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RESEARCH ARTICLE

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/m³ and 71.00 μg/m³, 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

data collection and analysis, decision to publish, or preparation of the manuscript.

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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 (PM₁₀), 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 $\mu\text{g}/\text{m}^3$ 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 $\mu\text{g}/\text{m}^3$, 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.

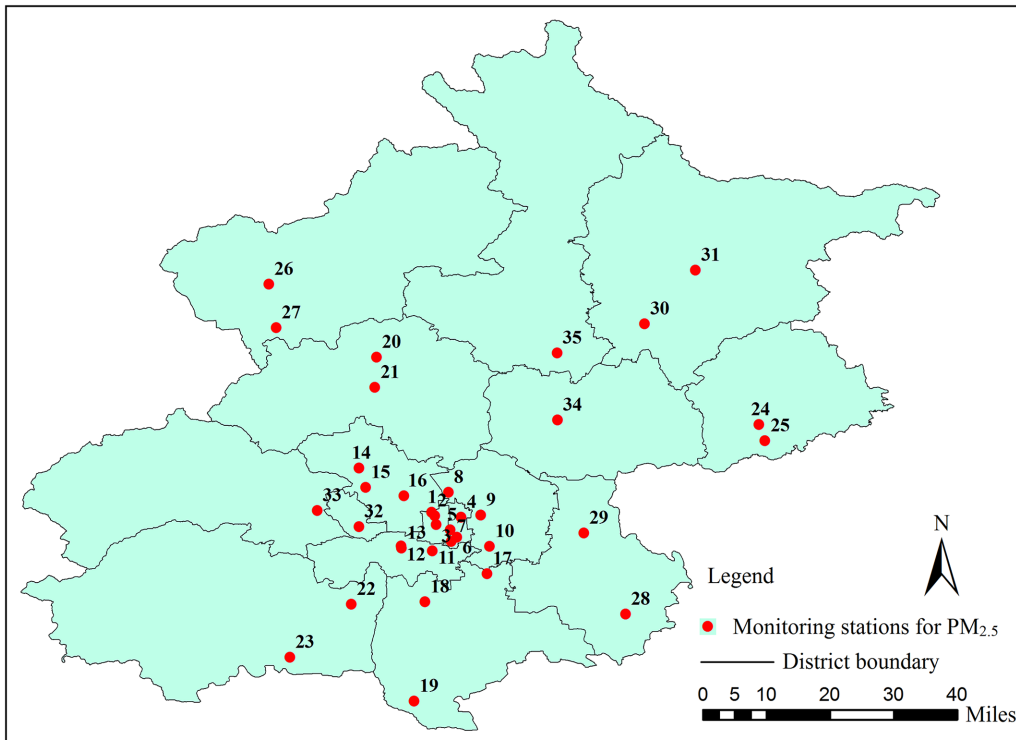
Daily mean concentrations for each district and the whole city were calculated by averaging concentrations reported by all 35 stations, which is the same method used by BJEPD to report daily concentration of air pollutants to the public. Rates of missing values in the 16 districts were mostly low ranging 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/m³) and II (75 µg/m³) 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/m³), 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 PM_{2.5} 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) $\mu\text{g}/\text{m}^3$ | Median (IQR) $\mu\text{g}/\text{m}^3$ | Range $\mu\text{g}/\text{m}^3$ | 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 $\mu\text{g}/\text{m}^3$ in district *Miyun* to 107.63 $\mu\text{g}/\text{m}^3$ in district *Tongzhou*, greatly exceeding the yearly CAAQS (GB3095-2012) Grade I (15 $\mu\text{g}/\text{m}^3$) and II standards (35 $\mu\text{g}/\text{m}^3$) for all districts in Beijing (Table 1). The citywide mean concentration of 88.07 $\mu\text{g}/\text{m}^3$ 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 $\mu\text{g}/\text{m}^3$) standard during more than 60% of days (a non-attainment rate of 60%) and Grade II (75 $\mu\text{g}/\text{m}^3$) 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 | | |
| Day of week | Weekdays vs. Weekends | | | Z | P |
| | | | | -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.

doi:10.1371/journal.pone.0141642.t002

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/m³) 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 months (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 1 pm 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

