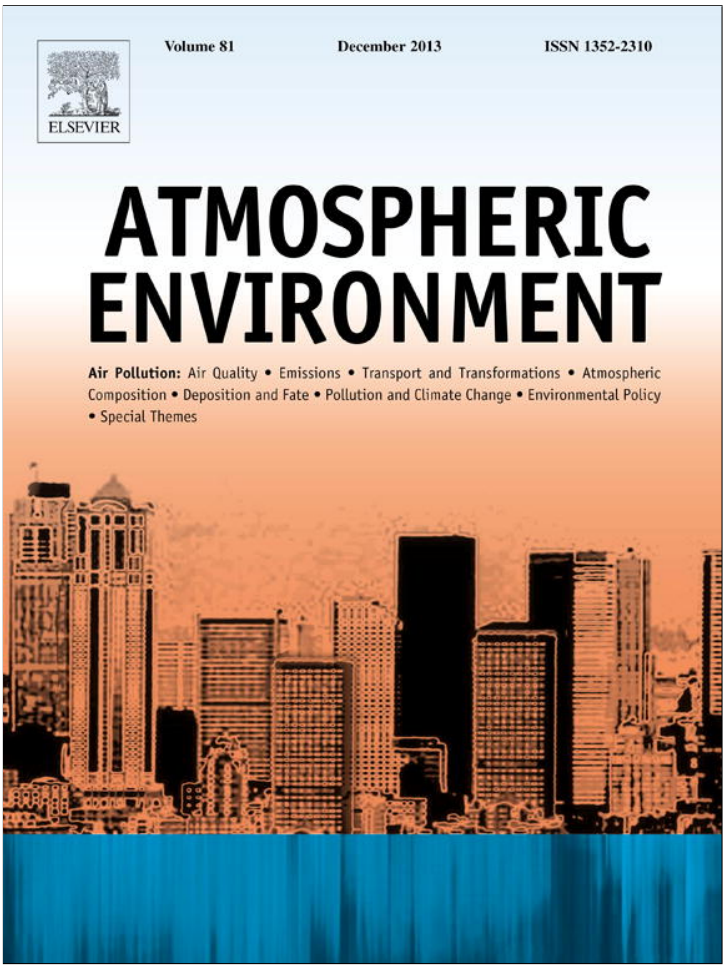

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Time-series analysis of mortality effects from airborne particulate matter size fractions in Beijing

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HIGHLIGHTS

- Feature of mortality impacts from PM size fractions is initial evaluated in Beijing.
- An obvious seasonal pattern of $PM_{2.5}$ acute effect has been found.
- Modifying effect of $PM_{2.5}$ by temperature exists in the study period.
- The level of RR assume an significantly tendency in recent years.

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ABSTRACT

Evidence concerning the health risk of fine and coarse particles is limited in developing Asian countries. The modifying effect between particles and temperature and season also remains unclear. Our study is one of the first to investigate the acute effect of particles size fractions, modifying effects and interannual variations of relative risk in a developing megacity where particulate levels are extraordinarily high compared to other Asian cities. After controlling for potential confounding, the results of a time-series analysis during the period 2005–2009 show that a 10 μg m^{-3} increase in $PM_{2.5}$ levels is associated with a 0.65% (95% CI: 0.29–0.80%), 0.63% (95% CI: 0.25–0.83%), and 1.38% (95% CI: 0.51–1.71%) increase in non-accidental mortality, respiratory mortality, and circulatory mortality, respectively, while a 10 μg m^{-3} increase in PM_{10} is similarly associated with increases of 0.15% (95% CI: 0.04–0.22%), 0.08% (95% CI: 0.01–0.18%), and 0.44% (95% CI: 0.12–0.63%). We did not find a significant effect of $PM_{2.5-10}$ on daily mortality outcomes. Our analyses conclude that temperature and particulates, exposures to both of which are expected to increase with climate change, might act together to worsen human health in Beijing, especially in the cool seasons. The level of the estimated percentage increase assume an escalating tendency during the study period, in addition to having a low value in 2008, and after the Olympic Games, the values increased significantly as the temporary atmospheric pollution control measures were terminated mostly.

1. Introduction

Adverse health effects of ambient airborne particles are of major concern to environmental health regulators. In recent years a tremendous amount of research on health effects of particles has been published and evidence is well established that even small concentrations of fine particles in air breathed by humans contribute significantly to their morbidity and mortality (Styer et al., 1995; Lee and Schwartz, 1999; Samet et al., 1997; Pope, 2000; Tie et al., 2009). A growing mass of compelling evidence has demonstrated the strong associations of acute effects between particulate matter (PM) and non-accidental mortality (NAM), respiratory mortality (RM), and circulatory mortality (CM) (Brunekreef and Holgate, 2002; Dominici et al., 2000, 2006; Yi et al., 2010; Chen et al., 2011; Lopez-Villarrubia et al., 2011). Most of these studies

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have shown that adverse health effects are consistently associated with PM_{10}, or inhalable particles (defined as particulate matter less than 10 μm in aerodynamic diameter). However, few studies have evaluated the association between $PM_{2.5-10}$ (particles with an aerodynamic diameter between 2.5 and 10 μm) and health outcomes (Health Effects Institute, 2004; Kan et al., 2007). $PM_{2.5}$ (particles with an aerodynamic diameter less than 2.5 μm) has only been studied recently in the developed countries, with only a small number of studies in Asian countries (Health Effects Institute, 2004). The pollution level, source, formation process, and composition of $PM_{2.5}$ may not be common to both PM_{10} and $PM_{2.5-10}$, and we hypothesizes that their biological effects and toxicities are also different. It is well known that the concentration of atmospheric pollutants in Beijing declined significantly and achieved the "Green Olympics" control goal of air quality, but no studies have yet tried to explore the variation of PM effect on human health before and after the Beijing 2008 Olympics in this city.

