PM$_{2.5}$ Spatiotemporal Variations and the Relationship with Meteorological Factors during 2013-2014 in Beijing, China

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Abstract

Objective

Limited information is available regarding spatiotemporal variations of particles with median aerodynamic diameter < 2.5 μm (PM$_{2.5}$) at high resolutions, and their relationships with meteorological factors in Beijing, China. This study aimed to detect spatiotemporal change patterns of PM$_{2.5}$ from August 2013 to July 2014 in Beijing, and to assess the relationship between PM$_{2.5}$ and meteorological factors.

Methods

Daily and hourly PM$_{2.5}$ data from the Beijing Environmental Protection Bureau (BJEPB) were analyzed separately. Ordinary kriging (OK) interpolation, time-series graphs, Spearman correlation coefficient and coefficient of divergence (COD) were used to describe the spatiotemporal variations of PM$_{2.5}$. The Kruskal-Wallis H test, Bonferroni correction, and Mann-Whitney U test were used to assess differences in PM$_{2.5}$ levels associated with spatial and temporal factors including season, region, daytime and day of week. Relationships between daily PM$_{2.5}$ and meteorological variables were analyzed using the generalized additive mixed model (GAMM).

Results

Annual mean and median of PM$_{2.5}$ concentrations were 88.07 μg/m^3 and 71.00 μg/m^3, respectively, from August 2013 to July 2014. PM$_{2.5}$ concentration was significantly higher in winter ($P < 0.0083$) and in the southern part of the city ($P < 0.0167$). Day to day variation of PM$_{2.5}$ showed a long-term trend of fluctuations, with 2–6 peaks each month. PM$_{2.5}$ concentration was significantly higher in the night than day ($P < 0.0167$). Meteorological factors

were associated with daily PM$_{2.5}$ concentration using the GAMM model (R^2 = 0.59, AIC = 7373.84).

Conclusion

PM$_{2.5}$ pollution in Beijing shows strong spatiotemporal variations. Meteorological factors influence the PM$_{2.5}$ concentration with certain patterns. Generally, prior day wind speed, sunlight hours and precipitation are negatively correlated with PM$_{2.5}$, whereas relative humidity and air pressure three days earlier are positively correlated with PM$_{2.5}$.

Introduction

Ambient air pollutants, especially particulate matter (PM), have attracted attention in recent years because their associated adverse health effects [1–8]. It has been established that long- and short-term exposure to PM, including particles with a median aerodynamic diameter < 2.5 μm (PM$_{2.5}$) and < 10 μm (PM$_{10}$), elevates the risk of cardiovascular and respiratory diseases and excess mortality [1–3]. Research suggests that PM$_{2.5}$ is very toxic and more harmful to human health than coarse particles (particles with a median aerodynamic diameter > 2.5 μm). When inhaled, PM$_{2.5}$ enters the bloodstream and translocated to vital organs including the liver, spleen, heart and the brain [9]. Adverse health outcomes from PM$_{2.5}$ inhalation include, among others: impaired pulmonary function, increased blood pressure, and cognitive deficit [4–6]. PM$_{2.5}$ can also lead to stroke, lung cancer, and some other illnesses [7, 8].

China has experienced rapid urbanization and industrialization, which has resulted in a dramatic increase in energy consumption and emission over the past several decades [10]. One of the environmental challenges is the frequent nationwide episodes of haze-fog. A recent study reported that the annual average concentration of PM$_{2.5}$ for almost all provincial capital cities in China exceeded 35 μg/m^3 during 2013–2014 [11, 12]. It appears that the threat is more serious in the capital city, Beijing, China, in part due to its large population size, increase number of vehicles and numerous active construction activities. For example, during 2004–2008, daily mean PM$_{2.5}$ concentration was 105 μg/m^3, and the latest study revealed that citywide cumulative number of exceedance days is generally high [13, 14]. The extremely high concentrations of PM$_{2.5}$ can lead to various negative health outcomes, several studies have shown that PM$_{2.5}$ has significant effects on cardiovascular and respiratory emergency room visits, as well as years of life lost in Beijing [13, 15, 16].

Considering the multiple deleterious health effects of PM$_{2.5}$, data with high spatial and temporal resolution are needed to accurately evaluate the status and health risks associated with PM$_{2.5}$ exposure. However, access to pre-existing PM$_{2.5}$ data from the Beijing Environmental Protection Bureau (BJEPB) has not possible since most of the PM$_{2.5}$ data for the previous years were not documented. It was until October 2012 that the hourly monitoring data of PM$_{2.5}$ was released. This data was sampled from 35 sites which is a representative of the whole city. Although the spatiotemporal distribution of PM$_{2.5}$ using these data was reported in one study, continuous concentrations of PM$_{2.5}$ at high temporal resolution were unavailable [14]. Other investigators reported long-term variation of PM$_{2.5}$, but their results were generally based on discrete points or indirect estimation [17, 18]. Several studies have explored the relationship between meteorological factors and PM$_{2.5}$ in Beijing and found that meteorological factors may be important in PM$_{2.5}$ variation. However, only few of these studies have examined the

correlation between wind speed and relative humidity and PM$_{2.5}$. Additionally, most of these studies have not fully explored the impact of various meteorological variables on PM$_{2.5}$ [19–21].

The purpose of the present study is to examine the spatiotemporal variations of PM$_{2.5}$ in Beijing, using officially released data from 35 stations during a one-year period from August 2013 to July 2014, and to assess the relationships between daily PM$_{2.5}$ and meteorological factors.

Methods

Source of PM$_{2.5}$ and meteorological factors

Since the end of September 2012, daily average and hourly real-time ambient air pollutant data have been gradually released to the public by the BJEPB, based on the 35 automatic monitoring stations established in the 16 districts of Beijing city (Fig 1). Daily average (August 2013 through July 2014) and hourly real-time (December 2013 through November 2014) of PM$_{2.5}$ concentration data were collected from the Centre of the City Environmental Protection Monitoring Website Platform, BJEPB (www.bjmemc.com.cn). In addition, meteorological data including daily mean wind speed (m/s), relative humidity (%), sunlight hours (h), temperature (°C), precipitation (mm) and air pressure (kPa) in the 16 districts were obtained from the Chinese Meteorological Bureau over the same period.

Daily mean concentrations for each district and the whole city were calculated by averaging concentrations reported by all 35 stations, which is the same method used by BJEPD to report daily concentration of air pollutants to the public. Rates of missing values in the 16 districts were mostly low ranging from 7.12% to 8.77%, except for *Mentougou* and *Huairou* which had higher levels of 17.81% and 10.41%, respectively. Some daily data were missing for all the districts mainly due to the technical problem on website maintenance during the study period. A Markov chain Monte Carlo (MCMC) multiple imputation method was used to impute missing values, and data from 339 days were available for analyses.

Spatiotemporal analysis of PM$_{2.5}$

To provide a more comprehensive picture of the current status and spatiotemporal variations of PM$_{2.5}$ pollution, daily and hourly concentration data were analyzed by different methods. Using the Chinese ambient air quality standards (CAAQS) as a reference, daily average PM$_{2.5}$ that exceeded Grades I (35 μg/m^3) and II (75μg/m^3) were selected.

Ordinary kriging (OK) interpolation [22, 23] was used to characterize PM$_{2.5}$ regional and seasonal variations, based on concentration data from the 35 monitoring stations. PM$_{2.5}$ summary statistics, space-time dependence functions and PM$_{2.5}$ estimates on a space-time grid were obtained to describe regional and seasonal variations. This was done using the Geostatistical Analyst Extension of ArcGIS (ArcMap, version 10.2.2). To explore PM$_{2.5}$ regional and seasonal variations, 16 districts were assigned to three areas: southern, northern and central (Table 1). Furthermore, 12 months were stratified into four seasons, spring (March, April and May), summer (June, July and August), autumn (September, October and November) and winter (December, January and February).

