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Authors for correspondence:
Song Xi Chen
e-mail: csx@gsm.pku.edu.cn
Hui Huang
e-mail: huanghui@math.pku.edu.cn

Assessing Beijing's PM$_{2.5}$ pollution: severity, weather impact, APEC and winter heating

Xuan Liang[1], Tao Zou[1], Bin Guo[4], Shuo Li[1], Haozhe Zhang[5], Shuyi Zhang[1], Hui Huang[2,3] and Song Xi Chen[1,2]

[1]Guanghua School of Management, [2]Center for Statistical Science, and [3]Department of Probability and Statistics, Peking University, Beijing 100871, People's Republic of China
[4]School of Economics, Sichuan University, Chengdu 610065, People's Republic of China
[5]Department of Statistics, Iowa State University, Ames, IA 50011, USA

By learning the PM$_{2.5}$ readings and meteorological records from 2010–2015, the severity of PM$_{2.5}$ pollution in Beijing is quantified with a set of statistical measures. As PM$_{2.5}$ concentration is highly influenced by meteorological conditions, we propose a statistical approach to adjust PM$_{2.5}$ concentration with respect to meteorological conditions, which can be used to monitor PM$_{2.5}$ pollution in a location. The adjusted monthly averages and percentiles are employed to test if the PM$_{2.5}$ levels in Beijing have been lowered since China's State Council set up a pollution reduction target. The results of the testing reveal significant increases, rather than decreases, in the PM$_{2.5}$ concentrations in the years 2013 and 2014 as compared with those in year 2012. We conduct analyses on two quasi-experiments—the Asia-Pacific Economic Cooperation meeting in November 2014 and the annual winter heating—to gain insight into the impacts of emissions on PM$_{2.5}$. The analyses lead to a conclusion that a fundamental shift from mainly coal-based energy consumption to much greener alternatives in Beijing and the surrounding North China Plain is the key to solving the PM$_{2.5}$ problem in Beijing.

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1. Introduction

Beijing and a substantial part of China are experiencing chronic air pollution. The main pollutants are fine particulate matter, and $PM_{2.5}$ in particular [1,2]. $PM_{2.5}$ consists of airborne particles with aerodynamic diameters of less than 2.5 μm. They are known to influence visibility, human health and even climate [3]. Epidemiological evidence shows that exposure to $PM_{2.5}$ can cause lung morbidity [4], serious respiratory and cardiovascular diseases, and even death [5–7].

There are studies on the chemical characteristics and formation mechanisms of $PM_{2.5}$ [8], in particular over Chinese cities [9–11]. Studies [1,12] reveal that the chemical composition of $PM_{2.5}$ varies significantly across regions of China, with changing physical attributes within polluting episodes [2,13]. Recent studies on the record-breaking episode of $PM_{2.5}$ pollution in January 2013 have discovered that meteorological conditions, secondary aerosols, local emissions and regional transportation contribute to the formation and development of $PM_{2.5}$ in Beijing. Chemical transport models are used to study the vertical and horizontal pattern of $PM_{2.5}$ dynamics [14,15]. Exploratory analyses have related anomalous wind and humidity conditions with high $PM_{2.5}$ concentration [3,16]. Contributions of local and regional emissions to Beijing's air pollution [17] have also been studied.

An important implication from these studies is that there are many non-ignorable sources of variability in the distribution and transmission patterns of $PM_{2.5}$, confounded by meteorological conditions, emissions at source and secondary chemical generation. Such uncertainties bring challenges in the assessment and monitoring of $PM_{2.5}$ in Beijing [18]. To diagnose and forecast air quality, deterministic models such as the Pollution Linked with Air-Quality and Meteorology Index [19] and the Community Multi-Scale Air Quality modelling system have been widely adopted in China [20,21]. These methods do not fully account for the uncertainties mentioned above. In addition, the relationship between $PM_{2.5}$ and the confounding factors remains unclear due to large variability in the observed $PM_{2.5}$ data. Quantification of this relationship requires data of sufficient time span together with comprehensive statistical analysis in order to measure the uncertainty and to adjust for the confounding factors. Statistical models, such as Bayesian hierarchical space–time models [22,23] and generalized additive models [24], were used to study ambient air pollution in the USA (see also [25]). Quasi-experiments (QEs) have been considered to evaluate the impact of $PM_{2.5}$ on human health [26]. As $PM_{2.5}$ concentration is highly influenced by meteorological conditions, we propose a statistical approach to adjust $PM_{2.5}$ distribution with respect to meteorological conditions, which can be used to monitor $PM_{2.5}$ pollution at a location. We show that the proposed adjustment can be used in conjunction with QEs to evaluate the impacts of emissions on $PM_{2.5}$ concentration.

Given that there are 22 million inhabitants in Beijing, and 300 million immediately to the south in the North China Plain (NCP), it is vital to measure the severity of the $PM_{2.5}$ pollution in Beijing. This is particularly relevant since China's State Council has a target of reducing $PM_{2.5}$ by at least 25% from the 2012 level by 2017 for Beijing [27]. This paper provides a set of statistical measures for key aspects of the $PM_{2.5}$ pollution, which can be used as benchmarks in evaluating the effectiveness of any pollution mitigation initiatives in Beijing and beyond.

Our analysis uses hourly $PM_{2.5}$ readings taken at the US Embassy in Beijing located at (116.47 E, 39.95 N), in conjunction with hourly meteorological measurements at Beijing Capital International Airport (BCIA), obtained from weather.nocrew.org. Both data series run from 1 January 2010 to 31 December 2014. Although the embassy and the airport are 17 km apart, they experience very much the same weather. The US Embassy started to announce hourly $PM_{2.5}$ readings from April 2008 at a different location. We did not use the data in 2008 and 2009 due to a large number of missing values and the embassy moving to its current location in 2009. China's Ministry of Environmental Protection (MEP) started to report $PM_{2.5}$ readings only from January 2013. We have hourly $PM_{2.5}$ data released by the Beijing Municipal Environmental Monitoring Center from May to December 2014 at two locations in Beijing, which are used to calibrate with the embassy data.

Our study appears to be the first that combines $PM_{2.5}$ and meteorological data for an extended time span (5 years) in studying China's $PM_{2.5}$ pollution. Due to the high variability and confounding by weather conditions, we believe that only data of sufficient length with high temporal frequency can produce accurate assessment of the severity of the air pollution and quantify its trend and pattern. These distinguish our analysis from existing short-term exploratory studies (e.g. [16]).

2. Basic statistical prognosis

We first provide a set of descriptive statistics on the extent of the $PM_{2.5}$ pollution in Beijing. According to the US (EPA) standard, $35\,\mu g\,m^{-3}$ (the European Union uses $25\,\mu g\,m^{-3}$) is the highest $PM_{2.5}$ level for acceptable air quality, while $150\,\mu g\,m^{-3}$ is widely viewed as very unhealthy and even hazardous.

We partition the $PM_{2.5}$ time series into three states: low PM state when $PM_{2.5} \leq 35\,\mu g\,m^{-3}$; polluting episode when $PM_{2.5} > 35\,\mu g\,m^{-3}$; and very high PM when $PM_{2.5} > 150\,\mu g\,m^{-3}$. To reduce the noise in the PM readings, we smooth the time series over 3 h moving windows. The smoothing is only used for the three states, and original hourly data are used in the subsequent analysis on $PM_{2.5}$. Figure 1a displays the average length of time persisted under the three states of $PM_{2.5}$ by year. Figure 1b reports the percentages of time in each of the three states by years and seasons. There were, on average, 88 polluting episodes per year between 2010 and 2014, about 1.7 per week, with an average length of around 73 h per episode. About 84% of the polluting episodes reached very high PM, which is not surprising given the rather long duration of the episodes. Among these episodes, the average length for having very unhealthy air (very high PM) was 25 h. The good quality air (low PM) in Beijing occupied 23% of the time, which was almost the same as that of the hazardous very high PM (22%). These percentages and statistics did not vary significantly over the 5 years. There were some seasonal variations in figure 1b with both the winter and autumn having longer low PM and very high PM periods than spring and summer, which is largely due to seasonal wind and emission patterns. These statistics portray a very severe situation for Beijing's air pollution.

To gain information on how representative the embassy data are, figure 1c displays the $PM_{2.5}$ series at the embassy and the two Beijing Municipal Environmental Monitoring Center (BMEMC) sites in Beijing: Nongzhanguan (Agriculture Exhibition Hall) and Dongsihuan Beilu (East Fourth Ring Road North). Both sites are very close to the embassy, with the former about 1.2 km south and the latter about 1.5 km southeast. The BMEPC data were from 11 May 2014, the date we started to collect data from the BMEPC website. Figure 1c shows that the readings were highly consistent among the three sites. Hence, results similar to those from figure 1a,b would also be attained from the BMEPC data.

3. Impacts of meteorological variables

Beijing residents have realized that wind tends to alleviate the air pollution, and long for stronger wind during the worst of the high $PM_{2.5}$ episodes. Lack of wind has been frequently blamed for high $PM_{2.5}$ in Beijing, far more often than anthropogenic activities that contribute to the pollution. We examine the influence of wind on $PM_{2.5}$ pollution by connecting the $PM_{2.5}$ data to the meteorological data. The weather data had 16 wind directions. Our study shows that the directions can be grouped into five broad categories: northwest (NW), which includes W, WNW, NW, NNW and N; northeast (NE), for NNE, NE and ENE; southeast (SE), covering E, ESE, SE, SSE and S; southwest (SW), having SSW, SW and WSW; and calm and variable (CV). The decision to allocate E to SE and W to NW was based on the locations of major polluting industries around Beijing.