Studies show that ambient temperature and particles are associated with human health and that they may interact to cause adverse effect, however the modified effect by temperature and season on the association between PM and mortality still remains inconsistent. Samet et al. (1998) found little evidence that weather conditions modified the effect of pollution, while Ren and Tong (2006) observed that temperature significantly modified the health effects of PM in Brisbane, Australia. Moreover, the modifying effects which were found in many cities were inconsistent with each other on this issue. For example, some have found increased adverse effects on warm days (Ren and Tong, 2006; Yi et al., 2010; Li et al., 2011, 2012), and others found an increased effect in the winter season (Ko et al., 2007a,b; Bell et al., 2008; Kan et al., 2008; Qian et al., 2012; Qiu et al., 2012). To date, this issue has not been addressed from the epidemiological point of view in Beijing, China.

In the present study, we explored the health effects of PM_{10}, $PM_{2.5}$, and $PM_{2.5-10}$ on several mortality outcomes in Beijing, China, during the period 2005–2009. We developed 4 models for estimating the short-term effects between particles and health outcomes. This type of time-series design is a major approach to evaluate acute effects of air pollution in epidemiological studies for the last decade. This paper reported a comprehensive characterization of Beijing particle pollution on human health and provided the scientific background for the further control of air pollution at Beijing.

2. Material and methods

2.1. Data collection

We conducted the study in the urban area of Beijing, the capital of China. Beijing has experienced rapid industrialisation and urbanisation since the 1980s and has become one of the world's largest megacities. The study site is located at 39°58′N, 116°22′E, between the north 3rd Ring Road and the north 4th Ring Road in a high-density residential area of Beijing. The data set used in this study consists of daily data of air pollution, meteorological conditions, and death records from 1 Jan 2005 to 31 Dec 2009.

Daily air pollution data and meteorological data, including PM_{10}, $PM_{2.5}$, $PM_{2.5-10}$ (calculated as the difference between measured hourly concentrations of PM_{10} and $PM_{2.5}$), temperature (T), and relative humidity (RH), were obtained from the Beijing-Tianjin-Hebei Atmospheric Environment Monitoring Network, which was established by the Institute of Atmospheric Physics, Chinese Academy of Sciences (IAP, CAS) (Xin et al., 2010). The 24-h average concentrations of PM_{10}, $PM_{2.5}$, and $PM_{2.5-10}$, which were measured using the Tapered Element Oscillating Microbalance (Franklin, MA, USA) method, were defined as non-missing if at least 75% of the hourly values of each variable were available covering the study period. Sampling methods and instrument protocols, as well as quality assurance/quality control (QA/QC) procedures for air quality monitoring, were be executed based on the Chinese National Environmental Protection Standard, Automated Methods for Ambient Air Quality Monitoring. The real-time data was collected and transferred via the internet.

The Air pollution Index (API), which is administered by the Beijing Environmental Protection Agency, is a reflection and evaluation method of air quality in China. Because of its simple and visualized features, the results are suitable for expressing the city's short-term air quality and its changing tendency.

The health outcome data in this study, comprising respiratory mortality (RM), circulatory mortality (CM), and non-accidental total mortality (NAM) (including RM and CM), were provided by the China Centers for Disease Control and Prevention and the Third Hospital of Peking University. The causes of death were coded according to the International Classification of Diseases, 10 (ICD-10).

2.2. Data analysis

Most epidemiological studies of air pollution effects use a time series design, in which day-to-day changes in mortality (usually in a single city) are related to day-to-day changes in air pollution exposure (Samet et al., 1997; Ren and Tong, 2006). It is assumed that the daily mortality counts typically follow an overdispersed Poisson distribution (Hastie and Tibshirani, 1990; Samet et al., 1997). In this study, Poisson generalized additive models (GAMs) were employed to explore the associations of PM_{10}, $PM_{2.5}$, $PM_{2.5-10}$, API, and temperature with health outcomes. We used cubic smoothing functions to control the confounding effects of secular trend, seasonality, day-of-the-week effect, public holidays, temperature and relative humidity.

Particulate pollution is reported to be the major air pollutant in Beijing for about 85% of days in the last 5 years (Beijing Municipal Environmental Protection Bureau 2010: http://www.bjepb.gov.cn/portal0/tab181/), and airborne particles and the main gaseous pollutants (i.e. NO_2, SO_2, O_3, and CO) are correlated and may interact with each other (Wong et al., 1999; Hong et al., 2002). So the single pollutant models are used in this study to estimate particles health effect. We first build basic models for various mortality outcomes without considering whether a modifying effect exists between temperature and PM or API. The independent models are described as follows,

$$\begin{aligned}\mathrm{Log}[\mathrm{E}(Yi)] &= \beta X_i + s(\mathrm{time}, b_s = "cr", df) + s(\mathrm{RH}_i, b_s = "cr", df) \\ &+ s(T_i, b_s = "cr", df) + \mathrm{as.factor}(\mathrm{DOW}) + \mathrm{Season} + \mathrm{Holiday} \\ &+ \alpha = \beta X_i + s(\mathrm{T}_i, b_s = "cr", df) + \mathrm{COVs} + \alpha \end{aligned} \tag{1}$$

where E (Yi) refers to the expected count at day i; X_i refers to air pollution variables such as PM_{10}, $PM_{2.5}$, $PM_{2.5-10}$, and API at day i; T_i refers to daily average temperature at day i; RH refers to daily average relative humidity at day i; s ( ) is the cubic smoothing spline; DOW denotes days of week on day i; α refers to the intercept; β is the coefficient; df is the degree of freedom of the smooth function; and COVs represents all other covariates in the model. Our study used the degrees provided by smoothness estimated automatically to reduce the human error in this study. Second, we used the nonparametric bivariate response model to identify the modifying effects of temperature and PM on health outcomes. The pollution effects across temperature level are estimated as done in some previous studies (Lipsett et al., 1997; Ren et al., 2006).