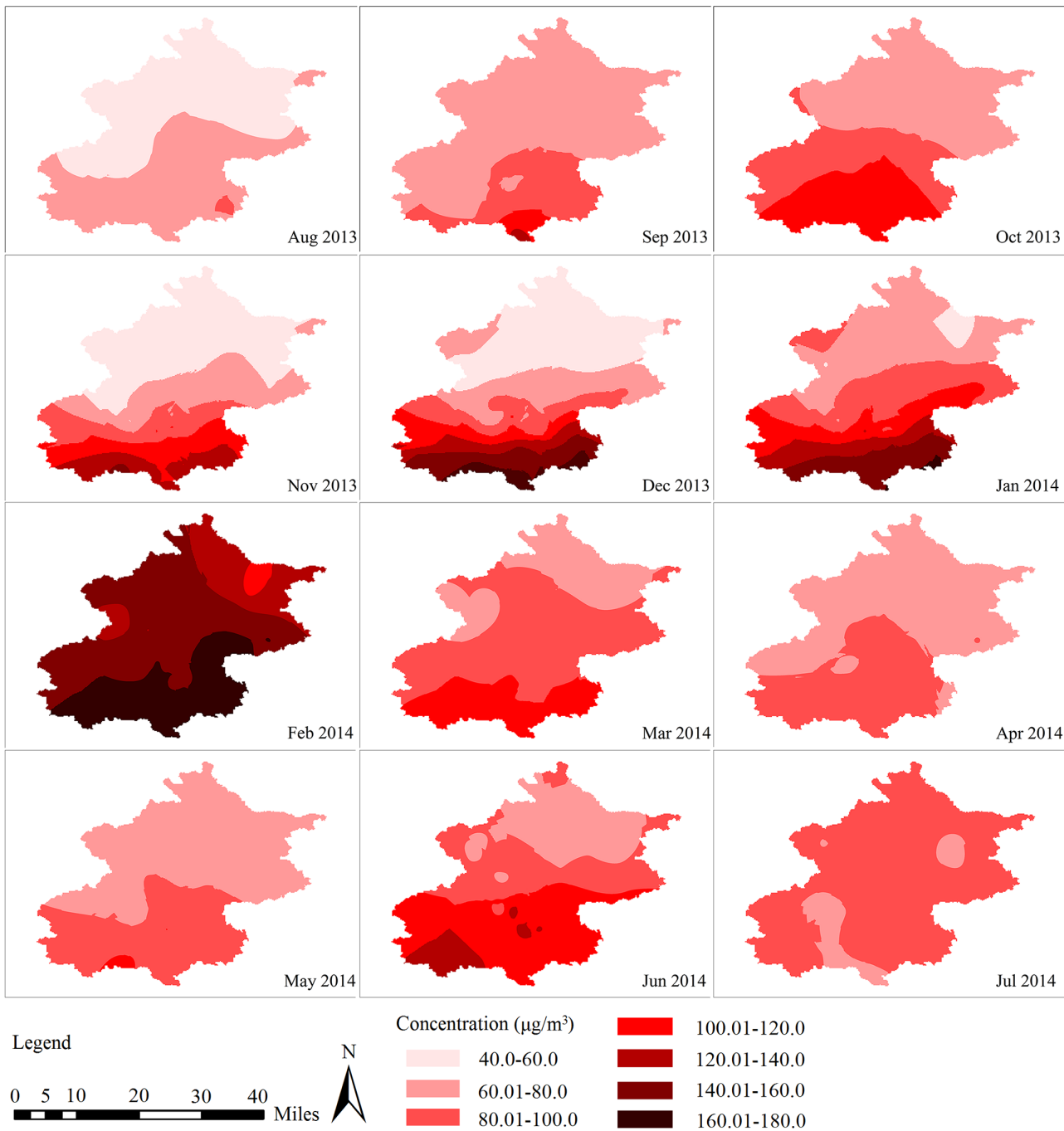


Fig 2. Regional and seasonal variations of PM_{2.5} in Beijing, 2013–2014.

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

Association between PM_{2.5} 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 PM_{2.5} concentrations