Figure 2a displays the distribution of wind direction and average wind speed at five regimes of $PM_{2.5}$ pollution: low PM ($\leq 35\,\mu g\,m^{-3}$), polluting episode ($> 35\,\mu g\,m^{-3}$), very high PM ($> 150\,\mu g\,m^{-3}$), the beginning and the ending periods of the polluting episodes, as well as at

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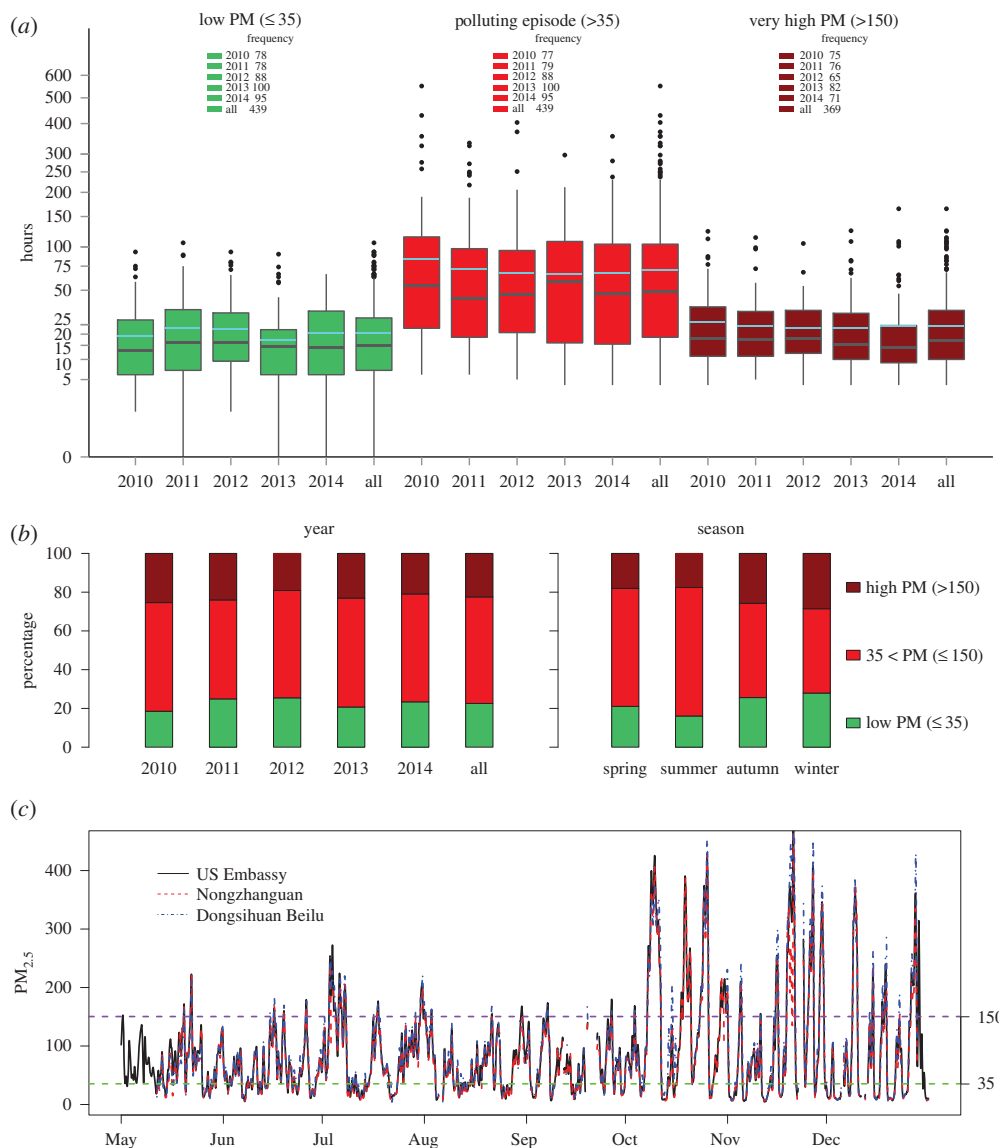
Figure 1. Summary statistics for $PM_{2.5}$ pollution in Beijing 2010–2014. (*a*) Box-plots for the time length of the three $PM_{2.5}$ states in Beijing. Black bars inside the boxes are the median hours, and the dashed bars are the averages. (*b*) Percentages of times in different years and seasons under the three states. (*c*) $PM_{2.5}$ readings at the US Embassy, Nongzhanguan and Dongsihuan Beilu from 1 May to 31 December 2014.

the overall baseline. A beginning (ending) period is determined by the hour that the $PM_{2.5}$ first surpasses (drops below) $35\,\mu g\,m^{-3}$, plus the 2 h before and after it. The baseline wind distribution in Beijing is dominated by NW and SE, with winters being dominated by NW and summers by SE, as shown in the electronic supplementary material, figure S1. However, during the low $PM_{2.5}$ and the ending period of the polluting episodes, there are far more northerly winds (around 80% for the low $PM_{2.5}$ and 84% for the ending period) than the baseline (43%) with much higher wind speed, and far less S and CV (only 7–10% and 9–11%, respectively, as compared with 35% and 21% at the baseline). We see a jump in SW, from a mere 5% at the baseline to 13% at the beginning of the polluting episodes, and a moderate increase of SE and CV during the polluting episodes. The very high PM periods are strongly associated with the increases in both CV and SE and a drop of NW in both percentage and velocity.

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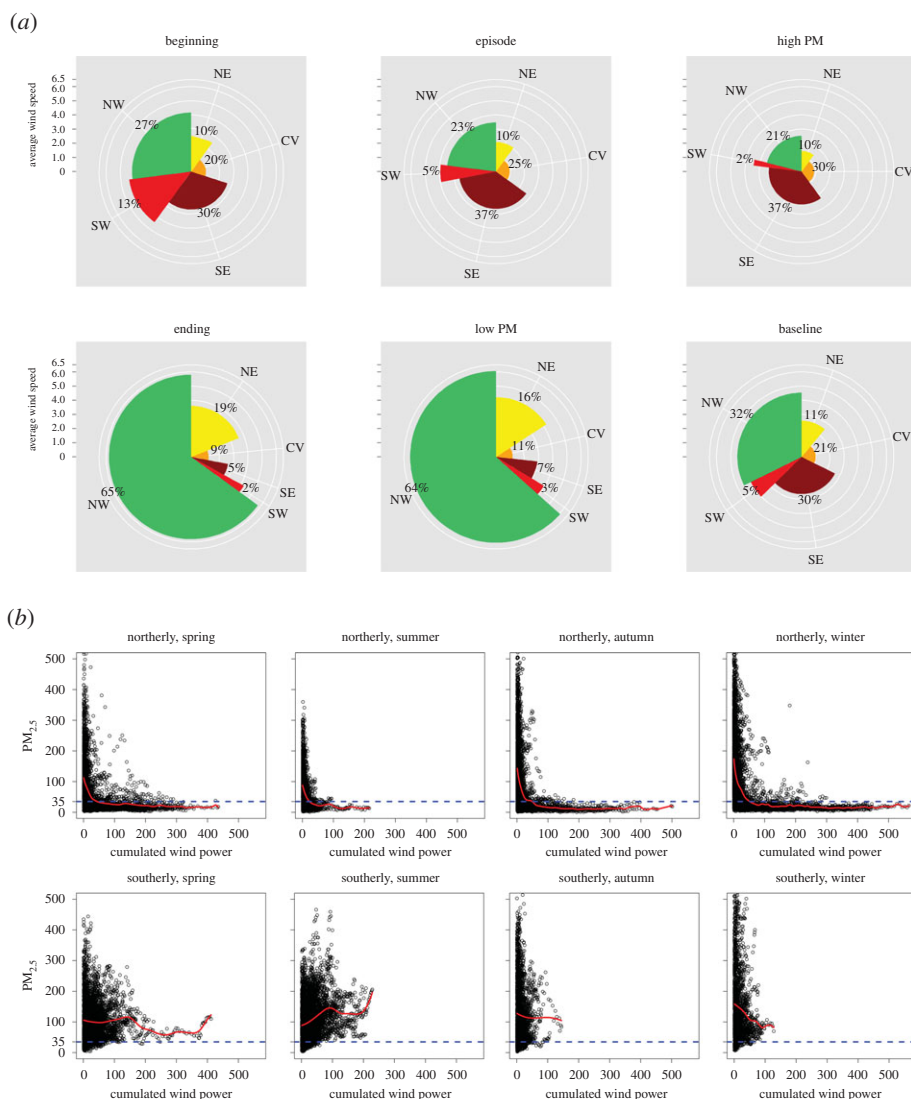
Figure 2. Impact of wind on PM$_{2.5}$. (*a*) The distribution of wind directions (via width of angles) and average speed (via length of radius) under different states of PM$_{2.5}$: beginning/ending, polluting episode (>35 μg m^{-3}), high PM (>150 μg m^{-3}), low PM (≤35 μg m^{-3}) and the baseline. All the wind distributions under the five regimes of PM$_{2.5}$ are significantly different from the baseline with almost zero p-values for the tests of independence. (*b*) PM$_{2.5}$ versus the cumulated wind power (CWP) at northerly and southerly winds for four seasons with fitted regression curves (red solid lines). The dashed lines mark 35 μg m^{-3}.

Situated at the northwest corner of the NCP, Beijing is hemmed in by Taihang Mountain to the west and Yan Mountain to the north, as shown in the electronic supplementary material, figure S2. The benefit of northerly wind is due to a lack of heavily polluting industry in the region north of Beijing. However, the mountains cause accumulation of the polluted air under a southerly wind. The south and the east of Beijing on the NCP are dense with heavy industries, which consume enormous amounts of coal and other fossil fuels. The annual coal consumption in the NCP was more than 1 billion tonnes in 2012, constituting 25% of China's and 15% of the world's consumption, in a densely populated region that accounts for only 5.6% of China's land area [28]. Additionally, there are more than 5 million cars in Beijing, which also contribute to its air pollution.

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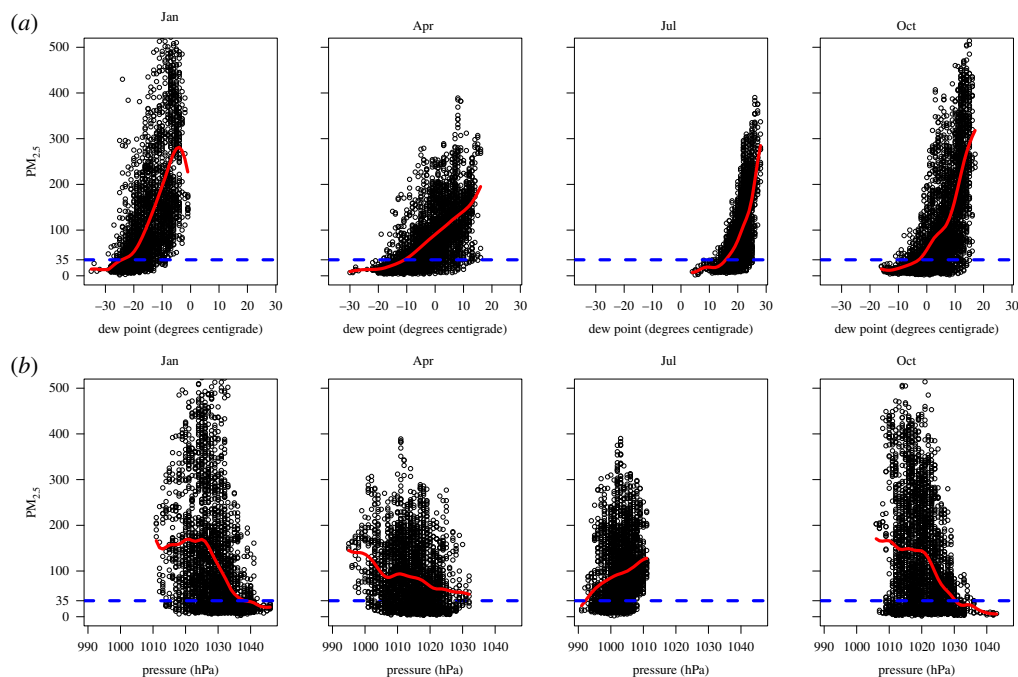
Figure 3. (*a*) PM$_{2.5}$ versus dew point; (*b*) PM$_{2.5}$ versus pressure; for four months: January, April, July and October using the 5 years of data from 2010 to 2014.