Table 1
Summary statistics of air pollution, meteorological conditions, and death outcomes in Beijing, China, 2005–2009.

Variable		Mean ± SD	Minimum	P_{25}	Median	P_{75}	Maximum
Air pollution	PM_{10}/μg m^{-3}	126 ± 87	7	66	107	164	900
	$PM_{2.5}$/μg m^{-3}	75 ± 54	2	36	64	105	435
	$PM_{2.5-10}$/μg m^{-3}	50 ± 59	1	20	41	72	827
	API	96 ± 55	12	64	87	111	500
Meteorological	T/°C	15 ± 11	−10.1	4.2	16.0	24.4	33.7
Condition	RH/%	44 ± 19	8	27	43	59	91
Death outcome	NAM	146 ± 49	39	127	153	178	260
	RM	70 ± 29	8	56	74	89	154
	CM	16 ± 7	3	11	15	20	51

Calculations were based on daily values using hourly observations. SD means standard deviation. P_{25} and P_{75} mean the 25th and 75th percentile. Abbreviations: $PM_{2.5}$, particles with an aerodynamic diameter less than 2.5 μm; PM_{10}, particles with an aerodynamic diameter less than 10 μm; $PM_{2.5-10}$, particles with an aerodynamic diameter between 2.5 and 10 μm; API, Air pollution Index; T, temperature; RH, relative humidity; NAM, non-accidental mortality; RM, respiratory mortality; CM, circulatory mortality.

$$\mathrm{Log}[\mathrm{E}(Yi)] = s(X_i, T_i) + s(X_i, b_s = \text{"}cr\text{"}, df) + s(T_i, b_s = \text{"}cr\text{"}, df) + \mathrm{COVs} + \alpha \tag{2}$$

Then, we replace the pollution term in Equation (1) as a seasonal interaction term. The third model which reflect the obvious changes throughout the year can be defined as,

$$\mathrm{Log}[\mathrm{E}(Y_i)] = b_s \sin(2\,pi\,\mathrm{time}/365)X_{i-l} + b_c \cos(2\,pi\,\mathrm{time}/365)X_{i-l} + b_o X_{i-l} + s(\mathrm{time}, bs = \text{"}cr\text{"}, df) + s(\mathrm{RH}_i, bs = \text{"}cr\text{"}, df) + s(\mathrm{T}_i, bs = \text{"}cr\text{"}, df) + \mathrm{COVs} + \alpha \tag{3}$$

where b_s, b_c, and b_0 are estimated coefficients.

Similarly, a categorical variable with five categories (5 years) is created to investigate the changes of PM effect over the recent 5 years. The whole data are classed into five periods by different years. We then add a product term of the pollutant concentrations and dummy variables into the core model to test the possible variations from year-to-year changes. The equation is as follows:

$$\mathrm{Log}[\mathrm{E}(Yi)] = \beta_{2005} I_{2005} X_i + \beta_{2006} I_{2006} X_i + \beta_{2007} I_{2007} X_i + \beta_{2008} I_{2008} X_i + \beta_{2009} I_{2009} X_i + s(\mathrm{time}, bs = \text{"}cr\text{"}, df) + s(\mathrm{RH}_i, bs = \text{"}cr\text{"}, df) + s(T_i, bs = \text{"}cr\text{"}, df) + \mathrm{COVs} + \alpha \tag{4}$$

where I_{2005}, I_{2006}, I_{2007}, I_{2008}, I_{2009} = 0/1 indicator variables representing the year of 2005, 2006, 2007, 2008, and 2009, respectively. β_{2005}, β_{2006}, β_{2007}, β_{2008}, β_{2009} = regression coefficients regarding the relation between $PM_{2.5}$ and death counts for a given year. We adjust for the same covariates (COVs) as in Model (1), (2), (3), and (4). To explore the acute effect and modifying effect on mortality in Beijing, we estimate the relative risk (RR) to denote the combinations of air pollutant and human health across different temperature and season levels. To capture the cumulative and delayed effects of PM and API on mortality risk, different lag structures for each particulate size fraction are examined: (1) single-day lags up to day 10, and (2) moving averages up to 10 days. The results is expressed in terms of the percentage increase in daily non-accidental mortality, respiratory mortality, and circulatory mortality for a 10 μg m^{-3} and 10 point increment of pollutant concentrations, and respective 95% confidence interval (95% CI). All model analyses are conducted in the statistical environment R, version 2.11.1, using the mgcv package, 1.6-–2 (R Development Core Term 2011: http://www.r-project.org).

3. Results

3.1. Data description

We examine the distribution of each variable by time during the study periods. Table 1 provides distributional characteristics of air pollution ($PM_{2.5}$, PM_{10}, and $PM_{2.5-10}$), weather (*T* and RH), and health outcome variables (NAM, RM, and CM). The results show considerable variation in each of variables.

The annual mean concentrations of $PM_{2.5}$ and PM_{10} are 75 μg m^{-3} and 126 μg m^{-3}, and are much higher than the National Grade II standard level for ambient air quality which will be implemented in China by 2016, with the annual concentration of 35 μg m^{-3} and 70 μg m^{-3} for $PM_{2.5}$ and PM_{10}, respectively. The exceeding days and exceeding rates of the 24-h average are 508, 28.4%, and 720, 40.3%, respectively. That means PM pollution (especially fine particles) is still a severe environmental problem in Beijing. $PM_{2.5}$ accounts for a substantial part of the PM_{10} in Beijing, and the ratio of $PM_{2.5}$ to PM_{10} ranges from 30% to 89%, with an average of 59%. PM_{10} is significantly positively correlated with $PM_{2.5-10}$ ($r = 0.80$) and $PM_{2.5}$ ($r = 0.76$), but $PM_{2.5}$ and $PM_{2.5-10}$ are weakly correlated ($r = 0.23$). Fig. 1 shows the distribution of PM concentrations and API points across four seasons. It is obvious that PM_{10} and $PM_{2.5-10}$ are higher on cool days (i.e. spring and winter), with mean concentrations of 140 μg m^{-3} and 70 μg m^{-3}, respectively. In addition to slightly higher in summer (79 μg m^{-3}), $PM_{2.5}$ shows no significant seasonal variations. API is the lowest in summer (83), with an annual average point of 96 for the full year.