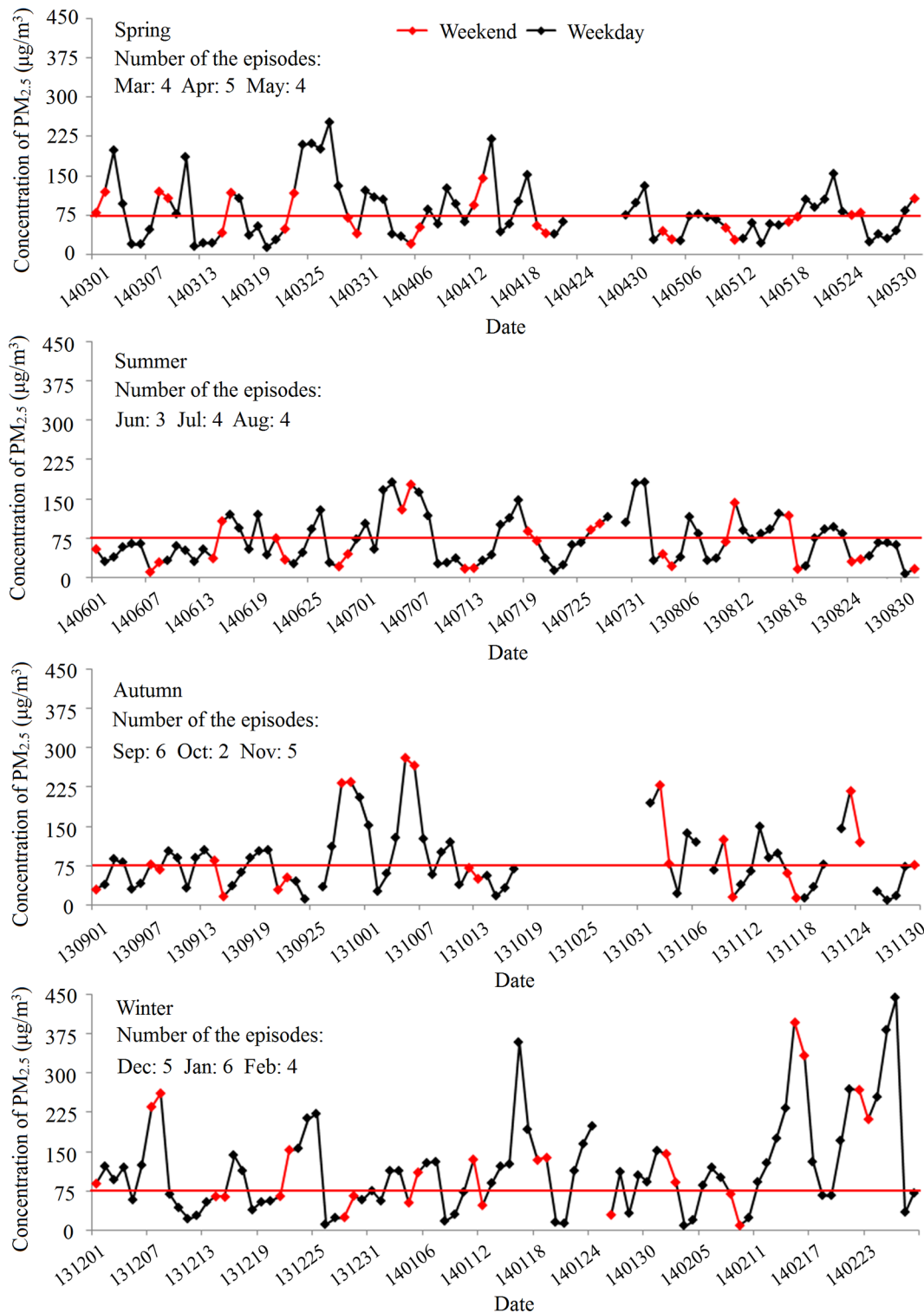


Fig 3. Day to day variations of PM_{2.5} in different seasons, Beijing, 2013–2014.