Wind speed is known to be influential in PM$_{2.5}$ for studies on the US data [29,30]. Our analysis reveals that a more effective variable is the CWP, which takes into account wind velocity in a particular direction. It reflects the fact that it is sustained wind from a fixed direction that reduces or increases the pollution. CWP is the cumulated wind speed from the start of the wind direction to the time of interest. When the wind direction changes, the CWP under a new direction starts to cumulate again. For calm wind, we use $0.445 \, \mathrm{m \, s^{-1}}$ as the unit of cumulation. Figure 2*b* displays PM$_{2.5}$ versus CWP under a northerly (NW and NE) and southerly (SE and SW) wind for the four seasons. It shows clearly that PM$_{2.5}$ can be significantly reduced by a northerly wind in all seasons. In contrast, a southerly wind does not reduce pollution; rather it generally increases it, particularly in the summer. Electronic supplementary material, figure S3, shows more information about the influence of a northerly wind.

In additional to wind, other meteorological variables are influential in the PM$_{2.5}$. Figure 3*a,b* displays the scatter plots of PM$_{2.5}$ versus dew point and pressure, respectively, for four selected months: January, April, July and October, based on the data from the years 2010 to 2014. The figure also gives fitted non-parametric regression curves, which indicate a clear increasing trend of PM$_{2.5}$ as the dew point increases, and a largely decreasing trend as the pressure increases except in July. The pressure distribution in July was much lower than that in the other three months. The meteorological variables are mutually correlated. Indeed, a decrease in the dew point and an increase in the pressure are usually accompanied by the arrival of the northerly wind, which brings in drier and fresher air. We need a systematical approach to model the overall effect of the weather variables on PM$_{2.5}$, which we deal with in the next section.

4. Modelling meteorological effects

As shown in the previous section, airborne PM$_{2.5}$ concentration is confounded with the meteorological variables. This means that different weather conditions can induce different PM$_{2.5}$ readings even if the underlying emissions are the same. For the purpose of the air-quality assessment, we need to compare the pollution levels in different periods under the same

meteorological conditions in order to be fair. However, the weather cannot be controlled. So, we must adjust for weather conditions. We are very much in the field of observational studies [31], where adjustment with respect to certain baseline variables is required in order to attain fair comparison on an outcome variable.

We provide a non-parametric framework for adjusting the mean and quantiles of the $PM_{2.5}$ distribution with respect to weather conditions in a period, say a month in a year, so that the adjusted means and quantiles of the same month in different years are comparable. The adjusted means and quantiles are needed to provide statistical evidence in order to assess if the pollution is getting better or worse.

Let Y_{ijt} be the $PM_{2.5}$ reading at an hour t in month j and year i, and X_{ijt} be a vector consisting of air pressure (P), dew point (D), temperature (T), precipitation (Prec) and CWP (C) such that $X_{ijt} = (P_{ijt}, T_{ijt}, D_{ijt}, Prec_{ijt}, C_{ijt})$. Prec refers to the cumulative rain or snow hours according to the weather description from BCIA data. Relative humidity is not included as it can be determined by temperature and dew point according to a physical relationship [32]. We denote wind direction as W_{ijt} such that $W_{ijt} = 1, 2, 3, 4$ for NW, NE, S and CV, respectively, after merging SE and SW winds with the southern wind (S).

Let U_{ijt} be the emission in the area that contains the site. One may use energy consumption as a proxy for U_{ijt}. However, as there are only monthly energy consumption statistics at the national level in China, and those at the provincial level are annual only, we have to treat U_{ijt} as a latent variable.

An underlying non-parametric regression model [33] for month j of year i is

$$Y_{ijt} = \tilde{m}_{ij}(X_{ijt}, W_{ijt}, U_{ijt}) + \epsilon_{ijt}, \quad t = 1, \ldots, n_{ij}, \tag{4.1}$$

where $\tilde{m}_{ij}(x, w, u) = E(Y_{ijt}|X_{ijt} = x, W_{ijt} = w, U_{ijt} = u)$ is the regression function, ϵ_{ijt} is the residual that satisfies $E(\epsilon_{ijt}|X_{ijt}, W_{ijt}, U_{ijt}) = 0$, and n_{ij} is the number of observations in month j of year i. The residuals $\{\epsilon_{ijt}\}$ are assumed to be stationary and weakly dependent [34] to reflect the dynamic nature of $PM_{2.5}$ data.

As U_{ijt} is latent, we consider a reduced model of (4.1)

$$Y_{ijt} = m_{ij}(X_{ijt}, W_{ijt}) + e_{ijt}, \quad t = 1, \ldots, n_{ij}, \tag{4.2}$$

where $m_{ij}(x, w) = E(Y_{ijt}|X_{ijt} = x, W_{ijt} = w)$ is the regression function based on the meteorological variables only, and $\{e_{ijt}\}$ are stationary and weakly dependent residuals. The electronic supplementary material reports diagnostic tests on the stationarity and weakly dependent assumptions of $\{e_{ijt}\}$ based on the fitted residuals of model (4.2), which shows adequate support for the assumptions.

An auto-regressive alteration of model (4.2) that better captures the dynamic of $PM_{2.5}$ is

$$Y_{ijt} = \beta Y_{ij,t-1} + g_{ij}(X_{ijt}, W_{ijt}) + \eta_{ijt}, \tag{4.3}$$

where η_{ijt} is stationary and weakly dependent such that $E(\eta_{ijt}|Y_{ij,t-1}, X_{ijt}, W_{ijt}) = 0$ and $g_{ij}(X_{ijt}, W_{ijt})$ is the non-parametric part of the model. This model is useful in developing detailed forecasting models for $PM_{2.5}$, but not so much for the adjustment with respect to the meteorological variables. The latter is the main purpose of this paper.

In the above three models, we do not assume specific parametric forms for the regression functions with respect to the meteorological variables, but want to learn from data non-parametrically. There are more than 700 hourly observations in each month (except February) for 5 years, which are sufficient for carrying out the statistical learning. The monthly time span allows enough data in the analysis under a comparable meteorological regime, especially with respect to the temperature and dew point. Longer than a month may introduce excessive heterogeneity in the weather conditions, and shorter than a month may have problems with a small sample size leading to unreliable estimates.

There may be a daily cycle of $PM_{2.5}$ concentration, not accounted for in the above models. Temperature may be viewed as a proxy for daily variation in $PM_{2.5}$, as well as for daily patterns

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in the boundary layer height (BLH), another variable that can influence the $PM_{2.5}$ density. BLH data are not available, although there are re-assimilated BLH values every 6 h in quite sparsely distributed grid points. Our analysis on the residuals did not reveal any daily or weekly cycles.

For the purpose of adjusting the $PM_{2.5}$ distribution with respect to the meteorological condition, we will concentrate on model (4.2). We consider a kernel smoothing estimator for $m_{ij}(x, w) = E(Y_{ijt}|X_{ijt} = x, W_{ijt} = w)$ at each given wind direction w. Specifically, we use the Nadaraya–Watson kernel estimator (NP) [33]:

$$\hat{m}_{ij}(x, w) = \frac{\sum_{t=1}^{n_{ij}} K_h(x - X_{ijt}) Y_{ijt} I(W_{ijt} = w)}{\sum_{t=1}^{n_{ij}} K_h(x - X_{ijt}) I(W_{ijt} = w)}, \tag{4.4}$$

where $I(W_{ijt} = w)$ is the indicator function for a wind direction, $K_h(z) = (K(z_1/h_1) \cdots K(z_q/h_q)/h_1 h_2 \cdots h_q)$ is a product kernel generated by a univariate kernel function $K(\cdot)$ and q is the dimension of continuous variable X_{ijt}. The smoothing bandwidths are $h_1, \ldots, h_q$. Throughout the paper, the Gaussian kernel $K(u) = (2\pi)^{-1/2} \exp(-u^2/2)$ is used. The bandwidths reflect different scales in the meteorological variables which contribute to the response. The local linear kernel estimator [35] can be employed as well, without changing the methodology proposed. We use cross-validation [33] to find suitable smoothing bandwidths.

The partially linear model (4.3) can be estimated using a combination of kernel smoothing and least-squares regression, where the smoothing bandwidths can be obtained by a version of cross-validation (see [36] for details).

Table 1 reports the monthly RMSEs obtained by fitting models (4.2) and (4.3) to each monthly sample. It also reports the raw standard deviation of $PM_{2.5}$ for each month. The table shows a huge reduction in the raw RMSE by fitting the non-parametric model (4.2). The average percentage of reduction for the 60 months was 78.2% with a standard deviation of 8.3%. The partially linear model (4.3) further reduced the RMSE of (4.2) by an average of 45.8% with a standard deviation 16.5%.

For the desired adjustment to the mean and the quantiles of the $PM_{2.5}$ distribution, model (4.2) is more useful than the auto-regressive partially linear model (4.3). The latter is needed when building more specific models for forecasting the $PM_{2.5}$ level. The rationale in presenting model (4.3) and its fitting results is to show that adding the auto-regressive part can improve the fit to the data.

The bandwidths obtained by the cross-validation for each monthly model (4.2) contain information on the importance of variables in explaining the $PM_{2.5}$ concentration. If a covariate is redundant, the bandwidth selected by the cross-validation will diverge to the upper bound of the allowable range with probability tending to 1 (see [37] for details). As all the meteorological variables, except the wind direction, are continuous, the upper bound is infinity.

