sector: Retrospective Decomposition Analysis Based on
541	STIRPAT Model in 2000–2015[J]. Sustainability, 2017c, 9(10): 1744.
542	Ma M, Yan R, Cai W. An extended STIRPAT model-based methodology for
543	evaluating the driving forces affecting carbon emissions in existing public building
544	sector: evidence from China in 2000–2015[J]. Natural Hazards, 2017b, 89(2): 741-
545	756.
546	Ma M, Yan R, Du Y, et al. A methodology to assess China's building energy
547	savings at the national level: An IPAT-LMDI model approach[J]. Journal of cleaner
548	production, 2017a, 143: 784-793.
549	Moroń W, Rybak W. NOx and SO2 emissions of coals, biomass and their blends
550	under different oxy-fuel atmospheres[J]. Atmospheric Environment, 2015, 116: 65-71.
551	National Bureau of Statistics of the People's Republic of China. Hebei
552	Economic Yearbook 2014[M]; China Statistical Press:Beijing, China, 2014.
553	National Bureau of Statistics of the People's Republic of China. Beijing
554	statistical yearbook 2014[M]; China Statistical Press:Beijing, China, 2014.
555	National Bureau of Statistics of the People's Republic of China. Tianjin
556	statistical yearbook 2014[M]; China Statistical Press:Beijing, China, 2014.
557	Shuai C, Shen L, Jiao L, et al. Identifying key impact factors on carbon emission:
558	evidences from panel and time-series data of 125 countries from 1990 to 2011[J].
559	Applied Energy, 2017, 187: 310-325.

560 561	Su B, Thomson E. China's carbon emissions embodied in (normal and processing) exports and their driving forces, 2006–2012[J]. Energy Economics, 2016, 59: 414-422.
562 563	Tobler W R. A computer movie simulating urban growth in the Detroit region[J]. Economic geography, 1970, 46(sup1): 234-240.
564 565 566	Wang X, Ding X, Fu X, et al. Aerosol scattering coefficients and major chemical compositions of fine particles observed at a rural site in the central Pearl River Delta, South China[J]. Journal of Environmental Sciences, 2012, 24(1): 72-77.
567 568 569	Wang Y S, Yao L, Wang L L, et al. Mechanism for the formation of the January 2013 heavy haze pollution episode over central and eastern China[J]. Science China Earth Sciences, 2014, 57(1): 14-25.
570 571	Wang Z, Fang C, Guang X U, et al. Spatial-temporal characteristics of the PM_(2.5) in China in 2014[J]. Acta Geographica Sinica, 2015 (in Chinese).
572 573 574	Wu J, Li J, Peng J, et al. Applying land use regression model to estimate spatial variation of PM2.5 in Beijing, China[J]. Environmental Science and Pollution Research, 2015, 22(9): 7045-7061.
575 576	Wu X, Chen Y, Guo J, et al. Spatial concentration, impact factors and prevention-control measures of PM2.5 pollution in China[J]. Natural Hazards, 1-18.
577 578 579	Xu B, Lin B. Regional differences of pollution emissions in China: contributing factors and mitigation strategies[J]. Journal of Cleaner Production, 2015, 112(4):1454-1463.
580 581 582	Yan D, Lei Y, Li L, et al. Carbon emission efficiency and spatial clustering analyses in China's thermal power industry: Evidence from the provincial level[J]. Journal of Cleaner Production, 2017.
583 584 585	Yan R, Ma M, Pan T. Estimating energy savings in Chinese residential buildings from 2001 to 2015: A decomposition analysis[J]. Journal of Engineering Science & Technology Review, 2017, 10(1).
586 587	Zhang A, Qi Q, Jiang L, et al. Population exposure to PM 2.5 in the urban area of Beijing[J]. PloS one, 2013, 8(5): e63486.
588 589 590	Zhang Y J, Hao J F, Song J. The CO2 emission efficiency, reduction potential and spatial clustering in China's industry: evidence from the regional level[J]. Applied Energy, 2016, 174: 213-223.

Zhao B, Wang P, Ma J Z, et al. A high-resolution emission inventory of primary
pollutants for the Huabei region, China[J]. Atmospheric Chemistry & Physics, 2012,
11(1):20331-20374.

Zhao X, Wang X, Ding X, et al. Compositions and sources of organic acids in
fine particles (PM2.5) over the Pearl River Delta region, south China[J]. Journal of
Environmental Sciences, 2014, 26(1): 110-121.

Zhao Y, Liu Y, Ma J, et al. Heterogeneous reaction of SO2 with soot: The roles
of relative humidity and surface composition of soot in surface sulfate formation[J].
Atmospheric Environment, 2017, 152: 465-476.

600Zou B, Wang M, Wan N, et al. Spatial modeling of PM 2.5, concentrations with601a multifactoral radial basis function neural network[J]. Environmental Science and

602 Pollution Research, 2015, 22(14):10395-404.