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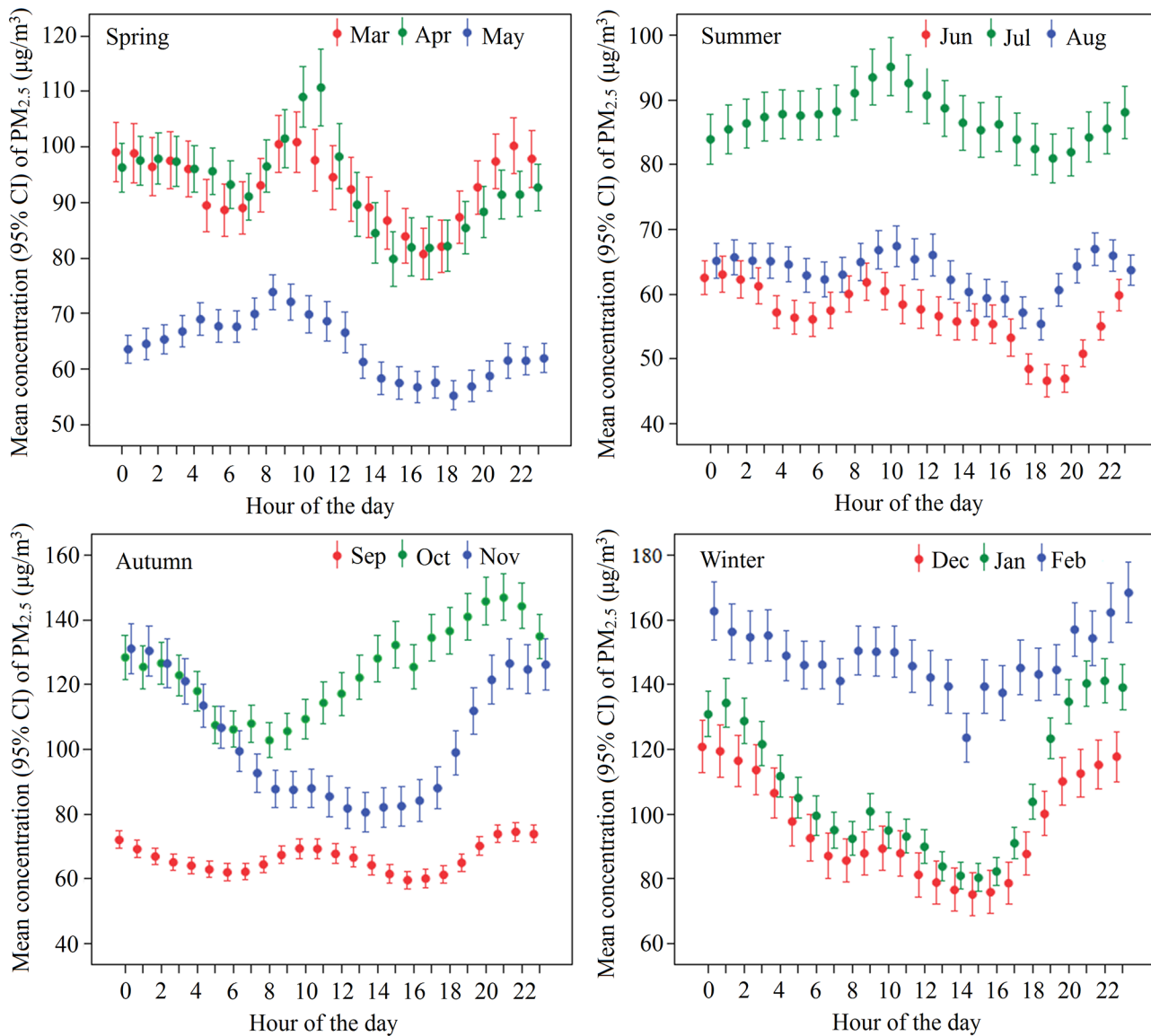


Fig 4. Diurnal variations of PM_{2.5} in different months, Beijing, 2013–2014.

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(S1 Table). 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 PM_{2.5} 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,

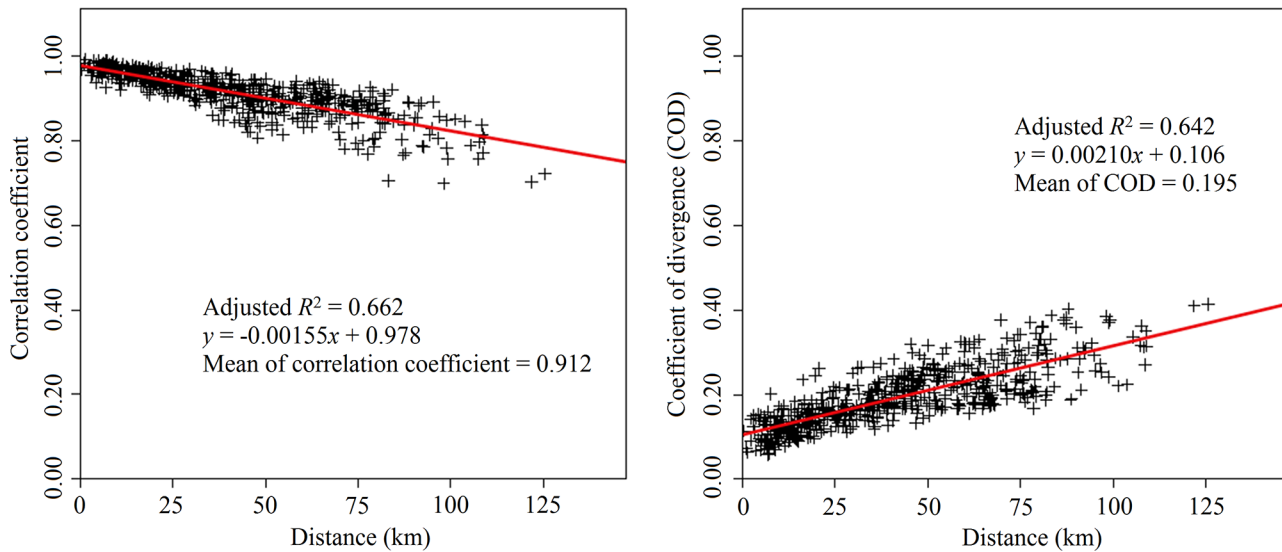


Fig 5. Correlation coefficient and COD versus distance between the stations.

doi:10.1371/journal.pone.0141642.g005

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 form.