We checked on the cross-validation bandwidths selected under each of the four wind directions (NW, NE, S and CV) for each month and employed 15 000 as the threshold to judge whether a variable is redundant or not. The use of 15 000 was made by the observation that the CV bandwidths were either small, in the range of 0.2–100 (mostly from 0.2 to 10), or above 15 000. The electronic supplementary material, figure S4, reports the frequency of each variable which was selected for being useful among the 60 months under each wind direction. It shows that dew point and pressure were the most influential, followed by temperature and CWP. Rain and snow were significant in the summer and winter, respectively. It is not surprising to see that CWP was less influential under the CV (calm and variable wind) than the other wind directions, as it is hard to cumulate for this wind type.

There are six covariates in model (4.2). As rightly raised by a referee, the curse of dimensionality may be encountered in the kernel estimation of the regression surface, despite having around 720 observations per month. We have tried to develop more specific parametric or semi-parametric models than (4.2) for the relationship between $PM_{2.5}$ readings and the meteorological variables. However, it is quite challenging to obtain the relationship, as $PM_{2.5}$ values are highly dependent on the wind direction and are highly nonlinear with respect to the meteorological variables, and

Table 1. RMSEs by fitting the non-parametric regression (NP) model (4.2) and the partially linear regression (PL) model (4.3), for each month in 2010–2014. The standard deviation without any models for the monthly samples is reported in the columns headed 'raw'.

(a)

month	2010			2011			2012		
	raw	NP	PL	raw	NP	PL	raw	NP	PL
1	93.94	10.07	5.42	46.31	8.26	7.42	131.47	8.44	7.31
2	84.83	34.86	26.47	143.18	24.50	10.67	78.39	13.22	5.81
3	84.29	13.58	6.91	73.74	8.02	6.10	86.69	14.41	10.43
4	73.52	4.55	2.92	67.01	11.17	13.41	69.12	11.36	5.83
5	59.04	10.86	6.97	51.59	6.94	8.30	56.00	20.13	14.70
6	52.26	11.00	9.36	76.05	20.15	10.24	68.50	15.48	8.51
7	72.49	16.00	10.39	80.13	16.99	8.74	56.67	16.84	6.58
8	67.32	9.90	8.01	54.27	21.48	8.40	59.20	21.19	4.63
9	78.43	28.44	12.57	85.13	26.36	8.24	52.43	11.70	5.13
10	124.59	21.45	11.05	122.91	20.37	10.49	92.44	30.96	11.40
11	133.69	41.00	9.33	89.62	25.35	13.65	85.21	30.38	12.05
12	114.89	23.83	11.69	107.23	11.85	8.90	96.97	5.81	5.84

(b)

month	2013			2014		
	raw	NP	PL	raw	NP	PL
1	168.95	36.74	16.50	110.98	26.34	17.05
2	117.27	25.44	15.33	145.35	14.83	8.94
3	104.80	16.26	7.40	97.73	9.44	4.09
4	58.60	6.24	5.59	57.58	13.67	6.29
5	55.85	13.75	6.75	45.03	10.44	9.64
6	68.76	16.37	8.04	41.81	9.64	6.10
7	43.67	11.89	5.07	65.05	13.76	6.15
8	40.59	9.11	5.78	44.48	14.31	8.56
9	65.11	21.38	11.22	47.83	10.74	5.73
10	95.05	20.41	8.53	118.08	28.11	6.58
11	92.56	19.41	10.38	109.71	23.78	11.12
12	107.36	22.72	15.93	93.88	23.18	12.16

there is significant confounding among the covariates too. We note that, for the purpose of the air quality assessment, a parametric or semi-parametric model may not be necessary, as the ultimate purpose of the kernel smoothing is to obtain the adjusted means and quantiles rather than the regression function. Hence, the kernel smoothing and the associated curse of dimensionality will have fewer effects on the inference of the adjusted means and quantiles. Having said the above, the non-parametric model (4.2) should be viewed as the first step in our model building exercise as it provides a benchmark for further development of more specific models. Linear or additive models based on functional principal component analysis [38,39] can be applied to reveal more

explicit association between the $PM_{2.5}$ process and meteorological covariates. The development of more interpretable models is an area of our future research.

5. Adjustment to meteorological conditions

As a completely randomized experiment that controls the weather is not possible, the observed $PM_{2.5}$ has to be adjusted with respect to the observed meteorological conditions.

We first present the adjustment to the monthly means of $PM_{2.5}$ with respect to the meteorological conditions for the 5 years. Let $f_{.j}(x,w)$ be the joint density function of the meteorological variables (X_{ijt}, W_{ijt}) in month j of all years. The adjusted mean $PM_{2.5}$ in month j and year i is

$$
\mu_{ij} = \sum_{w=1}^{4} \int E(Y_{ijt}|X_{ijt} = x, W_{ijt} = w) f_{.j}(x,w) \, dx
$$

$$
= \sum_{w=1}^{4} \int m_{ij}(x,w) f_{.j}(x,w) \, dx. \tag{5.1}
$$

Substituting the kernel estimator $\hat{m}_{ij}(x,w)$ given in (4.4), the estimator of the adjusted mean is

$$
\hat{\mu}_{ij} = \left(\sum_{a=1}^{n_{.j}} n_{aj} \right)^{-1} \sum_{a=1}^{n_{.j}} \sum_{t=1}^{n_{aj}} \sum_{w=1}^{4} \hat{m}_{ij}(X_{ajt}, W_{ajt}) I(W_{ajt} = w), \tag{5.2}
$$

where $n_{.j}$ is the number of years that month j is observed, which is 5 for our analysis. It can be shown that $\hat{\mu}_{ij}$ is a consistent and asymptotically unbiased estimator of the true μ_{ij}, using a similar technique to [40].

The essence of the adjustment is to use the data in the jth month of year i to estimate the regression function m_{ij}, and then substitute the meteorological information of all 5 years for month j to obtain the estimator of μ_{ij}, as suggested by (5.2). This is the approach used in observational studies [31] for carrying out adjustment based on covariates to a response so as to remove the bias due to a lack of randomness in the study.

As the observed data are dependent time series, the block bootstrap method [41] is needed to estimate the variance of $\hat{\mu}_{ij}$. The block bootstrap resamples blocks of observations to retain the dependence in the original data series. For each month j in year i, let $Z_t = (X_{ijt}, W_{ijt})$, $t = 1, \ldots, T = n_{ij}$. We considered moving blocks with length l: $B_1 = (Z_1, \ldots, Z_l), \ldots, B_{T-l+1} = (Z_{T-l+1}, \ldots, Z_T)$, $B_{T-l+2} = (Z_{T-l+2}, \ldots, Z_T, Z_1), \ldots, B_T = (Z_T, Z_1, \ldots, Z_{l-1})$. The wrapping at the boundary ensures that each of the original observations appears with equal chance in a bootstrapped sample. Then we independently resampled T/l blocks from a total of T blocks, which were joined together to form a resampled monthly times series $\{X_{ijt}^b, W_{ijt}^b\}$ for the bth replication of month j in year i.

Based on the resampled weather variables $\{(X_{ijt}^b, W_{ijt}^b)\}$, we impute the corresponding $PM_{2.5}$ via an estimated model (4.2): $Y_{ijt}^{b*} = \hat{m}_{ij}(X_{ijt}^b, W_{ijt}^b) + \hat{\epsilon}_{ijt}^{b*}$, where $\hat{\epsilon}_{ijt}^{b*}$ is resampled from a two-point distribution [42],

$$
\hat{\epsilon}_{ijt}^{b*} = \begin{cases} \hat{\epsilon}(X_{ijt}^b, W_{ijt}^b) \dfrac{1-\sqrt{5}}{2} & \text{with probability} \quad \dfrac{1+\sqrt{5}}{2\sqrt{5}} \\[2ex] \hat{\epsilon}(X_{ijt}^b, W_{ijt}^b) \dfrac{1+\sqrt{5}}{2} & \text{with probability} \quad 1 - \dfrac{1+\sqrt{5}}{2\sqrt{5}}, \end{cases} \tag{5.3}
$$

and $\hat{\epsilon}(X_{ijt}^b, W_{ijt}^b)$ is a residual estimator via a kernel smoothing of $\hat{\epsilon}_{ijt}^2 = \{Y_{ijt} - \hat{m}_{ij}(X_{ijt}, W_{ijt})\}^2$

$$
\hat{\epsilon}^2(x,w) = \frac{\sum_{t=1}^{n_{ij}} K_h(x - X_{ijt}) \hat{\epsilon}_{ijt}^2 I(W_{ijt} = w)}{\sum_{t=1}^{n_{ij}} K_h(x - X_{ijt}) I(W_{ijt} = w)}. \tag{5.4}
$$

Matching the resampled meteorological data and the imputed $PM_{2.5}$ gives us the bootstrap resampled data series $\{(Y_{ijt}^{b*}, X_{ijt}^{b}, W_{ijt}^{b})\}$ for $i = 1, \ldots, 5, t = 1, \ldots, n_{ij}$. We re-estimate the regression function to obtain $\hat{m}_{ij}^{b}(x, w)$ by (4.4), which then gives rise to the adjusted mean for the bth bootstrap replication

$$\hat{\mu}_{ij}^{b} = \left(\sum_{a=1}^{n_{.j}} n_{aj} \right)^{-1} \sum_{a=1}^{n_{.j}} \sum_{t=1}^{n_{aj}} \sum_{w=1}^{4} \hat{m}_{ij}^{b}(X_{ajt}^{b}, W_{ajt}^{b}) I(W_{ajt}^{b} = w). \tag{5.5}$$

The bootstrap estimate of the standard deviation of $\hat{\mu}_{ij}$ is

$$\hat{\sigma}_{ij} = \sqrt{\frac{\sum_{b=1}^{B}(\hat{\mu}_{ij}^{b} - \hat{\mu}_{ij})^2}{B-1}}. \tag{5.6}$$

Similarly to the adjusted mean in (5.1), we define the adjusted distribution function of $PM_{2.5}$ for month j of year i as

$$G_{ij}(y) = \sum_{w=1}^{4} \int F_{ij}(y|x, w) f_{.j}(x, w) \, dx,$$

where $F_{ij}(y|x, w) = E(I\{Y_{ijt} \leq y\}|X_{ijt} = x, W_{ijt} = w)$ is the conditional distribution and $I\{Y_{ijt} \leq y\}$ is the indicator function. In order to estimate $G_{ij}(y)$, we first estimate $F_{ij}(y|x, w)$ by the kernel smoothing method under each wind direction:

$$\hat{F}_{ij}(y|x, w) = \frac{\sum_{t=1}^{n_{ij}} K_h(x - X_{ijt}) I(W_{ijt} = w) G_{h_0}(Y_{ijt} - y)}{\sum_{t=1}^{n_{ij}} K_h(x - X_{ijt}) I(W_{ijt} = w)},$$

where $G_{h_0}(x) = \int_{-\infty}^{x/h_0} K(u) \, du$ is the integration of the univariate kernel K. Then, we estimate $G_{ij}(y)$ by

$$\hat{G}_{ij}(y) = \left(\sum_{a=1}^{n_{.j}} n_{aj} \right)^{-1} \sum_{a=1}^{n_{.j}} \sum_{t=1}^{n_{aj}} \sum_{w=1}^{4} \hat{F}_{ij}(y|X_{ajt}, W_{ajt}) I(W_{ajt} = w).$$

For any $q \in (0, 1)$, the adjusted qth percentile is estimated by $\hat{G}_{ij}^{-1}(q)$. The standard error of the estimated percentiles can be obtained by mimicking the block bootstrap procedure for the adjusted mean.