3.2. Regression results

Fig. 2 shows the relative risk (RR) (95% CI) of daily mortality per 10 μg m^{-3} increase in $PM_{2.5}$, PM_{10}, $PM_{2.5-10}$, and API on different

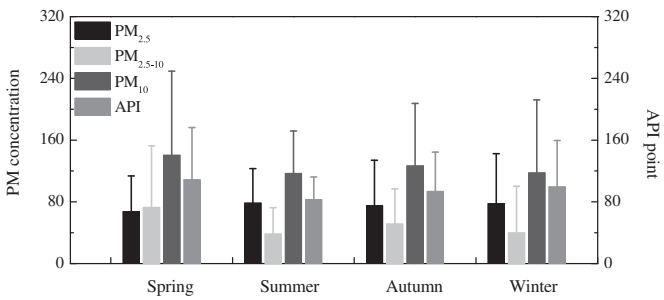

Fig. 1. Distribution [mean (SD)] of PM pollution concentrations (μg m^{-3}) and API point across different seasons in Beijing, 2005–2009. Calculations were based on daily values using hourly observations. Abbreviations: $PM_{2.5}$, particles with an aerodynamic diameter less than 2.5 μm; PM_{10}, particles with an aerodynamic diameter less than 10 μm; $PM_{2.5-10}$, particles with an aerodynamic diameter between 2.5 and 10 μm; API, Air pollution Index. Error bars represent standard deviation.

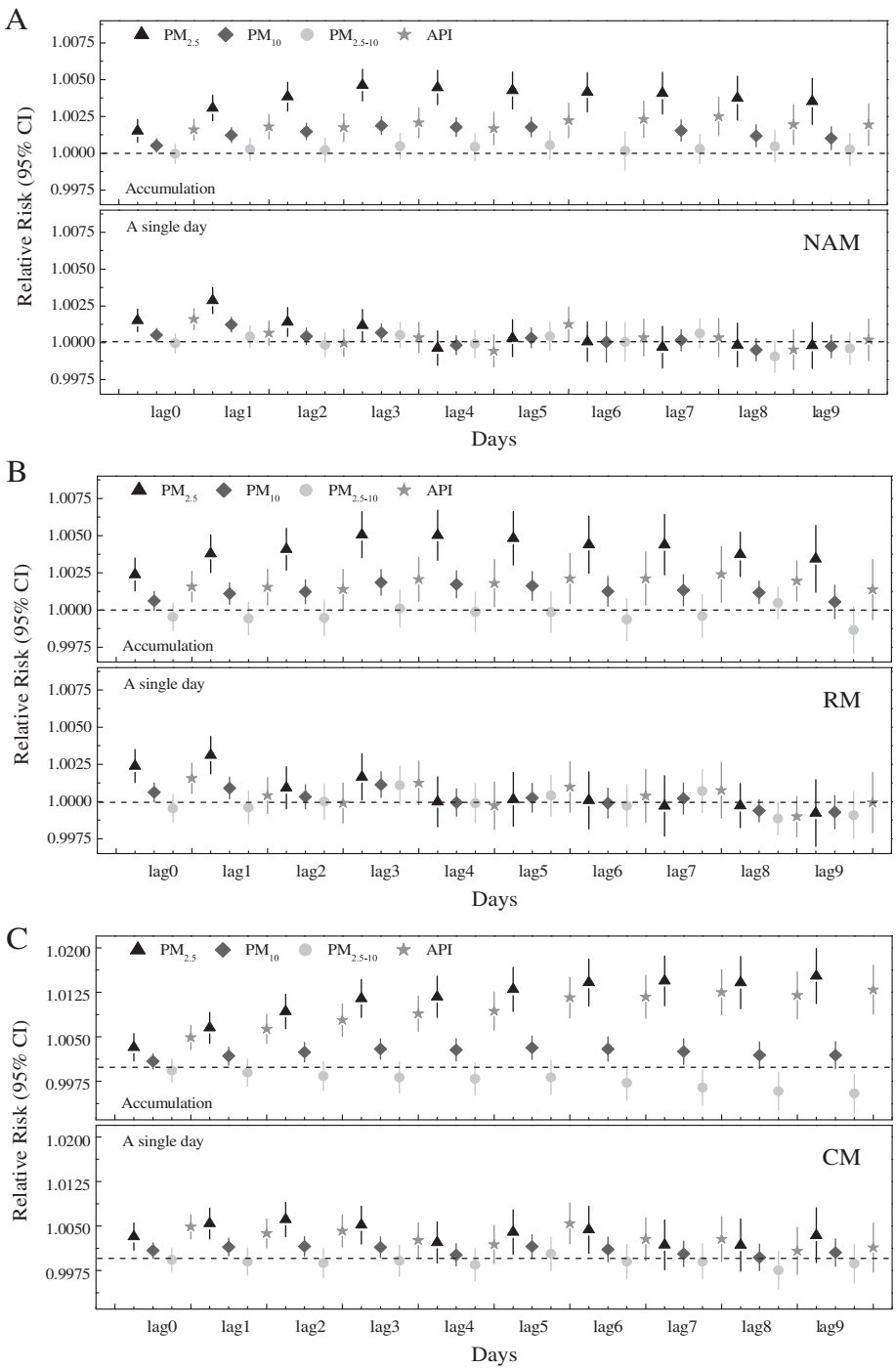

Fig. 2. Distribution of relative risk across lags of different independent variables for non-accidental mortality (A); respiratory mortality (B); circulatory mortality (C). Points represent central estimates, error bars represent 95% confidence intervals (95% CI), and dotted lines indicate RR = 1.00.

lags. On the whole, the effects of $PM_{2.5}$ are much higher than PM_{10} and API, whereas the estimated values of $PM_{2.5–10}$ which are the lowest almost remain to be 1.0. For NAM, RM, and CM, we find significant effects of both $PM_{2.5}$ and PM_{10}, however, there are no significant associations found between $PM_{2.5–10}$ and these outcomes in all lags we examined. The effect estimates of the exposure over the precious several days are larger than those considering only a single day's exposure. It can be observed that the effects of $PM_{2.5}$ and PM_{10} on NAM and RM are the most significant in single-day lag1 and multi-day lag3 (i.e. 4-day moving average). But for CM, the effect of $PM_{2.5}$ and PM_{10} increases observably as its duration increases, until they remain stable in certain values which occurs around 1.01.