Discussion

The study shows that Beijing has serious PM_{2.5} pollution citywide throughout the year [12–14]. We observed great 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

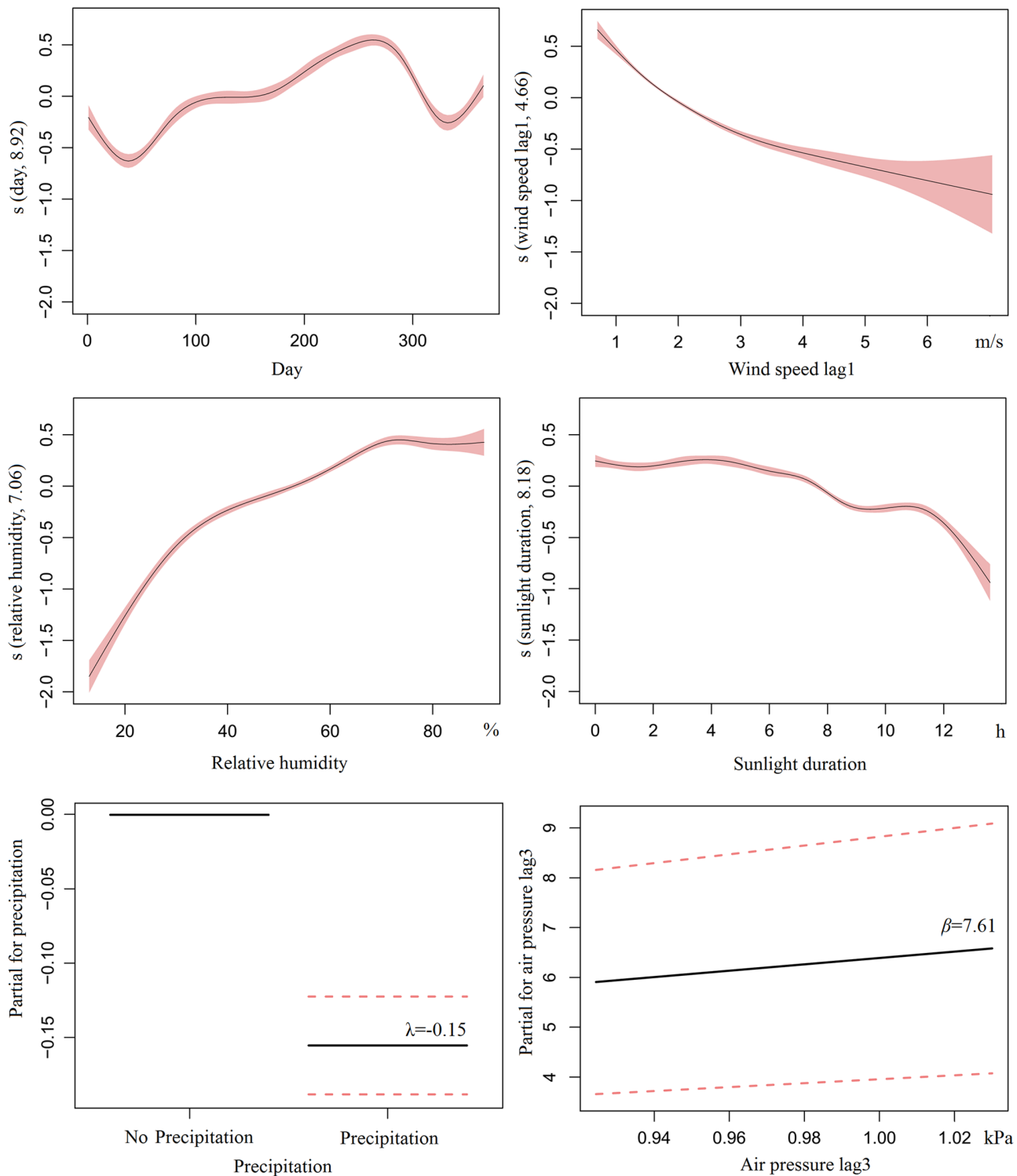


Fig 6. Exposure-response curves for PM_{2.5} and meteorological variables, Beijing, 2013–2014.

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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, O₃, SO₂, 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)

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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.

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