In addition, day to day variation of PM$_{2.5}$ citywide during the year was displayed as a time-series figure. The number of PM$_{2.5}$ pollution episodes (periods with concentrations > 75 μg/m^3), episode duration, and interval between two episodes were calculated. Diurnal variations of PM$_{2.5}$ in each month were developed into time-series figures by averaging the concentrations at various time points.
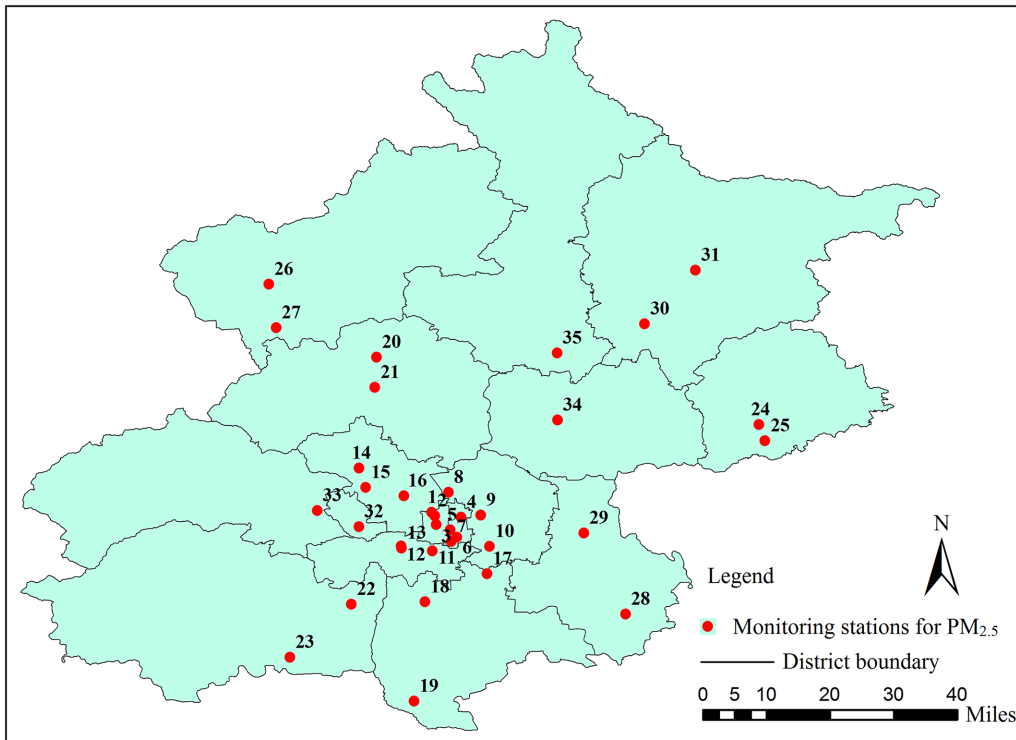
code	Station name	code	Station name
1	XiZhiMenBeiDaJie	19	JingXiNanLiuLiHe
2	XiChengGuanYuan	20	MenTouGouLongQuanZhen
3	XiChengWanShouXiGong	21	YanQingZhen
4	DongChengDongSi	22	JingXiBeiBaDaLing
5	QianMenDongDaJie	23	ChangPingDingLing
6	DongChengTianTan	24	ChangPingZhen
7	YongDingMenNeiDaJie	25	JingDongNanYongLeDian
8	ChaoYangAoTiZhongXin	26	TongZhouXinCheng
9	ChaoYangNongZhanGuan	27	YiZhuangKaiFaQu
10	DongSiHuanBeiLu	28	DaXingHuangCunZhen
11	NanSanHuanXiLu	29	JingNanYuFa
12	FengTaiHuaYuan	30	MiYunZhen
13	FengTaiYunGang	31	JingDongBeiMiYunShuiKu
14	HaiDianBeiBuXinQu	32	PingGuZhen
15	HaiDianBeiJingZhiWuYuan	33	JingDongDongGaoCun
16	HaiDianWanLiu	34	ShunYiXinCheng
17	ShiJingShanGuCheng	35	HuaiRouZhen
18	FangShanLiangXiang		

Note: ArcGIS (ArcMap, version 10.2.2) was used to create the map

Fig 1. Locations of the 35 PM2.5 monitoring stations in Beijing.

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Table 1. Distribution of PM$_{2.5}$ concentrations in the 16 districts of Beijing, 2013–2014.

District	Regional category	Mean(SD) µg/m³	Median (IQR) µg/m³	Range µg/m³	Non-attainment days and rates (%) for grade I	Non-attainment days and rates (%) for grade II
Yanqing	North	67.95(61.06)	52.00(67.00)	5.0–459.0	216 (63.7)	108 (31.9)
Changping	North	73.30(66.36)	55.00(72.00)	5.0–432.0	224 (66.1)	130 (38.3)
Miyun	North	67.79(61.74)	51.00(68.00)	4.0–481.0	205 (60.5)	117 (34.5)
Huairou	North	77.80(65.80)	59.00(72.00)	5.0–402.0	226 (69.1)	136 (41.6)
Pinggu	North	79.23(66.25)	61.00(74.50)	4.0–475.0	242 (71.4)	150 (44.2)
Shunyi	North	83.55(70.27)	65.00(80.00)	5.0–509.0	241 (71.5)	149 (44.2)
Haidian	Center	85.15(70.47)	71.00(77.00)	4.0–430.0	248 (73.2)	161 (47.5)
Shijingshan	Center	87.99(69.32)	72.00(73.00)	6.0–408.0	254 (76.3)	158 (47.4)
Xicheng	Center	88.26(71.60)	70.50(78.00)	6.0–449.0	253 (74.6)	161 (47.5)
Chaoyang	Center	89.24(71.33)	71.50(81.00)	5.0–464.0	262 (77.3)	165 (48.7)
Dongcheng	Center	91.56(72.97)	75.00(81.00)	3.0–457.0	266 (78.5)	168 (49.6)
Fengtai	Center	96.50(76.59)	79.00(82.00)	6.0–511.0	268 (79.3)	184 (54.4)
Mentougou	South	77.41(64.88)	65.50(66.75)	5.0–403.0	211 (70.3)	124 (41.3)
Fangshan	South	106.97 (79.33)	89.00(91.00)	6.0–492.0	285 (84.3)	194 (57.4)
Daxing	South	106.20 (83.77)	86.00(93.50)	7.0–493.0	280 (82.6)	192 (56.6)
Tongzhou	South	107.63 (85.87)	88.00(92.00)	4.0–537.0	287 (84.9)	193 (57.1)

SD: standard deviation; IQR: inter-quartile range.

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To assess PM$_{2.5}$ spatial heterogeneity, Spearman correlation coefficients and coefficients of divergence (COD) were calculated for each monitoring station pair, and compared with the distance between the stations [24, 25]. A low COD value indicates small differences between stations PM$_{2.5}$ concentrations, while a value close to 1 signifies greater disparity between concentrations.

Kruskal-Wallis H and Bonferroni correction tests were used to assess differences in PM$_{2.5}$ levels associated with spatial and temporal factors, including season, area, and daytime. Week-day/weekend differences were tested by Mann-Whitney U test. All statistical tests were two-sided, and *P*-values less than 0.05 were considered statistically significant.

Modeling association between PM$_{2.5}$ and meteorological factors

Because scatter plots showed that not all meteorological variables were linearly correlated with PM$_{2.5}$, a generalized additive mixed model (GAMM) was used to explore the effects of meteorological factors on daily PM$_{2.5}$ concentrations. This model can use both additive parametric terms and nonparametric function to formulate covariate effects and add random effects to the additive predictor, accounting for over dispersion and correlation [26, 27]. District-level daily PM$_{2.5}$ concentration data were used as the dependent variable, and corresponding district-level meteorological factors were used as independent variables. Lagged (1–3 days earlier) effects of meteorological factors were checked, because the prior weather conditions may influence the subsequent concentrations of PM$_{2.5}$ [28]. Meteorological variables that had the strongest correlation with PM$_{2.5}$ from lag0 (current value) to lag3 (value 3 days earlier) with Spearman correlation coefficient $r_s > 0.2$ were entered in the final model. The Akaike Information Criteria (AIC) and adjusted R^2 were used to select the appropriate variables and models.