Fig. 3 graphically presents the exposure–response relationships between PM and three mortality outcomes. We can conclude that the exposure–response curves associated with $PM_{2.5}$ and PM_{10} exposure present positive nonlinear relationships, although the risks did not increase monotonically. The curves which manifest an obvious J-shaped pattern tend to become nonlinear and flat at higher PM concentrations. It is becoming more clear that estimated

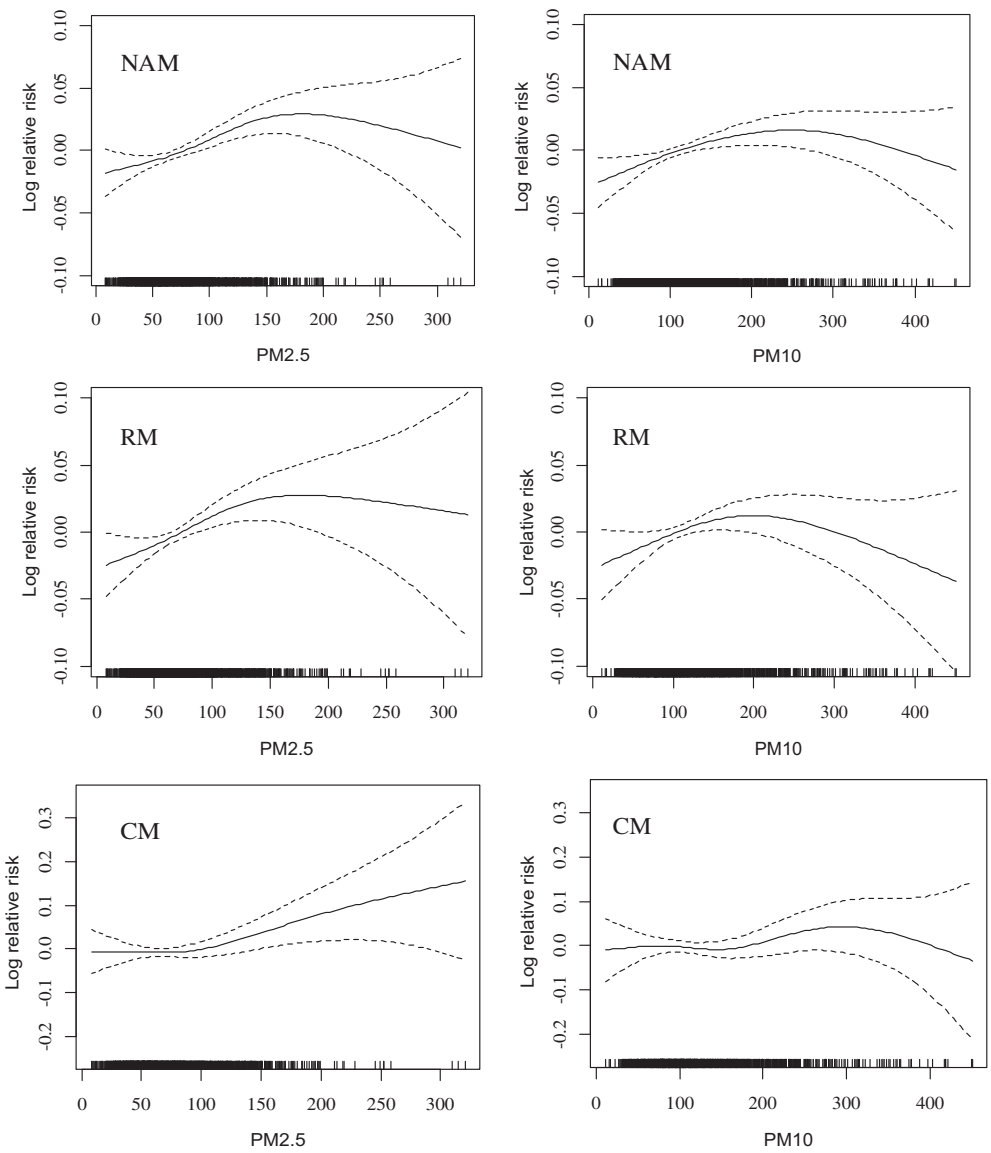

Fig. 3. Dose-response association of mean $PM_{2.5}$ and PM_{10} with different health outcomes in Beijing, China, 2005–2009. Dotted lines indicate 95% confidence interval. Abbreviations: NAM, non-accidental mortality; RM, respiratory mortality; CM, circulatory mortality.

log-RR remain to be stable values while PM_{10} are greater than 230 μg m^{-3} or PM_{25} are greater than 150 μg m^{-3}, even on a downward trend. That means the deaths caused by PM are increasing relaxed under these stages.

Fig. 4 depicts the modifying effect of $PM_{2.5}$, PM_{10} by temperature on each health outcome (NAM, RM, and CM) for the previous 3-day moving average. The estimated death numbers are higher at extreme high or low temperature levels, especially at low levels. Obviously, the modifying effects exist across different temperature levels in the study area, with the lightest adverse effect appeared approximately at 24 °C of where there is a trough for the three health outcomes. It can also be obtained that the effects of particles on human health are smaller than the one caused by temperature, especially during extreme conditions.