Let us consider how to measure the effect of emissions by revisiting model (4.1). Suppose that the regression function in (4.1) is additive with respect to the meteorological variable (X_{ijt}, W_{ijt}) and the emission level U_{ijt}, such that

$$Y_{ijt} = \tilde{m}_{ij,1}(X_{ijt}, W_{ijt}) + \tilde{m}_{ij,2}(U_{ijt}) + \epsilon_{ijt}.$$

If the emission U_{ijt} is independent of weather conditions (X_{ijt}, W_{ijt}), it can be shown that

$$m_{ij}(x, w) = E(Y_{ijt}|X_{ijt} = x, W_{ijt} = w) = \tilde{m}_{ij,1}(x, w) + E\{m_{ij,2}(U_{ijt})\}$$

and the adjusted mean in (5.1) can be written as

$$\mu_{ij} = \sum_{w=1}^{4} \int E(Y_{ijt}|X_{ijt} = x, W_{ijt} = w) f_{.j}(x, w) \, dx$$

$$= \sum_{w=1}^{4} \int \tilde{m}_{ij,1}(x, w) f_{.j}(x, w) \, dx + E\{m_{ij,2}(U_{ijt})\}. \tag{5.7}$$

The result in (5.7) implies that, under the additive and independence assumptions, the adjusted average $PM_{2.5}$ is the sum of two terms: one due to weather conditions and the other due to emissions. This may be used to obtain the effect of emission by taking the difference in μ_{ij}. For instance, if the weather impact in January is the same among all the years considered, then $\mu_{i,1} - \mu_{j,1}$ will reflect the different emission levels between years i and j on $PM_{2.5}$. Likewise, if the

weather impacts between two neighbouring time periods are roughly the same, the difference in the adjustment means informs the difference in the effects of emissions between the two periods. The latter approach is used in our studies on the impacts of the emission control for the Asia-Pacific Economic Cooperation (APEC) meeting and winter heating in §7.

We note here that the emission level U_{ijt} does not measure aerosol abundance, but the original emission level directly related to energy consumption, as otherwise the assumption of it being independent of the meteorological variable would be unreasonable, as secondary aerosols are impacted by weather conditions. Of course, the independent assumption does not hold in a longer time scale, as the emission level has an annual cycle. However, for the monthly time frame that we are considering the assumption is reasonable.

6. Five-year assessment

We applied the adjustment on the mean and the quantiles of the $PM_{2.5}$ with respect to the meteorological variables outlined in the previous section to the 60 months in the years 2010–2014. Figure 4a presents the monthly adjusted and raw averages of $PM_{2.5}$, along with 95% confidence intervals for the adjusted means based on the standard errors obtained via the block bootstrap with block size $l = 12$ h. The confidence intervals were obtained by extending 1.96 times the standard deviation above and below the adjusted mean. We observe substantial differences between the adjusted and the raw averages. Among the 60 months in the 5 years, 28 months had raw averages outside the intervals. This demonstrates that it is necessary to adjust the averages as those without any adjustments would be unfairly impacted by the weather.

As more robust alternatives to the mean, figure 4b gives the adjusted percentiles of monthly $PM_{2.5}$ at 90th, 75th, 50th (median), 25th and 10th percentiles, along with their 95% CI obtained via the block bootstrap method with $l = 12$. It is observed that there were much smaller monthly variations for the three lower percentiles. The variation increased in the two higher percentiles. We find that the medians were significantly above the threshold 35 $\mu g\,m^{-3}$ in 56 months and were mostly ranged between 70 and 100 $\mu g\,m^{-3}$.

The monthly 75th percentiles were significantly higher than 90 $\mu g\,m^{-3}$ in all but three months. The pattern displayed by the 90th and 75th percentiles showed a much higher level of $PM_{2.5}$ pollution from October to March. This reflects the extra emissions due to the winter heating season in North China from November to March [16], and the biomass burning by farmers in October [13]. There are also increased mineral aerosols due to transported dust in winter and spring [43].

The State Council of China set $PM_{2.5}$ reduction targets for various regions of China in October 2013. Those for Beijing and NCP are (i) a 25% reduction by 2017 from the 2012 level and (ii) an annual average $PM_{2.5}$ in Beijing below 60 $\mu g\,m^{-3}$.

To check on progress towards the reduction targets, figure 5 gives the differences in adjusted averages, medians and 90th percentiles between 2013 and 2012 and between 2014 and 2012. The exact numerical values are given in the electronic supplementary material, table S1. Figure 5 reveals that the reduction in the average $PM_{2.5}$ from 2012 happened in two months in 2013, and only one of them (August) was significant at 2.5% ($p < 0.025$). Six months in 2014 had reductions in averages over 2012, and only two of them (May and June) were significant at 2.5%. In contrast, among the 10 months in 2013 which had increased averages over their 2012 counterparts, four months' increases were significant at 2.5%. In 2014, among the six months with higher averages, four of them were significantly higher than their counterparts in 2012 at 2.5%. We note that most of the increases in 2013 and 2014 were much larger in magnitude than most of the decreases in the 2 years.

The situation for the medians was similar to that of the averages reported above. Only two months in 2014 had a significantly smaller median than 2012, and five months had a significantly larger median at 2.5% significance. In 2013, there was no significant reduction, whereas significant increases happened in two months. There were improvements about the 90th percentiles in three months in year 2014 and one month in 2013. However, significant increases in the 90th percentiles
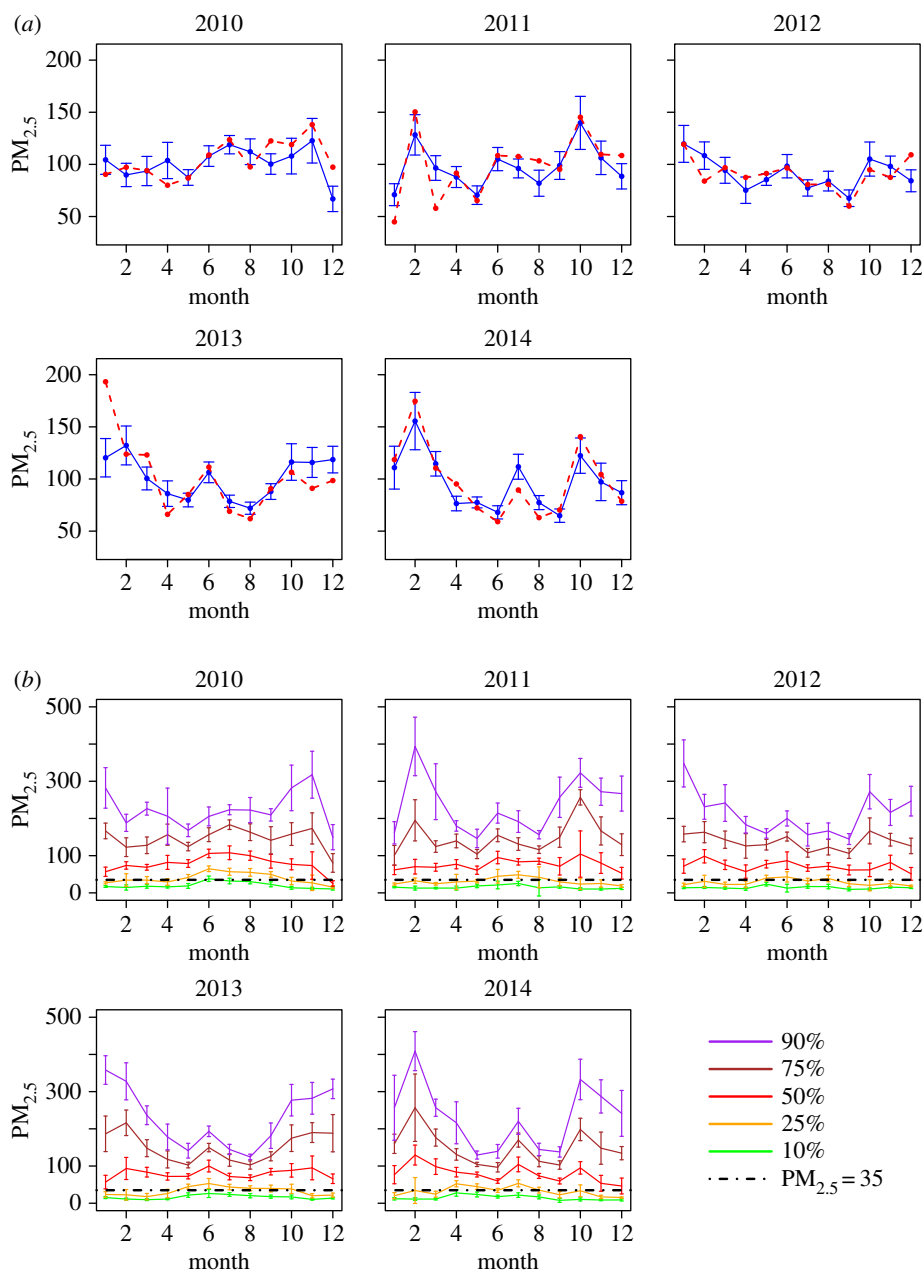
Figure 4. Adjusted average and percentiles of $PM_{2.5}$. (*a*) The adjusted monthly averages (solid lines) and raw averages (dashed lines). (*b*) The adjusted monthly 90th (purple), 75th (brown), 50th (median, red), 25th (yellow) and 10th (green) percentiles of $PM_{2.5}$ concentration with the black dashed line of $PM_{2.5} = 35 \, \mu g \, m^{-3}$. The bars are 1.96 times the standard deviations above and below the estimates.