The conditional probability distribution of PM$_{2.5}$ concentrations approximately followed a Gamma distribution according to QQ plot and was tested by one-sample Kolmogorov-Smirnov test, so a logarithm-linked function for PM$_{2.5}$ concentration was used in the GAMM model. Cubic splines were used as the nonparametric function of the covariates, which were potentially not linearly correlated to log-transformed PM$_{2.5}$ [29]. Day of the year was introduced to control temporal effects on PM$_{2.5}$ concentration. An automatic choice was adopted to determine the most appropriate parameters (degrees of freedom, knots) for the splines, based on generalized cross-validation (GCV). In addition, since PM$_{2.5}$ concentration depends linearly on its own previous values and on a stochastic term, an autoregressive structure $ARMA(p,q)$ was introduced in the model to describe the regression [30]. Optimal values of p and q were determined by AIC and autocorrelation function (ACF). The initial model is

$$\log(E(Y_{i,t})) = \alpha + s_1(Day_i) + s_2(WS_{i,t}) + s_3(RH_{i,t}) + s_4(T_{i,t}) + s_5(SH_{i,t}) + \lambda DOW(P_{i,t})$$
$$+ \beta AP_{i,t} + \mu Z_i + \tau_t$$

Where $Y_{i,t}$ is the concentration of PM$_{2.5}$ in district i (i = 1 to 16) on day t (t = 1 to 339). Each s represents a cubic splines smoothing function for meteorological factors including wind speed (WS), relative humidity (RH), temperature (T) and sunlight hours (SH), which exhibit non-linear relationships with log-transformed daily PM$_{2.5}$ concentration. $s(Day_i)$ was used to control for temporal trend. Since precipitation (P) followed an extreme skewed distribution and air pressure (AP) was linearly correlated with PM$_{2.5}$, a dichotomous form of precipitation and linear term of air pressure were introduced in the model. Z_i is a random intercept for district i and τ_t is the autoregression term. All analyses were conducted using statistical software R (version 3.1.2), and package "mgcv" was used for the GAMM modeling. All statistical tests were two-sided, and P-values less than 0.05 were considered statistically significant.

Results

Overview of PM$_{2.5}$ pollution in Beijing

Annual mean PM$_{2.5}$ concentrations ranged from 67.79 μg/m^3 in district *Miyun* to 107.63 μg/m^3 in district *Tongzhou*, greatly exceeding the yearly CAAQS (GB3095-2012) Grade I (15 μg/m^3) and II standards (35 μg/m^3) for all districts in Beijing ([Table 1](#)). The citywide mean concentration of 88.07 μg/m^3 also exceeded the standards. [Table 1](#) lists the number of non-attainment days (defined as days with PM$_{2.5}$ concentration exceeding standards) and rates for the 16 districts based on the daily CAAQS (GB3095-2012) standards. All the 16 districts experienced PM$_{2.5}$ pollution that exceeded daily Grade I (35 μg/m^3) standard during more than 60% of days (a non-attainment rate of 60%) and Grade II (75 ug/m^3) standard during over 30% of days (a non-attainment rate of 30%) of the year.

Spatiotemporal variations of PM$_{2.5}$ pollution

Bonferroni test was used to assess seasonal and regional differences in PM$_{2.5}$ levels, and the mean difference was significant at the 0.0083 and 0.0167 levels, respectively ([Table 2](#)). PM$_{2.5}$ pollution in Beijing had pronounced seasonal and regional variations ([Fig 2](#)). It was significantly higher in winter ($P < 0.0083$) and lower in summer ($P < 0.0083$). There was no statistically significant difference in PM$_{2.5}$ concentration between spring and autumn ($MD = -5.615$, $P = 0.024$). PM$_{2.5}$ concentration in the southern part of the city was significantly higher than the northern area ($MD = 29.492$, $P < 0.0167$). Observed PM$_{2.5}$ levels also revealed a pronounced spatial gradient, increasing from north to south in most months, except in July 2014.

Table 2. Significance tests of PM$_{2.5}$ levels for different season, region, daytime and day of week.

Variable		Kruskal-Wallis H test		Bonferroni test	
		χ^2	P	MD	P
Season*	Spring vs. Summer	367.720	0.000	9.945	0.000
	Spring vs. Autumn			-5.615	0.024
	Spring vs. Winter			-38.140	0.000
	Summer vs. Autumn			-15.560	0.000
	Summer vs. Winter			-48.085	0.000
	Autumn vs. Winter			-32.525	0.000
Region #	South vs. North	304.553	0.000	29.492	0.000
	North vs. Center			-16.254	0.000
	Center vs. South			-13.238	0.000
Daytime#	7 pm-6 am vs. 7 am-12 am	69.991	0.000	12.839	0.001
	7 pm-6 am vs. 1 pm-6 pm			7.855	0.000
	7 am-12 am vs. 1 pm-6 pm			4.985	0.135
				Mann-Whitney U test	
				Z	P
Day of week	Weekdays vs. Weekends			-0.145	0.885

*: The mean difference is significant at the 0.0083 level for Bonerroni test.

#: The mean difference is significant at the 0.0167 level for Bonerroni test.

This pattern was more obvious in cold months (November 2013 to January 2014), with extremely high concentrations in the southern part of the Beijing city.

The day to day pattern of PM$_{2.5}$ concentrations from August 2013 to July 2014 showed a long-term trend of fluctuations (Fig 3). A total of 52 episodes of PM$_{2.5}$ pollution (> 75 μg/m^3) were observed during the year (13 in spring, 11 in summer, 13 in autumn and 16 in winter) with 2–6 episodes each month. An episode usually lasted 1–7 days, and intervals between episodes were 1–14 days (missing days were not included in the calculation). Mann-Whitney U test was used to assess weekday/weekend difference, but no statistically significant difference was found (Fig 3 and Table 2) ($Z = -0.145$, $P = 0.885$).

Hourly average PM$_{2.5}$ concentration in each month had a diurnal pattern represented by one or two peaks. For given days, if the PM$_{2.5}$ concentration increased from a value lower than the monthly mean to one higher than that mean, those days were regarded as a single peak until the concentration fell below the mean (Fig 4). Over 7 months (February to April, June to September), there were 2 peaks, 1 in the forenoon, and the other in the early night time. For the other 5 month (May, and October to January), the peak was either in the forenoon or early night time. The lowest PM$_{2.5}$ levels were in the afternoon, except during October. Bonferroni test was used to assess hourly difference in PM$_{2.5}$ levels, and the mean difference was significant at the 0.0167 level (Table 2). It shows that PM$_{2.5}$ concentration at night (7 pm through 6 am) was significantly higher than in the daytime (7 am through 12 am and 1pm through 6 pm) ($P < 0.0167$), but there were no statistically significant difference between forenoon (7 am through 12 am) and afternoon (1 pm through 6 pm) ($MD = 4.985$, $P = 0.136$).