Fig. 5 shows the seasonal variability of health effect for $PM_{2.5}$ on daily mortality in recent years. Table 2 shows the effect of PM and API on daily mortality across different seasons. A clear seasonal pattern of $PM_{2.5}$ acute effects exist in Beijing, with a high of about 0.92% (at the $P < 0.01$ level) in winter and a low of 0.05% (at the $P < 0.1$ level) in summer on total mortality, respectively. As expected, the effect is greater in the cool season (i.e. spring and winter) than in the warm season (i.e. summer and autumn) for $PM_{2.5}$, PM_{10}, and API, despite the maximum concentration of $PM_{2.5}$ is observed in warm season in the study periods. In general, most of seasonal estimates have passed the statistically significance (at the $P < 0.05$ level) for $PM_{2.5}$ and API, but for PM_{10}, half of the estimates did not show significantly associations in the analyses. On cool days, which is 2–13 times higher than the one on warm days, a 10 μg m^{-3} increase is associated with a maximum increase in NAM, RM, and CM of 0.92% (95% CI: 0.45–1.24%), 0.89% (95% CI: 0.53–1.13%), and 1.93% (95% CI: 1.72–2.10%) for $PM_{2.5}$, respectively. The corresponding estimates in NAM, RM, and CM for API are 0.64% (95% CI: 0.31–0.95%), 0.59% (95% CI: 0.35–0.89%), and 1.59% (95% CI: 0.98–1.93%), respectively.

Fig. 6 depicts the interannual variability of the mass concentration of $PM_{2.5}$ and the estimated percent increase for $PM_{2.5}$ in daily mortality from 2005 to 2009. It can be seen that the $PM_{2.5}$ levels generally declines in recent years in Beijing, whereas the

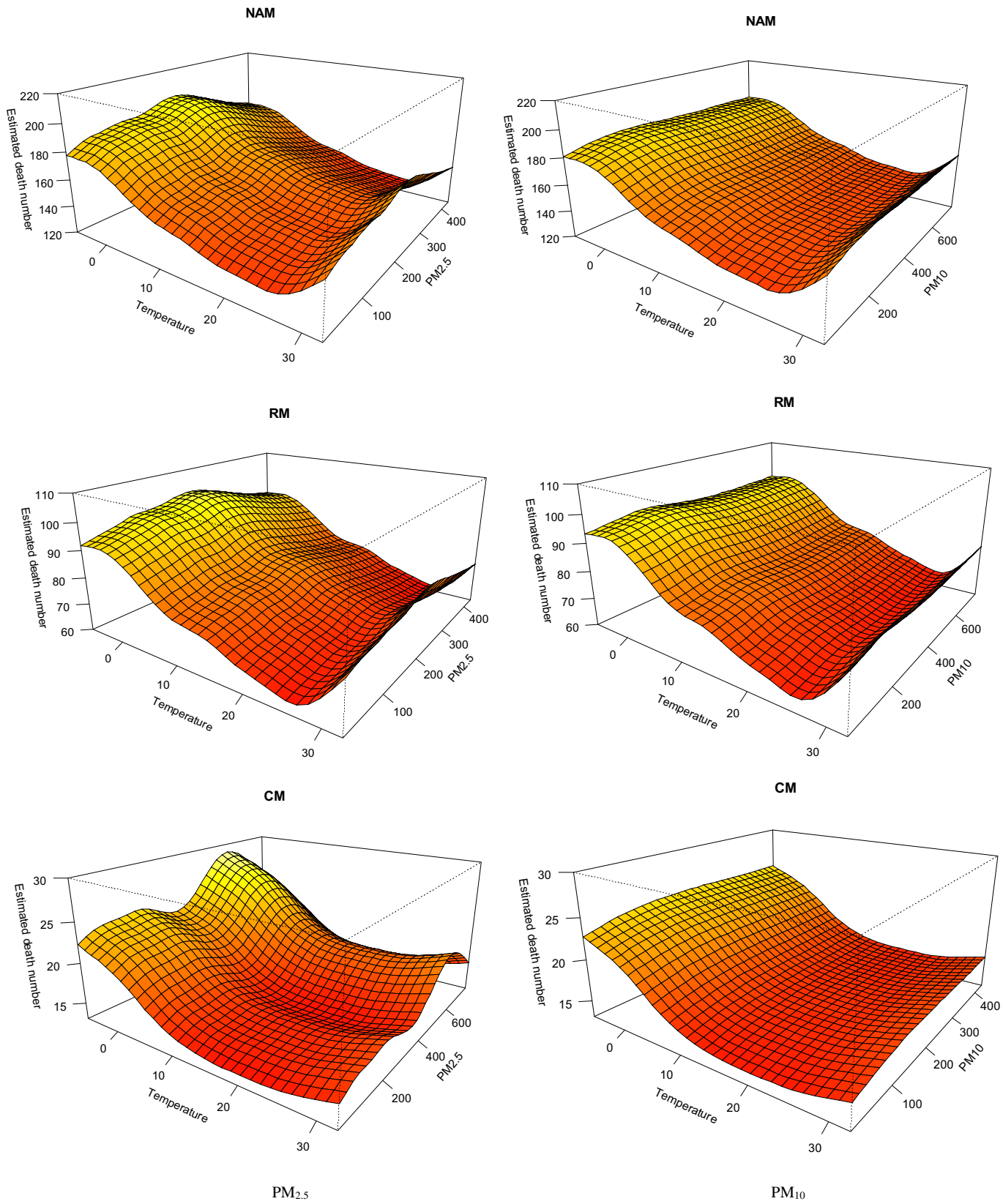

Fig. 4. Bivariate response surfaces of PM and temperature for the three health outcomes in Beijing, 2005–2009. Abbreviations: NAM, non-accidental mortality; RM, respiratory mortality; CM, circulatory mortality.