happened in four months in both 2013 and 2014. Moreover, the adjusted annual averages (standard error) from 2010 to 2014 were 101.3 $\mu g \, m^{-3}$ (1.99), 97.6 $\mu g \, m^{-3}$ (2.08), 91.5 $\mu g \, m^{-3}$ (1.72), 101.2 $\mu g \, m^{-3}$ (1.85) and 96.9 $\mu g \, m^{-3}$ (2.09), respectively, indicating that the $PM_{2.5}$ pollution in Beijing has actually got worse since 2012. The electronic supplementary material, table S2, gives the annual adjusted means and percentiles and their standard errors. The percentiles in 2013 and 2014 were all significantly higher than their 2012 counterparts except the 10th percentile in 2014, lending support to the above conclusion based on the averages.
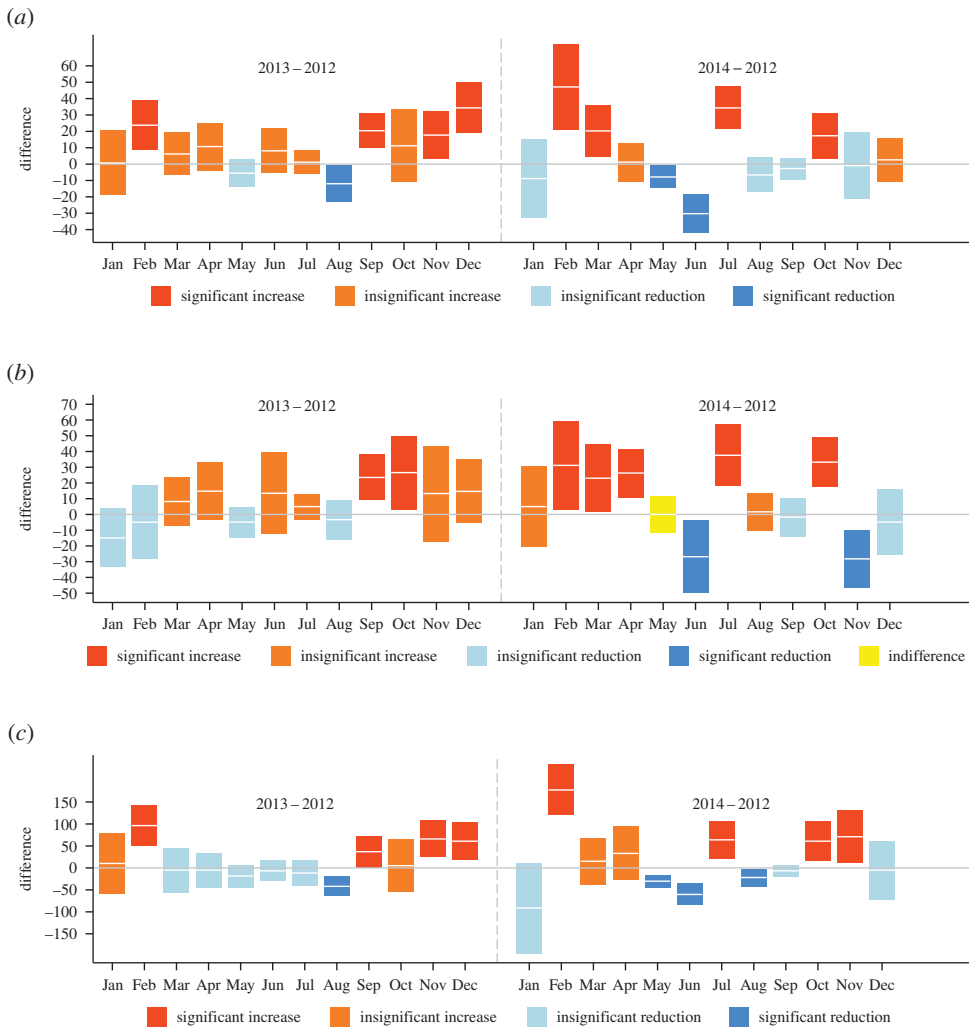
Figure 5. Differences in PM$_{2.5}$ between 2013 and 2012, and between 2014 and 2012. (*a*) Differences in the adjusted averages. (*b*) Differences in the adjusted medians. (*c*) Differences in the adjusted 90th percentiles. 'Difference' means the 2013/2014 value of a month minus the 2012 value of the same month. The boxes are 1.96 times the standard deviations above and below the differences (white line). Significant increases (decreases) correspond to boxes which are entirely above (below) the horizontal line of zero; and insignificant increases (decreases) correspond to boxes which intercept the horizontal line. A significant increase (reduction) means the *p*-value is less than 0.025 for the one-sided test for the difference being positive (negative).

7. Two quasi-experiments

QEs [26,44] may be used in conjunction with observational studies to gauge the effect of the emissions U_{ijt}, information about which is not available. We have two opportunities for QEs. One was the 2014 APEC meeting in Beijing on 8–11 November. The second QE is the annual winter heating season which runs usually from 15 November to 15 March in Beijing and the NCP. These two experiments offer opportunities to assess the impacts of emissions on PM$_{2.5}$ and provide potential policy options to combat the pollution.

To ensure the air quality for the APEC meeting, the Chinese government had ordered a temporary closure of factories in Beijing and the northern part of the NCP from 3 to 12 November, with half of the cars in Beijing being ordered to stay off the roads according to the last digit of the licence plate being even or odd. From 6 to 12 November, the control zone was enlarged to include
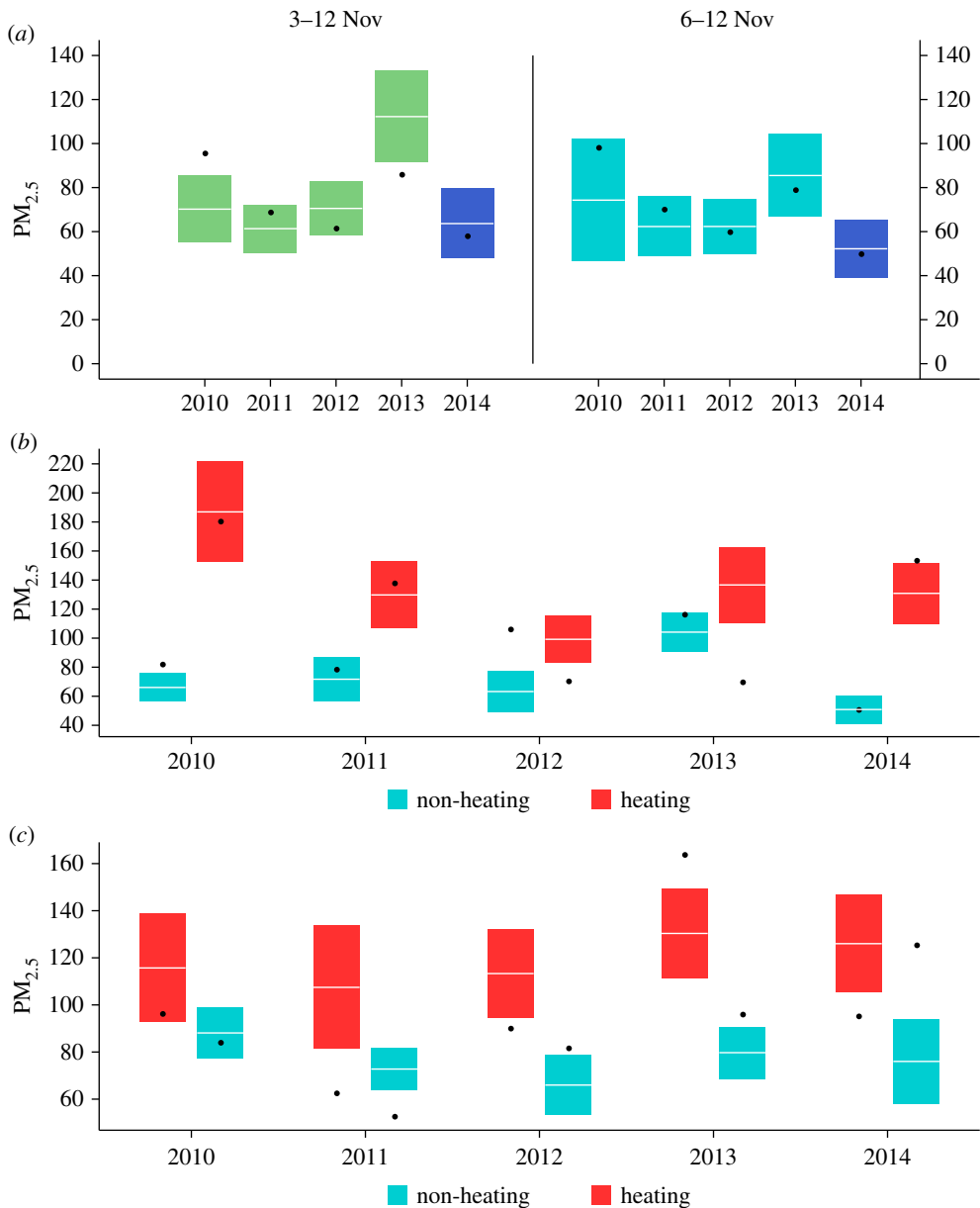
Figure 6. Adjusted averages in the QEs. (*a*) Adjusted averages in the periods of APEC. (*b*) Adjusted averages in the non-heating and heating periods in November. (*c*) Adjusted averages in the non-heating and heating periods in March. The boxes are 1.96 times the standard deviations above and below the estimates (white line). The black dots are the averages without the adjustment.

the entire NCP and part of the neighbouring Shanxi and Inner Mongolia provinces. Schools and public offices were closed in this second period until 12 November with much suppressed economic and social activities. To evaluate the impacts of the APEC measures on Beijing's air quality, we conducted the proposed adjustment to the average $PM_{2.5}$ levels for the two time periods, 3–12 November and 6–12 November, by considering the meteorological conditions over the two periods in the 5 years from 2010 to 2014. The left panel of figure 6*a* shows that the 2014 average in the first period of 3–12 November was significantly less than the averages in 2013 with the *p*-value being almost zero. However, the reduction in 2014 was not statistically significant

at the 5% level when compared with the 2010, 2011 and 2012 averages (the p-values were 0.28, 0.61, 0.25, respectively). For the second period of 6–12 November (the right panel in figure 6a), the p-values for testing the adjusted mean in 2014 being less than the corresponding periods in 2010–2013 were 0.06, 0.11, 0.07 and 0, respectively, which were much lower than those in the first period. Although only the p-value when comparing with 2013 was significant at 5%, the p-values for the other three years were only slightly higher than 0.05, which reflected the large variation in the weather conditions. Thus, the administrative actions to enlarge the control zone on emissions did have some effect in reducing the $PM_{2.5}$ level in the second period. The $PM_{2.5}$ reduction was averaged at 25.3% compared with the previous four years; see electronic supplementary material, table S3, for detailed numerical values about the averages and percentiles. That the APEC emission control measures were more effective in reducing the pollution in the second time period may be due to the fact that it takes time for the raw pollutants to become $PM_{2.5}$ in secondary reactions.