The spatial heterogeneity of PM$_{2.5}$ was examined by calculating correlation coefficients and CODs for daily average concentrations at 595 station pairs. Mean values of the two coefficients for all station pairs were 0.912 and 0.195, respectively (Fig 5). Fig 5 shows that correlation coefficients declined with increasing distance between stations, whereas CODs increased with
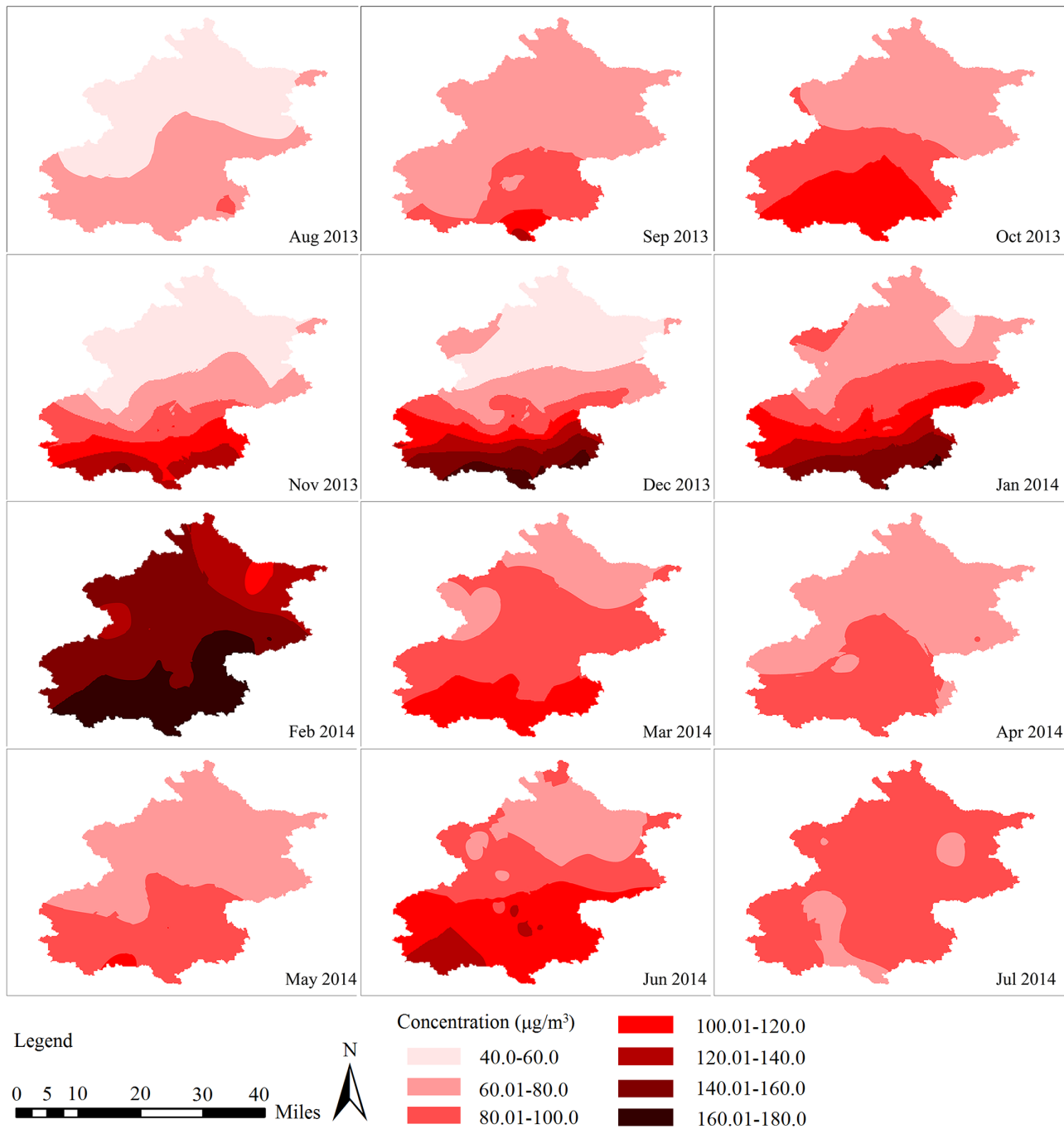
Fig 2. Regional and seasonal variations of PM2.5 in Beijing, 2013–2014.

increasing distance between stations. Slopes of both fit lines in Fig 5 were significantly different from zero ($P < 0.05$).

Association between PM2.5 and meteorological factors

Correlation analysis showed that prior day wind speed ($r_s = -0.48$, $P < 0.01$) and air pressure 3 days earlier ($r_s = 0.26$, $P < 0.01$) were highly correlated with the current PM2.5 concentrations
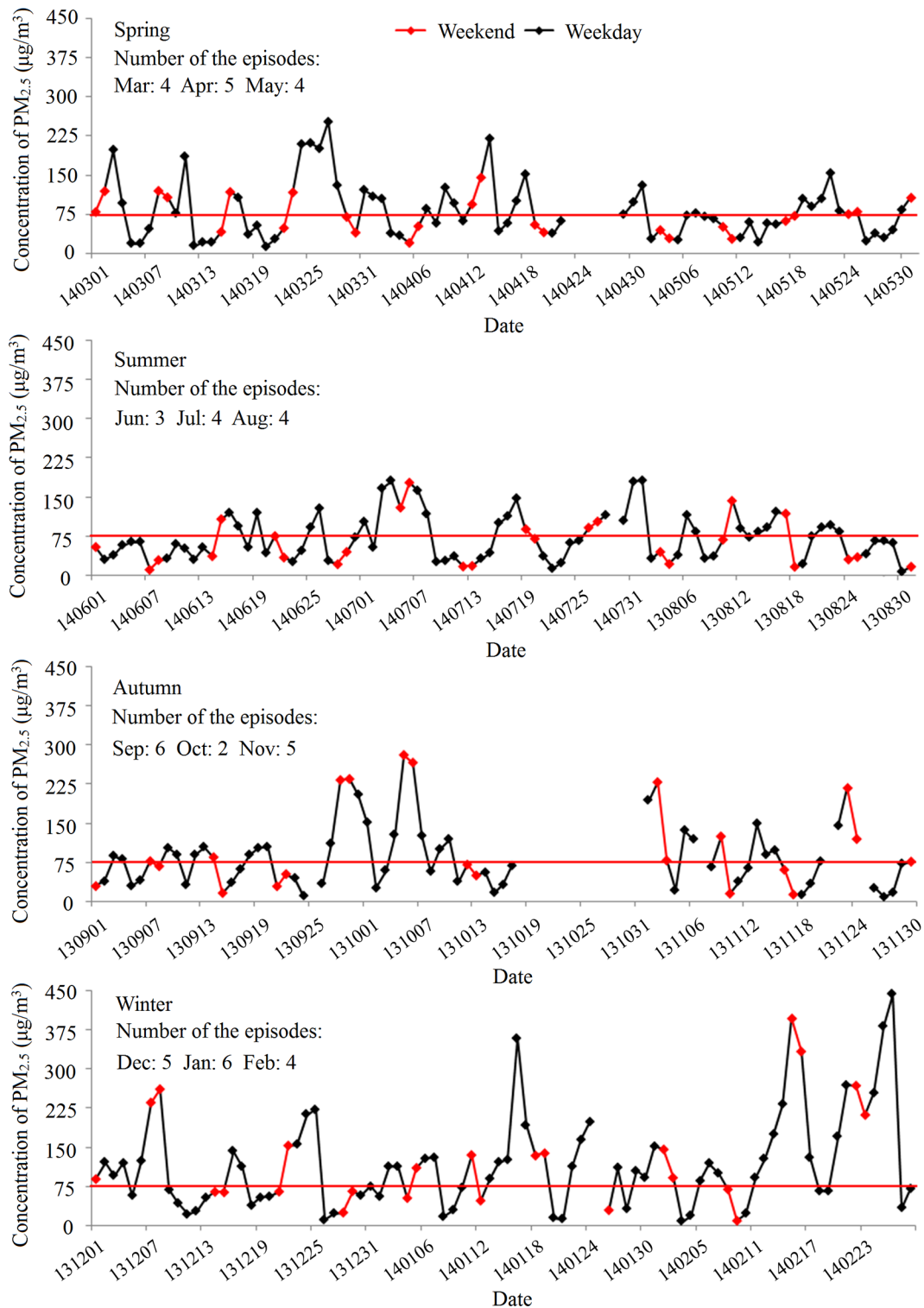
Fig 3. Day to day variations of PM$_{2.5}$ in different seasons, Beijing, 2013–2014.