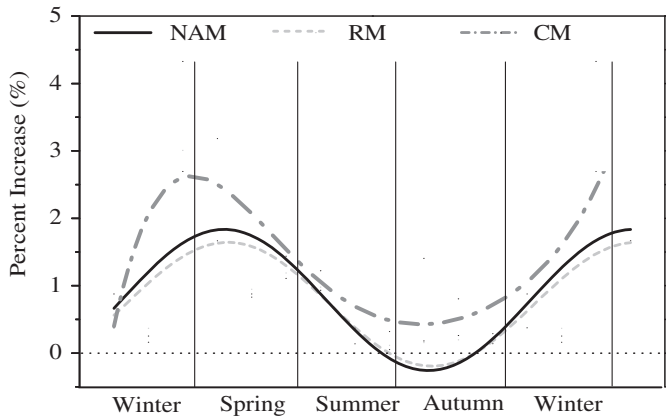

Fig. 5. The seasonal variability of the estimated percent increases in daily mortality of $PM_{2.5}$ in recent years. These curves resulted from seasonal interaction model (Equation (3)). Abbreviations: NAM, non-accidental mortality; RM, respiratory mortality; CM, circulatory mortality; spring, March through May; summer, June through August; autumn, September through November; winter, December through February.

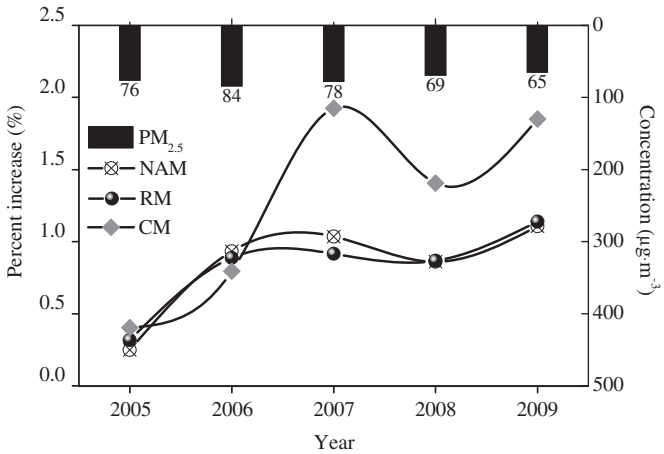

Fig. 6. The inter-annual variability of the estimated percent increases in daily mortality of $PM_{2.5}$ in recent years. These curves were fitted by statistical regression equations. Column means the annual average concentration of $PM_{2.5}$ in Beijing. Abbreviations: NAM, non-accidental mortality; RM, respiratory mortality; CM, circulatory mortality.

relative risk shows different change trends. Overall, the level of the estimated percentage increase assumes an escalating tendency during the study period, in addition to having a low value in 2008 because strict atmospheric pollution control measures were implemented in Beijing–Tianjin–Hebei region before the Olympics games (Xin et al., 2010, 2012). But it bounced off the bottom and started a new uptrend after 2008. The values which show the largest effect in 2009, rose sharply again after the Beijing Olympics. Under this serious current situation, a 10 μg m^{-3} increase of $PM_{2.5}$ corresponded to more than 1.00% increase of non-accidental mortality, respiratory mortality, and circulatory mortality.

4. Discussion

The present study suggests the existence of a short-term association spreading over several succeeding days between $PM_{2.5}$, PM_{10} or $PM_{2.5-10}$ and the risk of mortality for non-accidental, respiratory and circulatory mortality during 2005–2009. In general, the health effect of $PM_{2.5}$ remains strongly significant, whereas a null association is found between $PM_{2.5-10}$ and daily mortality. By using product models we also find seasonal patterns and interannual patterns of PM health effect, which shows that low temperatures could enhance the $PM_{2.5}$ effect on cause-specific population mortality and the level of adverse effect assume an escalating tendency from 2005 to 2009 in Beijing.

As it is shown in Table 1 and Fig. 2, in the current analysis, a 10 μg m^{-3} increase in the 4-day moving average concentrations of $PM_{2.5}$ and PM_{10} corresponded to 0.65% (95% CI: 0.29–0.80%), 0.63% (95% CI: 0.25–0.83%), and 1.38% (95% CI : 0.51–1.71%); 0.15% (95% CI: 0.04–0.22%), 0.08% (95% CI: 0.01–0.18%), and 0.44% (95% CI: 0.12–0.63%) increase of non-accidental mortality, respiratory mortality, and circulatory mortality, respectively. A 10 point increase of API corresponded to 0.50% (95% CI: 0.19–0.61%), 0.43% (95% CI: 0.14–0.59%), and 1.15% (95% CI: 0.41–1.47%) increases, respectively. Our results are consistent with the only other study for

Table 2
The estimated mean percentage of change in daily mortality associated with a 10 μg m^{-3} increase of PM across different seasons, 2005–2009.