For the second QE on the effect of heating, we chose the two weeks from the start of the heating date in November and the two weeks just prior to the termination of heating in March as the treatment periods. As controls (non-heating periods), we chose the two weeks just before the start of heating in November and the two weeks right after the end of heating in March. The heating period can start earlier than 15 November or be extended after 15 March depending on the temperature at that time. For instance, it was started on 3 November in 2012; and ended on 22, 18, 17 March in 2010, 2012 and 2013, respectively. Figure 6b,c displays the pairwise adjusted averages for the heating and non-heating periods in November and March, respectively, for the last 5 years. The figures show startling increases in $PM_{2.5}$ due to heating in both November and March, with the increases being highly statistically significant in all heating seasons (the p-values were all ≤ 0.015). The increases in November due to heating ranged from 31% (in 2013) to 183% (in 2010). The increase of 157% in 2014 was partly due to the APEC measures and should not be taken as a norm. The reduction in March due to the cessation of heating ranged from 24% to 42%. In other words, the increase due to heating in March ranged from 31% to 72%. The electronic supplementary material, tables S4 and S5, gives the exact numeric values for the adjusted means and percentiles for each heating and non-heating period, which strongly support heating having a significant effect on $PM_{2.5}$ in Beijing. The table also reports the original unadjusted averages and percentiles for the heating treatment and the non-heating control. This shows that the effect of heating would not have been revealed in many seasons if the adjustments were not carried out.

8. Incorporating other chemicals

So far our analysis is solely based on the dependence between $PM_{2.5}$ and the meteorological variables without considering the atmospheric chemical reaction that generates $PM_{2.5}$. As rightly pointed out by a referee, urban aerosol formation is significantly contributed to by secondary formation processes as elaborated in [8]. To extend our analysis to include potential secondary formation, we face a hurdle in that the US Embassy only measures the $PM_{2.5}$ concentration without other chemical compounds. To get around the problem, we consider hourly recordings of three pollutants—SO_2, NO_2 and CO—at a neighbouring MEP site, Nongzhanguan, which is 1.2 km south of the embassy. Measurements of more extended chemical elements are not available in most of the MEP sites, except O_3. Our preliminary analysis reveals that O_3 has little explaining power for $PM_{2.5}$. Hence, we did not include it in the analysis. Although the MEP site at Nongzhanguan had started measuring $PM_{2.5}$ and the chemical compounds in early 2013, the data in 2013 suffered from a high percentage of missing values. Therefore, we only analysed the 2014 data.

We consider adding a linear term for the three chemicals to the non-parametric model (4.2) and the auto-regressive model (4.3):

$$Y_{ijt} = \beta_1 SO_{2ijt} + \beta_2 NO_{2ijt} + \beta_3 CO_{ijt} + m_{ij}(X_{ijt}, W_{ijt}) + e_{ijt}, \quad t = 1, \ldots, n_{ij} \tag{8.1}$$

Table 2. RMSEs by fitting the non-parametric model (4.2), the non-parametric auto-regressive model (4.3) and models (8.1) and (8.2) by adding a linear regressive part for the three chemicals, respectively, for each month in 2014. The column headed 'raw' is the monthly sample standard deviation of $PM_{2.5}$ without any model.

year-month	RMSE					RMSE reduction (%)			
	raw	(4.2)	(8.1)	(4.3)	(8.2)	(4.2)	(8.1)	(4.3)	(8.2)
2014-1	110.98	26.34	18.04	17.05	15.10	76.26	83.75	84.63	86.39
2014-2	145.35	14.83	12.16	8.94	7.78	89.80	91.63	93.85	94.65
2014-3	97.73	9.44	3.76	4.09	3.12	90.34	96.16	95.81	96.81
2014-4	57.58	13.67	6.50	6.29	5.01	76.26	88.71	89.08	91.30
2014-5	45.03	10.44	11.84	9.64	9.03	76.81	73.72	78.59	79.94
2014-6	41.81	9.64	7.95	6.10	4.97	76.95	80.99	85.41	88.10
2014-7	65.05	13.76	9.63	6.15	5.77	78.85	85.20	90.55	91.12
2014-8	44.48	14.31	9.01	8.56	7.43	67.84	79.74	80.76	83.29
2014-9	47.83	10.74	8.39	5.73	5.33	77.55	82.47	88.02	88.85
2014-10	118.08	28.11	13.13	6.58	5.56	76.20	88.88	94.43	95.29
2014-11	109.71	23.78	13.72	11.12	10.80	78.32	87.50	89.87	90.16
2014-12	93.88	23.18	11.76	12.16	10.34	75.31	87.47	87.05	88.99
average	81.46	16.52	10.49	8.53	7.52	78.37	85.52	88.17	89.57

and

$$Y_{ijt} = \beta_0 Y_{ij,t-1} + \beta_1 SO_{2ijt} + \beta_2 NO_{2ijt} + \beta_3 CO_{ijt} + g_{ij}(X_{ijt}, W_{ijt}) + \eta_{ijt}. \qquad (8.2)$$

We use the parametric linear function to avoid further issues with the curse of dimensionality.

Table 2 reports RMSEs for fitting models (4.2), (4.3), (8.1) and (8.2) and the percentages of reduction in relative RMSEs of the four models. We do not report the R^2-values of the models as they were all above 0.93, which can exaggerate the goodness-of-fit of models. The table shows that the meteorological variables explained on average 78% of the uncertainty, as measured by the RMSEs, of the $PM_{2.5}$ as secondary formation of $PM_{2.5}$ requires certain meteorological conditions. The RMSEs were further reduced for models (8.1) and (8.2) of the order of 8–10%, indicating that more uncertainty was explained by incorporating the three chemicals. The relative amount of reductions provides a measure of the uncertainty missed by considering only the meteorological variables. In addition, the reductions from (4.3) to (8.2) were less than those from (4.2) to (8.1). This was because the lagged $PM_{2.5}$ term (the auto-regressive part) in (4.3) and (8.2) already contained information about the three chemicals.

The estimates of the regression coefficients of the three chemical elements together with their p-values [45] for being significantly different from zero are reported in the electronic supplementary material, tables S6–S8. The results show that, for model (8.1), SO_2 tended to be significant in the winter months, and NO_2 and CO were significant in almost all the months of the year. The significance of SO_2 in winter was likely to be due to the excessive coal burning for heating. The significance of NO_2 and CO for the whole year indicates that the pollution from motor vehicles plays an important role in air pollution in Beijing, which cannot be captured fully by considering meteorological conditions alone. The fitting results for model (8.2) in the electronic supplementary material, table S8, show that, when the lagged $PM_{2.5}$ was added to the model, the three chemicals became less significant, which was consistent with the findings in table 2.

9. Discussion

By employing statistical learning based on 5 years' data, we propose a set of statistical measures to quantify the severity of $PM_{2.5}$ pollution in Beijing. We provide a methodological framework to produce adjusted means and percentiles for objectively comparing $PM_{2.5}$ concentrations that neutralize the impacts of meteorological conditions. The adjustment allows us to find significant effects of the emission reduction measures for the APEC meeting and annual winter heating on the $PM_{2.5}$. Our analyses have the following conclusions and broad implications.

All our analyses point to a conclusion that, up to the end of 2014, the $PM_{2.5}$ pollution in Beijing had not improved over the 2012 levels, and the air quality had in fact got worse.

Our analysis of the effect of heating on $PM_{2.5}$ reveals that winter heating contributes significantly to $PM_{2.5}$ abundance in Beijing. A step that various authorities in the NCP can take is to phase out the use of coal for heating and convert all the furnaces in cities and towns to natural gas. However, this action (even though it may be perceived as too radical) alone is not enough for eliminating the pollution. To appreciate this point, we note from our analysis that the heating has contributed a more than 50% increase (on average) in $PM_{2.5}$ in the winter months in Beijing since 2010. This means that one-third of the $PM_{2.5}$ in the heating months is due to heating. This leads to a best case scenario where using natural gas or other cleaner energy for heating in the NCP at best reduces the average $PM_{2.5}$ in the months of December, January and February by one-third. At the same time, we use the averages in the non-heating portion of November and March as the monthly average while keeping the other months' $PM_{2.5}$ levels the same. Then, the annual averages for 2013 and 2014 would be 88.2 $\mu g\,m^{-3}$ and 80.1 $\mu g\,m^{-3}$, respectively, which represent only 3.6% and 12.5% reductions, respectively, from the 2012 annual average of 91.5 $\mu g\,m^{-3}$. These two numbers are still well short of the 25% reduction target of 68.6 $\mu g\,m^{-3}$.

Despite the drastic APEC measures which basically shut down a significant portion of the economy in the NCP for 10 days, the average $PM_{2.5}$ in the period 6–12 November 2014 was 52.2 $\mu g\,m^{-3}$ (6.72) (electronic supplementary material, table S3), which was still much higher than the healthy threshold of 35 $\mu g\,m^{-3}$. This indicates that, under the existing energy consumption profile of the NCP, it is rather unlikely that Beijing will attain clean air of 35 $\mu g\,m^{-3}$ or lower over a prolonged period. Hence, transitions to alternative forms of industrial installations with lower emission profiles must be taken in order for Beijing to attain the standard of healthy air.