Fig 4. Diurnal variations of PM2.5 in different months, Beijing, 2013–2014.

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For relative humidity ($r_s = 0.38$, $P < 0.01$) and sunlight hours ($r_s = -0.51$, $P < 0.05$), the strongest correlation was in the day of PM2.5 measurement. Because correlation coefficients of temperature (lag0 − lag3) were all < 0.2 at various daytimes, they were not included in the final model. For dichotomous variable precipitation, the model without a lagged term had the smallest AIC and largest adjusted R^2. Thus, meteorological variables including prior day wind speed (*WSlag1*), relative humidity (*RH*), sunlight hours (*SH*), precipitation (*P*) and air pressure 3 days earlier (*APlag3*) were entered in the final model. We selected the order of the autoregressive error term $p = 2$ and $q = 2$ with the smallest AIC, and the autocorrelation fall between [0.1,
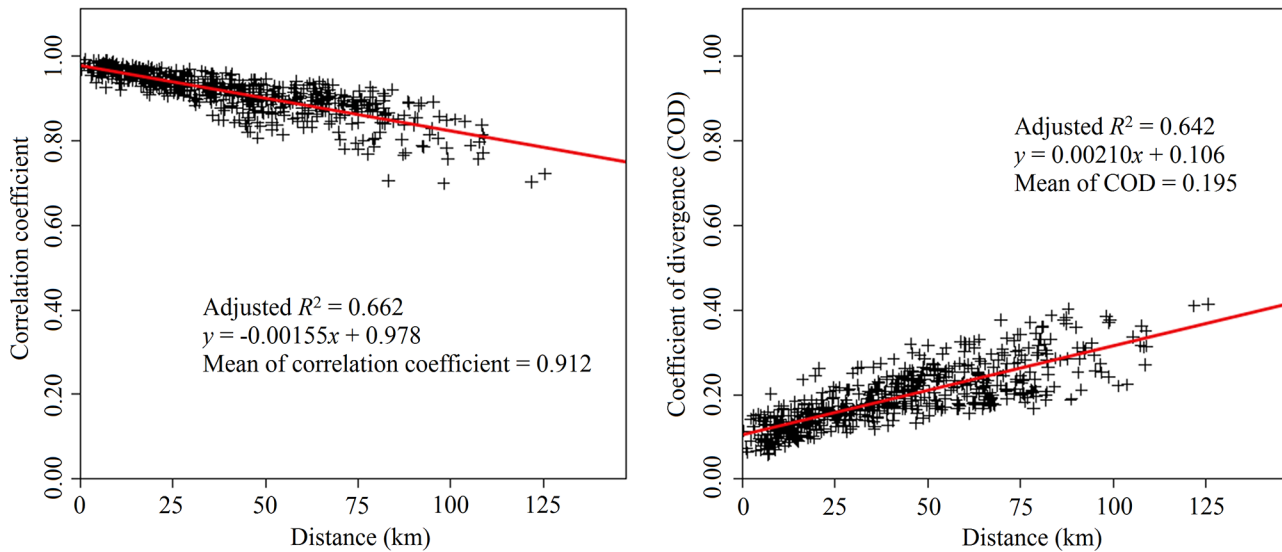
Fig 5. Correlation coefficient and COD versus distance between the stations.

0.1] from the ACF. The final model is

$$\log(E(Y_{i,t})) = \alpha + s_1(Day_i) + s_2(WSlag1_{i,t}) + s_3(RH_{i,t}) + s_4(SH_{i,t}) + \lambda DOW(P_{i,t}) + \beta APlag3_{i,t}$$
$$+ \mu Z_i + corARMA(2,2)$$

Overall effect size measured by the adjusted R^2 was 0.59 and goodness-of-fit assessed by the AIC was 7373.84 for the final GAMM model. The relationship between PM$_{2.5}$ and prior day wind speed was monotonically decreasing ([Fig 6]). Similarly, an overall downward tendency was found for PM$_{2.5}$ with increasing sunlight hours. On the contrary, PM$_{2.5}$ was positively correlated to relative humidity. For the dichotomous precipitation variable, PM$_{2.5}$ concentration was 85.68% (95% CI: 82.98%–88.47%) on days with precipitation, compared with those days of without precipitation. Air pressure had a 3-day lag effect on PM$_{2.5}$, which was positively correlated with log-transformed PM$_{2.5}$ concentration in linear from.

Discussion

The study shows that Beijing has serious PM$_{2.5}$ pollution citywide throughout the year [12–14]. We observed great spatial variations across the city [14, 31], with a pronounced increasing gradient from the north to the south. Southern Beijing is adjacent to seriously polluted cities in *Hebei* province and *Tianjin* [12, 32, 33]. Regional transportation may have a strong influence on southern suburbs, and aggravate PM$_{2.5}$ pollution. The northern part of Beijing is surrounded by mountains, and substantial green vegetation may be helpful to cleanse the air [34]. The lower population density in the northern suburbs, together with less human activity, may have contributed to the lower PM$_{2.5}$ concentration observed.

PM$_{2.5}$ concentration shows great seasonal variations, with the most severe PM$_{2.5}$ pollution in winter [12, 31]. Beijing has a northern temperate continental monsoon climate. The official residential heating season is from November to March. The elevated PM$_{2.5}$ level in winter is mainly from coal combustion and biomass burning for residential heating, as in the other northern cities in China [12, 32, 35]. Years ago, sandstorms were a serious problem, and usually reached Beijing in the spring. These storms involved long-range transport of desert dust, with
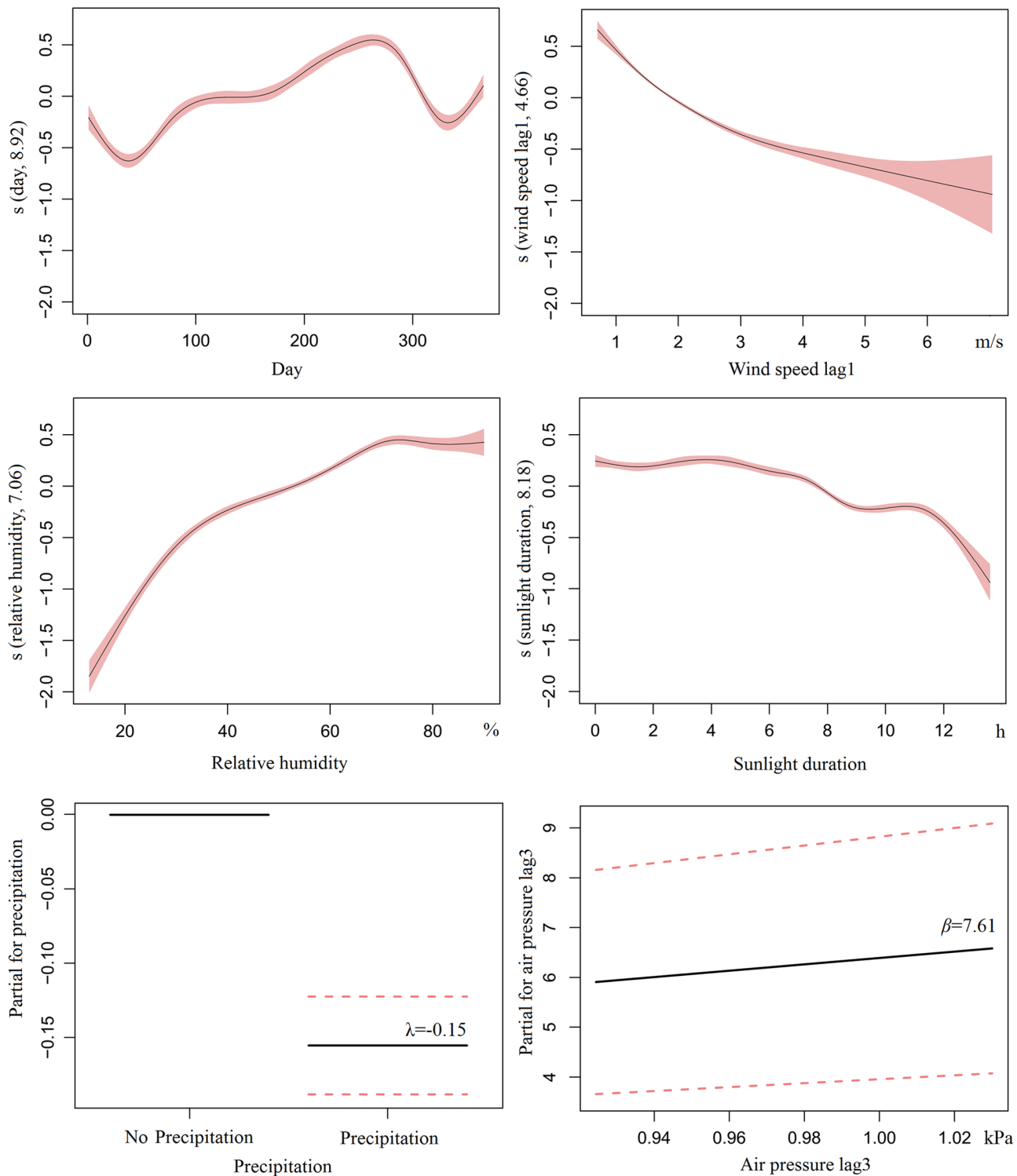
Fig 6. Exposure-response curves for PM$_{2.5}$ and meteorological variables, Beijing, 2013–2014.

mineral dust comprising 18.6% of PM$_{2.5}$ mass [16, 31]. However, there is no evidence indicating severe PM$_{2.5}$ pollution in spring. This may be as a result of the implemented Beijing and Tianjin Sandstorm Source Control Project which was set up in 2000 [36].

Regarding the weekly pattern, some studies found that air pollutant concentrations revealed a general weekend effect, with higher levels during the weekdays and lower ones during weekends [37, 38]. However, this pattern does not prevail in all cities, especially for PM$_{2.5}$ [39, 40]. Our results confirm no weekday/weekend difference for PM$_{2.5}$ concentrations in Beijing. Vehicle restrictions on weekdays may be an important explanation of this phenomenon. However, there were obvious periodic oscillations for PM$_{2.5}$, with 2–6 episodes each month. Because pronounced day to day variation of PM$_{2.5}$ may be less influenced by traffic intensity, which is uniform across weekdays and weekends, the concentration fluctuation may be largely caused by meteorological conditions such as solar radiation, formation of convectively mixed boundary layers, and wind [41, 42]. This is somehow also supported by a negative correlation between sunlight hours and PM$_{2.5}$ concentration, as well as a negative correlation between that concentration and wind speed (Fig 6). Furthermore, there are more episodes in winter and fewer in summer. Such variation is possibly due in part to seasonal variations of the air pollutant emission and the atmospheric boundary layer height.

Diurnal PM$_{2.5}$ variations were observed with one or two peaks in each month, similar to the results of other studies [39, 41, 43, 44] (Fig 5). The diurnal variations are dominated by the diurnal cycle of source emissions and the boundary layer height [44]. Generally, the forenoon peak is attributable to enhanced anthropogenic activity during morning rush hour, and decreasing PM$_{2.5}$ in the afternoon is mainly due to the developing boundary layer height, which provides a large volume for PM$_{2.5}$ dilution. Finally, a reduced boundary layer height with increased anthropogenic activity during the afternoon rush hour produces the early nighttime peak. In addition, the PM$_{2.5}$ diurnal variations vary by months. In the colder months (October to January), there are more coal combustion and biomass burning for residential heating, and boundary layer height generally decreases early in the afternoon because of less solar radiation, resulting in higher levels of PM$_{2.5}$ in early nighttime [45, 46].

PM$_{2.5}$ levels in Beijing were strongly correlated for all station pairs ($r_s > 0.70$), and distance was a powerful predictor of correlation [24]. However, 43.03% of COD values calculated for station-pairs' daily average concentrations of PM$_{2.5}$ were > 0.20, and those values were positively associated with distance, giving an approximate indication of spatial heterogeneity [47, 48]. This finding suggests that despite strong correlation among the stations, averaging PM$_{2.5}$ concentrations at multiple monitoring stations in Beijing may produce misclassification errors in epidemiological research (e.g., time-series epidemiologic studies evaluating relationships between PM$_{2.5}$ and health events).

Although the influence of meteorological conditions on the diffusion, dilution and accumulation of air pollutants has been widely recognized, it remains inconsistent when considering specific meteorological effects on PM$_{2.5}$ concentration. Previous studies have developed various meteorological predictive models for PM$_{2.5}$, with greater predictive powers judged by adjusted R^2 (0.79) or cross-variation R^2 (0.77) [49, 50]. Although model performance remains strong, the predictive ability of our model for PM$_{2.5}$ (adjusted $R^2 = 0.59$) was somewhat lower. The difference may be attributed to the use of additional selection, such as land use information [49, 50]. The reason why it cannot be explained fully by meteorological factors may be the complex and diverse human activities related to PM$_{2.5}$.

Among meteorological factors, most studies focused on wind speed, indicating that wind speed is negatively correlated with PM$_{2.5}$ [20, 21, 50–53], and this was also evident in this study. The lag effect of wind speed has also been considered in our study, and the result suggests that PM$_{2.5}$ is affected principally by prior day wind speed. For precipitation, our study is

also comparable to the other studies [50, 54]. Fig 6 shows that PM$_{2.5}$ concentration is nearly 10% lower on days with precipitation, compared with those days of no precipitation, owing to the fact that precipitation has a scavenging effect on air pollutants [54, 55].

The results about relative humidity on PM$_{2.5}$ pollution were not consistent. Using correlation analysis, some studies found that the relationship between relative humidity and PM$_{2.5}$ is negative or varies with seasons [20, 51–53]. After controlling for temporal tendency, our results showed that relative humidity is positively correlated with PM$_{2.5}$ according to the GAMM method [21]. The main reason could be that during high relative humidity, there is increased formation of secondary PM with large amounts of gas-phase chemical pollutants (CO, O$_3$, SO$_2$, and NO$_x$) [19, 56]. Such situations are also not conducive to air pollutant diffusion.

There have been few studies exploring the relationship between PM$_{2.5}$ and air pressure, as well as sunlight hours. Our results showed that air pressure has a delayed influence on PM$_{2.5}$ concentration, with a positive correlation. In general, certain weather conditions (e.g. precipitation) following low pressure environment may explain this phenomenon. However, evidence is insufficient and more quantitative research is needed to construct a detailed picture of the impact of air pressure on PM$_{2.5}$ concentration. There is a negative relationship between sunlight hours and PM$_{2.5}$, which may be attributed to a larger atmospheric volume for dilution through an increase in boundary layer height [57].

There are limitations in this study. The sampling stations in the study are not equally distributed and are sparse in some districts, and hence a better designed sampling method should be used in future studies. Furthermore, meteorological factors may have a long-term influence on PM$_{2.5}$. We selected only factors that had strong correlation with PM$_{2.5}$ for modeling. We therefore call for future studies in Beijing to investigate the complicated relationship between PM$_{2.5}$ and meteorological conditions over longer period.

Conclusions

This study provides baseline information for a comprehensive understanding of the current PM$_{2.5}$ pollution in Beijing. The results indicate that PM$_{2.5}$ concentration has strong spatiotemporal variations. PM$_{2.5}$ pollution is more severe in winter and decreased from the south to the north part of the city. Day to day variations of PM$_{2.5}$ show a long-term trend of fluctuations with 2–6 peaks in each month. Diurnal PM$_{2.5}$ variations are observed, with peaks in the forenoon or early nighttime, or both. There is spatial heterogeneity across the observing stations in Beijing. Meteorological factors influence PM$_{2.5}$ concentration in particular forms. Generally, prior day wind speed, sunlight hours and precipitation are negatively correlated with PM$_{2.5}$, whereas relative humidity and air pressure 3 days earlier are positively correlated with PM$_{2.5}$.