References

1. Zhang X, Wang Y, Niu T, Zhang X, Gong S, Zhang Y, Sun J. 2012 Atmospheric aerosol compositions in China: spatial/temporal variability, chemical signature, regional haze distribution and comparisons with global aerosols. *Atmos. Chem. Phys.* **12**, 779–799. (doi:10.5194/acp-12-779-2012)
2. Guo S *et al*. 2014 Elucidating severe urban haze formation in China. *Proc. Natl Acad. Sci. USA* **111**, 17 373–17 378. (doi:10.1073/pnas.1419604111)

3. Huang K, Zhuang G, Wang Q, Fu J, Lin Y, Liu T, Han L, Deng C. 2014 Extreme haze pollution in Beijing during January 2013: chemical characteristics, formation mechanism and role of fog processing. *Atmos. Chem. Phys. Discuss.* **14**, 7517–7556. (doi:10.5194/acpd-14-7517-2014)

4. Donaldson K, Li X, MacNee W. 1998 Ultrafine (nanometre) particle mediated lung injury. *J. Aerosol Sci.* **29**, 553–560. (doi:10.1016/S0021-8502(97)00464-3)

5. Pope III CA, Burnett RT, Thun MJ, Calle EE, Krewski D, Ito K, Thurston GD. 2002 Lung cancer, cardiopulmonary mortality, and long-term exposure to fine particulate air pollution. *JAMA* **287**, 1132–1141. (doi:10.1001/jama.287.9.1132)

6. Schwartz J, Neas LM. 2000 Fine particles are more strongly associated than coarse particles with acute respiratory health effects in schoolchildren. *Epidemiology* **11**, 6–10. (doi:10.1097/00001648-200001000-00004)

7. Chen R, Zhao Z, Kan H. 2013 Heavy smog and hospital visits in Beijing, China. *Am. J. Respir. Crit. Care Med.* **188**, 1170–1171. (doi:10.1164/rccm.201304-0678LE)

8. Zhang R, Wang G, Guo S, Zamora ML, Ying Q, Lin Y, Wang W, Hu M, Wang Y. 2015 Formation of urban fine particulate matter. *Chem. Rev.* **115**, 3803–3855. (doi:10.1021/acs.chemrev.5b00067)

9. Zhang Y, Hu M, Zhong L, Wiedensohler A, Liu S, Andreae M, Wang W, Fan S. 2008 Regional integrated experiments on air quality over Pearl River Delta 2004 (PRIDE-PRD2004): overview. *Atmos. Environ.* **42**, 6157–6173. (doi:10.1016/j.atmosenv.2008.03.025)

10. Chang S-Y, Chou CC, Liu S, Zhang Y. 2013 The characteristics of $PM_{2.5}$ and its chemical compositions between different prevailing wind patterns in Guangzhou. *Aerosol. Air Qual. Res.* **13**, 1373–1383. (doi:10.4209/aaqr.2012.09.0253)

11. Li J, Song Y, Mao Y, Mao Z, Wu Y, Li M, Huang X, He Q, Hu M. 2014 Chemical characteristics and source apportionment of $PM_{2.5}$ during the harvest season in eastern China's agricultural regions. *Atmos. Environ.* **92**, 442–448. (doi:10.1016/j.atmosenv.2014.04.058)

12. Yang F, Tan J, Zhao Q, Du Z, He K, Ma Y, Duan F, Chen G. 2011 Characteristics of $PM_{2.5}$ speciation in representative megacities and across China. *Atmos. Chem. Phys.* **11**, 5207–5219. (doi:10.5194/acp-11-5207-2011)

13. Huang R-J et al. 2014 High secondary aerosol contribution to particulate pollution during haze events in China. *Nature* **514**, 218–222. (doi:10.1038/nature13774)

14. He H, Tie X, Zhang Q, Liu X, Gao Q, Li X, Gao Y. 2015 Analysis of the causes of heavy aerosol pollution in Beijing, China: a case study with the WRF-Chem model. *Particuology* **20**, 32–40. (doi:10.1016/j.partic.2014.06.004)

15. Uno I, Sugimoto N, Shimizu A, Yumimoto K, Hara Y, Wang Z. 2014 Record heavy $PM_{2.5}$ air pollution over China in January 2013: vertical and horizontal dimensions. *SOLA* **10**, 136–140. (doi:10.2151/sola.2014-028)

16. Wang L, Zhang N, Liu Z, Sun Y, Ji D, Wang Y. 2014 The influence of climate factors, meteorological conditions, and boundary-layer structure on severe haze pollution in the Beijing-Tianjin-Hebei region during January 2013. *Adv. Meteorol.* **2014**, 685971. (doi:10.1155/2014/685971)

17. Sun Y, Jiang Q, Wang Z, Fu P, Li J, Yang T, Yin Y. 2014 Investigation of the sources and evolution processes of severe haze pollution in Beijing in January 2013. *J. Geophys. Res.* **119**, 4380–4398. (doi:10.1002/2014JD021641)

18. Zhang Q, He K, Huo H. 2012 Policy: cleaning China's air. *Nature* **484**, 161–162. (doi:10.1038/484161a)

19. Wang JZ, Yang YQ, Wang YQ, Zhang GZ. 2013 Sand dust weather and its variation in China based on PLAM index tracking method. *J. Meteorol. Environ.* **29**, 92–97. (doi:10.3969/j.issn1673-503X.2013.05.015)

20. Liu X-H, Zhang Y, Cheng S-H, Xing J, Zhang Q, Streets DG, Jang C, Wang W-X, Hao J-M. 2010 Understanding of regional air pollution over China using CMAQ, part I. Performance evaluation and seasonal variation. *Atmos. Environ.* **44**, 2415–2426. (doi:10.1016/j.atmosenv.2010.03.035)

21. Liu X-H, Zhang Y, Xing J, Zhang Q, Wang K, Streets DG, Jang C, Wang W-X, Hao J-M. 2010 Understanding of regional air pollution over China using CMAQ, part II. Process analysis and sensitivity of ozone and particulate matter to precursor emissions. *Atmos. Environ.* **44**, 3719–3727. (doi:10.1016/j.atmosenv.2010.03.036)

22. Cressie N, Wikle CK. 2011 *Statistics for spatio-temporal data*. Hoboken, NJ: John Wiley & Sons.

23. Sahu SK, Gelfand AE, Holland DM. 2006 Spatio-temporal modeling of fine particulate matter. *J. Agric. Biol. Environ. Stat.* **11**, 61–86. (doi:10.1198/108571106X95746)

24. Dominici F, McDermott A, Zeger SL, Samet JM. 2002 On the use of generalized additive models in time-series studies of air pollution and health. *Am. J. Epidemiol.* **156**, 193–203. (doi:10.1093/aje/kwf062)
25. Smith RL, Kolenikov S, Cox LH. 2003 Spatiotemporal modeling of $PM_{2.5}$ data with missing values. *J. Geophys. Res.* **108**, 4–27. (doi:10.1029/2002JD002914)
26. Dominici F, Greenstone M, Sunstein CR. 2014 Particulate matter matters. *Science* **344**, 257–259. (doi:10.1126/science.1247348)
27. Chinese State Council. 2013 *Atmospheric pollution prevention and control action plan*. Beijing, China: People's Press. [In Chinese.]
28. Department of Energy Statisticas, National Bureau of Statistics, People's Republic of China. 2013 *China energy statistics yearbook*. Beijing, China: Chinese Statistics Press. [In Chinese.]
29. Choi J, Fuentes M, Reich BJ. 2009 Spatial–temporal association between fine particulate matter and daily mortality. *Comp. Statist. Data Anal.* **53**, 2989–3000. (doi:10.1016/j.csda.2008.05.018)
30. Tai AP, Mickley LJ, Jacob DJ. 2010 Correlations between fine particulate matter ($PM_{2.5}$) and meteorological variables in the United States: implications for the sensitivity of $PM_{2.5}$ to climate change. *Atmos. Environ.* **44**, 3976–3984. (doi:10.1016/j.atmosenv.2010.06.060)
31. Rosenbaum PR. 2002 *Observational studies*. New York, NY: Springer.
32. Alduchov OA, Eskridge RE. 1996 Improved magnus form approximation of saturation vapor pressure. *J. Appl. Meteorol.* **35**, 601–609. (doi:10.1175/1520-0450(1996)035<0601:IMFAOS>2.0.CO;2)
33. Härdle W. 1990 *Applied nonparametric regression*. Cambridge, UK: Cambridge University Press.
34. Bosq D. 1998 *Nonparametric statistics for stochastic processes*. New York, NY: Springer.
35. Fan J, Gijbels J. 1996 *Local polynomial modelling and its applications: monographs on statistics and applied probability*. London, UK: CRC Press.
36. Härdle W, Liang H, Gao J. 2000 *Partially linear models*. Heidelberg, Germany: Physica-Varlag.
37. Hall P, Li Q, Racine JS. 2007 Nonparametric estimation of regression functions in the presence of irrelevant regressors. *Rev. Econ. Stat.* **89**, 784–789. (doi:10.1162/rest.89.4.784)
38. Yao F, Müller HG, Wang JL. 2005 Functional linear regression analysis for longitudinal data. *Ann. Statist.* **33**, 2873–2903. (doi:10.1214/009053605000000660)
39. Müller HG, Yao F. 2008 Data functional additive models. *J. Am. Statist. Assoc.* **103**, 1534–1544. (doi:10.1198/016214508000000751)
40. Chen SX, Qin J, Tang CY. 2013 Mann–Whitney test with adjustments to pretreatment variables for missing values and observational study. *J. R. Stat. Soc. Ser. B* **75**, 81–102. (doi:10.1111/j.1467-9868.2012.01036.x)
41. Davison AC, Hinkley DV. 1997 *Bootstrap methods and their application*. Cambridge, UK: Cambridge University Press.
42. Härdle W, Mammen E. 1993 Comparing nonparametric versus parametric regression fits. *Ann. Stat.* **21**, 1926–1947. (doi:10.1214/aos/1176349403)
43. Zhang X *et al*. 2013 Factors contributing to haze and fog in China. *Chin. Sci. Bull.* **58**, 1178–1187. [In Chinese.] (doi:10.1360/972013-150)
44. Chen Y, Ebenstein A, Greenstone M, Li H. 2013 Evidence on the impact of sustained exposure to air pollution on life expectancy from China's Huai River policy. *Proc. Natl Acad. Sci. USA* **110**, 12 936–12 941. (doi:10.1073/pnas.1300018110)
45. You J, Chen G. 2004 Block external bootstrap in partially linear models with nonstationary strong mixing error terms. *Can. J. Statist.* **32**, 335–346. (doi:10.2307/3316019)