Supporting Information

S1 Database. Data of daily PM$_{2.5}$ concentrations and meteorological variables.
(XLSX)

S2 Database. Data of hourly PM$_{2.5}$ concentrations.
(XLSX)

S1 Table. Correlation coefficient matrix of PM$_{2.5}$ and meteorological factors.
(XLSX)

The header above reads: PM$_{2.5}$ and Meteorological Factors

Acknowledgments

Author Contributions

Conceived and designed the experiments: FFH X. Li ASY XHG. Performed the experiments: FFH X. Li YXL LXT. Analyzed the data: FFH X. Li CW QX. Contributed reagents/materials/analysis tools: FFH X. Li QG JG SPC KC LL NG X. Liu KY. Wrote the paper: FFH X. Li WW.

References

1. Dockery DW, Pope CA 3rd. Acute respiratory effects of particulate air pollution. Annu Rev Public Health. 1994; 15: 107–32. PMID: 8054077

2. Lee BJ, Kim B, Lee K. Air pollution exposure and cardiovascular disease. Toxicol Res. 2014; 30(2): 71–5. doi: 10.5487/TR.2014.30.2.071 PMID: 25071915

3. Pelucchi C, Negri E, Gallus S, Boffetta P, Tramacere I, La Vecchia C. Long-term particulate matter exposure and mortality: a review of European epidemiological studies. BMC Public Health. 2009; 9: 453. doi: 10.1186/1471-2458-9-453 PMID: 19995424

4. Ailshire JA, Crimmins EM. Fine particulate matter air pollution and cognitive function among older US adults. Am J Epidemiol. 2014; 180(4): 359–66. doi: 10.1093/aje/kwu155 PMID: 24966214

5. Liang R, Zhang B, Zhao X, Ruan Y, Lian H, Fan Z. Effect of exposure to PM$_{2.5}$ on blood pressure: a systematic review and meta-analysis. J Hypertens. 2014; 32(11): 2130–41. PMID: 25250520

6. Wu S, Deng F, Hao Y, Wang X, Zheng C, Lv H, et al. Fine particulate matter, temperature, and lung function in healthy adults: findings from the HVNR study. Chemosphere. 2014; 108: 168–74. doi: 10.1016/j.chemosphere.2014.01.032 PMID: 24548647

7. Shin HH, Fann N, Burnett RT, Cohen A, Hubbell BJ. Outdoor fine particles and nonfatal strokes: systematic review and meta-analysis. Epidemiology. 2014; 25(6): 835–42. PMID: 25188557

8. Turner MC, Krewski D, Pope CA 3rd, Chen Y, Gapstur SM, Thun MJ. Long-term ambient fine particulate matter air pollution and lung cancer in a large cohort of never-smokers. Am J Respir Crit Care Med. 2011; 184(12): 1374–81. doi: 10.1164/rccm.201106-1011OC PMID: 21980033

9. Peters A, Veronesi B, Calderón-Garcidueñas L, Gehr P, Chen LC, Geiser M, et al. Translocation and potential neurological effects of fine and ultrafine particles a critical update. Part Fibre Toxicol. 2006; 3: 13. PMID: 16961926

10. Beijing Municipal Bureau of Statistics. Beijing Statistical Yearbook. Beijing: China Statistics Press; 2014.

11. Li M, Zhang L. Haze in China: current and future challenges. Environ Pollut. 2014; 189: 85–6. doi: 10.1016/j.envpol.2014.02.024 PMID: 24637256

12. Wang Y, Ying Q, Hu J, Zhang H. Spatial and temporal variations of six criteria air pollutants in 31 provincial capital cities in China during 2013–2014. Environ Int. 2014; 73: 413–22. doi: 10.1016/j.envint.2014.08.016 PMID: 25244704

13. Guo Y, Li S, Tian Z, Pan X, Zhang J, Williams G. The burden of air pollution on years of life lost in Beijing, China, 2004–08: retrospective regression analysis of daily deaths. BMJ. 2013; 347: f7139. doi: 10.1136/bmj.f7139 PMID: 24322399

14. Zhang A, Qi Q, Jiang L, Zhou F, Wang J. Population exposure to PM$_{2.5}$ in the urban area of Beijing. PloS One. 2013; 8(5): e63486. doi: 10.1371/journal.pone.0063486 PMID: 23658832

15. Guo Y, Jia Y, Pan X, Liu L, Wichmann HE. The association between fine particulate air pollution and hospital emergency room visits for cardiovascular diseases in Beijing, China. Sci Total Environ. 2009; 407(17): 4826–30. doi: 10.1016/j.scitotenv.2009.05.022 PMID: 19501385

16. Leitte AM, Schlink U, Herbarth O, Wiedensohler A, Pan XC, Hu M, et al. Size-segregated particle number concentrations and respiratory emergency room visits in Beijing, China. Environ Health Perspect. 2011; 119(4): 508–13. doi: 10.1289/ehp.1002203 PMID: 21118783

17. Yu Y, Schleicher N, Norra S, Fricker M, Dietze V, Kaminski U, et al. Dynamics and origin of PM$_{2.5}$ during a three-year sampling period in Beijing, China. J Environ Monit. 2011; 13(2): 334–46. doi: 10.1039/c0em00467g PMID: 21180709

18. Wang JF, Hu MG, Xu CD, Christakos G, Zhao Y. Estimation of citywide air pollution in Beijing. PloS One. 2013; 8(1): e53400. doi: 10.1371/journal.pone.0053400 PMID: 23320082

19. Song C, Pei T, Yao L. Analysis of the characteristics and evolution modes of PM$_{2.5}$ pollution episodes in Beijing, China during 2013. Int J Environ Res Public Health. 2015; 12(2): 1099–111. doi: 10.3390/ijerph120201099 PMID: 25648172

20. Zhang Q, Quan J, Tie X, Li X, Liu Q, Gao Y, et al. Effects of meteorology and secondary particle formation on visibility during heavy haze events in Beijing, China. Sci Total Environ. 2015; 502: 578–84. doi: 10.1016/j.scitotenv.2014.09.079 PMID: 25300022

21. Zhang H, Wang Y, Hu J, Ying Q, Hu XM. Relationships between meteorological parameters and criteria air pollutants in three megacities in China. Environ Res. 2015; 140: 242–54. doi: 10.1016/j.envres.2015.04.004 PMID: 25880606

22. Matheron G. Principles of geostatistics. Econ geol. 1963; 58: 1246–66.

23. Giakoumi A, Maggos TH, Michopoulos J, Helmis C, Vasilakos CH. PM2.5 and volatile organic compounds (VOCs) in ambient air: a focus on the effect of meteorology. Environ Monit Assess. 2009; 152 (1–4): 83–95. doi: 10.1007/s10661-008-0298-2 PMID: 18536869

24. Zhang Y, Li M, Bravo MA, Jin L, Nori-Sarma A, Xu Y, et al. Air quality in Lanzhou, a major industrial city in China: characteristics of air pollution and review of existing evidence from air pollution and health studies. Water Air Soil Pollut. 2014; 225(10): 2187. PMID: 25838615

25. Bravo MA, Bell ML. Spatial heterogeneity of PM10 and O3 in São Paulo, Brazil, and implications for human health studies. J Air Waste Manag Assoc. 2011; 61(1): 69–77. PMID: 21305890

26. Coull BA, Schwartz J, Wand MP. Respiratory health and air pollution: additive mixed model analyses. Biostatistics. 2001; 2(3): 337–49. PMID: 12933543

27. Xu M, Guo Y, Zhang Y, Westerdahl D, Mo Y, Liang F, et al. Spatiotemporal analysis of particulate air pollution and ischemic heart disease mortality in Beijing, China. Environ Health. 2014; 13: 109. doi: 10.1186/1476-069X-13-109 PMID: 25495440

28. Ito K, Thurston GD, Silverman RA. Characterization of PM2.5, gaseous pollutants, and meteorological interactions in the context of time-series health effects models. J Expo Sci Environ Epidemiol. 2007; 17 Suppl 2: S45–60. doi: 10.1038/sj.jes.7500627 PMID: 18079764

29. Wood SN. Generalized additive models: an introduction with R. New York: Chapman & Hall/CRC; 2006.

30. Clifford S, Low Choy S, Hussein T, Mengersen K, Morawska L. Using the generalised additive model to model the particle number count of ultrafine particles. Atmos Environ. 2011; 45(32): 5934–45.

31. Wang G, Cheng S, Li J, Lang J, Wen W, Yang X, et al. Source apportionment and seasonal variation of PM$_{2.5}$ carbonaceous aerosol in the Beijing-Tianjin-Hebei region of China. Environ Monit Assess. 2015; 187(3): 143. doi: 10.1007/s10661-015-4288-x PMID: 25716523

32. Chai F, Gao J, Chen Z, Wang S, Zhang Y, Zhang J, et al. Spatial and temporal variation of particulate matter and gaseous pollutants in 26 cities in China. J Environ Sci. 2014; 26(1): 75–82.

33. Ji D, Wang Y, Wang L, Chen L, Hu B, Tang G et al. Analysis of heavy pollution episodes in selected cities of northern China. Atmos Environ. 2012; 50: 338–48.

34. Liu J, Mo L, Zhu L, Yang Y, Liu J, et al. Removal efficiency of particulate matters at different underlying surfaces in Beijing. Environ Sci Pollut Res Int. 2015.

35. Xiao Q, Ma Z, Li S, Liu Y. The impact of winter heating on air pollution in China. PLoS One. 2015; 10 (1): e0117311. doi: 10.1371/journal.pone.0117311

36. State Forestry Administration of the People's Republic of China. China Forestry Development Report. 2013. Available from: http://www.forestry.gov.cn/.

37. Motallebi N, Tran H, Croes BE, Larsen LC. Day-of-week patterns of particulate matter and its chemical components at selected sites in California. J Air Waste Manag Assoc. 2003; 53(7): 876–88. PMID: 12880074

38. Blanchard CL, Tanenbaum S. Weekday/Weekend differences in ambient air pollutant concentrations in atlanta and the southeastern United States. J Air Waste Manag Assoc. 2006; 56(3): 271–84. PMID: 16573190

39. Hu J, Wang Y, Ying Q, Zhang H. Spatial and temporal variability of PM$_{2.5}$ and PM$_{10}$ over the North China Plain and the Yangtze River Delta, China. Atmos Environ. 2014; 95: 598–609.

40. Shen GF, Yuan SY, Xie YN, Xia SJ, Li L, Yao YK, et al. Ambient levels and temporal variations of PM$_{2.5}$ and PM$_{10}$ at a residential site in the mega-city, Nanjing, in the western Yangtze River Delta, China. J Environ Sci Health A Tox Hazard Subst Environ Eng. 2014; 49(2): 171–8. doi: 10.1080/10934529.2013.838851 PMID: 24171416

41. Cyrys J, Pitz M, Heinrich J, Wichmann HE, Peters A. Spatial and temporal variation of particle number concentration in Augsburg, Germany. Sci Total Environ. 2008; 401(1–3): 168–75. doi: 10.1016/j.scitotenv.2008.03.043 PMID: 18511107

42. Huang P, Zhang J, Tang Y, Liu L. Spatial and Temporal Distribution of PM$_{2.5}$ pollution in Xi'an city, China. Int J Environ Res Public Health. 2015; 12(6): 6608–25. doi: 10.3390/ijerph120606608 PMID: 26068090

43. Wallace J, Kanaroglou P. The effect of temperature inversions on ground-level nitrogen dioxide (NO2) and fine particulate matter (PM2.5) using temperature profiles from the Atmospheric Infrared Sounder (AIRS). Sci Total Environ. 2009; 407(18): 5085–95. doi: 10.1016/j.scitotenv.2009.05.050 PMID: 19540568

44. Liu Z, Hu B, Wang L, Wu F, Gao W, Wang Y. Seasonal and diurnal variation in particulate matter (PM10 and PM2.5) at an urban site of Beijing: analyses from a 9-year study. Environ Sci Pollut Res Int. 2015; 22(1): 627–42. doi: 10.1007/s11356-014-3347-0 PMID: 25096488

45. Guinot B, Roger J, Cachier H, Wang P, Bai J, Tong Y. Impact of vertical atmospheric structure on Beijing aerosol distribution. Atmos Environ. 2006; 40(27): 5167–80.

46. Miao S, Chen F, LeMone M, Tewari M, Li Q, Wang Y. An observational and modeling study of characteristics of Urban Heat Island and boundary layer structures in Beijing. J Appl Meteorol Climatol. 2008; 48(3): 484–501.

47. Wilson JG, Kingham S, Sturman AP. Intraurban variations of PM$_{10}$ air pollution in Christchurch, New Zealand: implications for epidemiological studies. Sci Total Environ. 2006; 367(2–3): 559–72. PMID: 16243380

48. Pinto JP Lefohn AS, Shadwick DS. Spatial variability of PM2.5 in urban areas in the United States. J Air Waste Manag Assoc. 2004; 54(4): 440–9. PMID: 15115373

49. Liu Y, Paciorek CJ, Koutrakis P. Estimating regional spatial and temporal variability of PM$_{2.5}$ concentrations using satellite data, meteorology, and land use information. Environ Health Perspect. 2009; 117 (6): 886–92. doi: 10.1289/ehp.0800123 PMID: 19590678

50. Yanosky JD, Paciorek CJ, Laden F, Hart JE, Puett RC, Liao D, et al. Spatio-temporal modeling of particulate air pollution in the conterminous United States using geographic and meteorological predictors. Environ Health. 2014; 13: 63. doi: 10.1186/1476-069X-13-63 PMID: 25097007

51. Vassilakos C, Saraga D, Maggos T, Michopoulos J, Pateraki S, Helmis CG. Temporal variations of PM$_{2.5}$ in the ambient air of a suburban site in Athens, Greece. Sci Total Environ. 2005; 349: 223–31. PMID: 16198683

52. Trivedi DK, Ali K, Beig G. Impact of meteorological parameters on the development of fine and coarse particles over Delhi. Sci Total Environ. 2014; 478(1–3): 175–83.

53. Akyüz M, Cabuk H. Meteorological variations of PM$_{2.5}$/PM$_{10}$ concentrations and particle-associated polycyclic aromatic hydrocarbons in the atmospheric environment of Zonguldak. J Hazard Mater. 2009; 170(1): 13–21. doi: 10.1016/j.jhazmat.2009.05.029 PMID: 19523758

54. Li F, Zhang C. Analysis on the relationship between PM$_{2.5}$ and precipitation in Xi'an. Zhong Guo Huan Jing Jian Ce. 2013; 29(6): 22–8. Chinese.

55. Li L, Qian J, Ou CQ, Zhou YX, Guo C, Guo Y. Spatial and temporal analysis of Air Pollution Index and its timescale-dependent relationship with meteorological factors in Guangzhou, China, 2001–2011. Environ Pollut. 2014; 190: 75–81. doi: 10.1016/j.envpol.2014.03.020 PMID: 24732883

56. Olivares G, Johansson C, Ström J, Hansson HC. The role of ambient temperature for particle number concentrations in a street canyon. Atmos Environ. 2007; 41(10): 2145–55.

57. Pal S, Lee TR, Phelps S, De Wekker SF. Impact of atmospheric boundary layer depth variability and wind reversal on the diurnal variability of aerosol concentration at a valley site. Sci Total Environ. 2014; 496: 424–34. doi: 10.1016/j.scitotenv.2014.07.067 PMID: 25105753