	NAM			RM			CM		
	Central effect	95% CI	*P*-value	Central effect	95% CI	*P*-value	Central effect	95% CI	*P*-value
$PM_{2.5}$									
Spring	**0.76**	(0.21, 0.95)	0.00627	**0.74**	(0.48, 1.02)	0.01791	**0.84**	(0.58, 1.10)	0.01065
Summer	0.05	(−0.12, 1.21)	0.06607	−0.09	(−0.23, 0.56)	0.47926	**0.33**	(0.09, 0.68)	0.03382
Autumn	**0.43**	(0.07, 0.68)	0.00928	0.45	(0.13, 0.62)	0.08024	**0.84**	(0.59, 1.07)	0.00061
Winter	**0.92**	(0.45, 1.24)	0.00463	**0.89**	(0.53, 1.13)	0.00031	**1.93**	(1.72, 2.10)	0.00809
Total	**0.65**	(0.29, 0.80)	0.00497	**0.63**	(0.28, 0.83)	0.00085	**1.38**	(0.81, 1.71)	0.03065
PM_{10}									
Spring	**0.22**	(0.16, 0.49)	0.00001	**0.16**	(0.11, 0.48)	0.01193	0.40	(0.11, 0.67)	0.06094
Summer	**−0.26**	(−0.31, 0.09)	0.01099	0.07	(−0.15, 0.23)	0.17467	−0.16	(−0.23, 0.63)	0.34012
Autumn	0.04	(−0.16, 0.26)	0.18464	−0.40	(−0.48, 0.05)	0.18528	0.15	(0.02, 0.29)	0.96358
Winter	0.13	(0.05, 0.49)	0.23512	0.07	(−0.15, 0.16)	0.42991	**0.57**	(0.25, 0.84)	0.00024
Total	0.15	(0.04, 0.32)	0.12047	0.08	(0.01, 0.18)	0.32191	0.44	(0.12, 0.63)	0.46226
API									
Spring	**0.41**	(0.12, 0.63)	0.00262	**0.32**	(0.17, 0.61)	0.01590	**0.70**	(0.14, 1.52)	0.00031
Summer	−0.12	(−0.34, 0.16)	0.63470	−1.25	(−2.20, −0.16)	0.91960	**0.40**	(0.28, 0.66)	0.00109
Autumn	**0.21**	(0.13, 0.43)	0.01250	**0.14**	(0.05, 0.59)	0.01790	**0.74**	(0.39, 0.92)	0.01530
Winter	**0.64**	(0.31, 0.95)	0.00293	**0.59**	(0.35, 0.89)	0.00015	**1.59**	(0.98, 1.93)	0.00000
Total	**0.26**	(0.08, 0.44)	0.03221	**0.23**	(0.11, 0.63)	0.01349	**0.82**	(0.38, 1.24))	0.00076

Statistically significant results ($P < 0.05$) were given in bold letters. Abbreviations: NAM, non-accidental mortality; RM, respiratory mortality; CM, circulatory mortality; $PM_{2.5}$, particles with an aerodynamic diameter less than 2.5 μm; PM_{10}, particles with an aerodynamic diameter less than 10 μm; API, Air pollution Index; 95% CI, 95% confidence interval.

Beijing which found a 10 μg m^{-3} increase of $PM_{2.5}$ was associated with a 0.66% increase in respiratory mortality from 2007 to 2008 (Chen et al., 2011), and higher than other cities in China, e.g. 0.36% of $PM_{2.5}$ and total mortality in Shanghai (Kan et al., 2007), 0.61% of $PM_{2.5}$ and respiratory mortality in Shanghai (Chen et al., 2011), 0.29% of $PM_{2.5}$ and respiratory mortality in Shenyang (Chen et al., 2011), and 0.42% of $PM_{2.5}$ and total mortality in Pearl River Delta (Xie et al., 2011). But compared with prior estimates in developed countries, our values are lower in Beijing. For example, a meta-analysis study based on Canada cities showed 1.2% increase of $PM_{2.5}$ and total mortality (Burnett et al., 2000), a multi-city analysis in 112 U.S. cities showed 1.00% increase in total mortality (Zanobetti and Schwartz, 2009), and a piecewise linear analysis showed 7.4% increase in respiratory mortality in two capital cities of the Canary Islands (Lopez-Villarrubia et al., 2011). First, the estimates of relative risk demonstrate the average level of the adverse effects over the entire range of particle concentrations. In our previous study we found that the greatest damage to human respiratory health occurs mainly within the concentration range of 20–60 μg m^{-3}, and excessively high $PM_{2.5}$ concentrations could make the adverse effect stable (Li et al., 2013). At higher concentrations, the risk of death per unit increase of pollutant concentrations often tended to be reduced, and the exposure-response curve of air pollution tends to become flat. Second, this difference on $PM_{2.5}$ health effect may be affected by the characteristics of particles' levels, population sensitivity to PM, age distribution, and chemical composition and toxicity (Zhou et al., 2011), which depends a lot on the level of industrialisation and urbanisation.

Although our estimates in Beijing are currently lower compared with prior studies in overseas developed countries and regions, the values are obviously on the rise. The level of the estimated percentage increase assumes an escalating tendency during the study period in addition to having a low value in 2008. The prevention and control measures for atmospheric pollution that were implemented jointly by the Beijing, Tianjin, and Hebei municipal governments made distinct successful contributions to the reduction of air pollution during this time. It was reported that the levels of $PM_{2.5}$ decrease of 48% compared to the prophase mean concentration before the Olympic Games, and the contribution of emission control measures contributed ≥62% of the decrease of $PM_{2.5}$ (Xin et al., 2012). As the temporary atmospheric pollution control measures were terminated mostly after the Olympic Games, the values of estimated percentages increased significantly even though the level of $PM_{2.5}$ has decreased. That means the government would make less toxicity of PM pollution only by more rigorous reduction measures in the Beijing–Tianjin–Hebei region. Unless strict measures are implemented now, the serious air pollution disaster such as the London smog of 1952 or the smog event in masi river of Belgium will unfortunately occur in Beijing.

We found no significant associations found between $PM_{2.5-10}$ and these outcomes in all lags we examined. The lack of a significant effect of $PM_{2.5-10}$ on daily mortality is consistent with most prior studies (Schwartz et al., 1996; Klemm and Mason, 2000; Anderson et al., 2001; Villeneuve et al., 2003; Kan et al., 2007), but in contrast to several others (Burnett et al., 2000; Cifuentes et al., 2000; Mar et al., 2000). Such observed differences may have been due to differences in regional coarse fraction composition (Mar et al., 2000). Ambient particles are chemically nonspecific and consist of various components and compounds (e.g. trace elements, elemental carbon, organic carbon, nitrate, sulphate, arsenic, ammonia, calcium, sodium, magnesium, potassium, and etc.), and the toxicity of each of these chemical components and their mixtures may vary (Zhou et al., 2011; Liu et al., 2013). $PM_{2.5-10}$ is typically mechanically generated by crushing or grinding, and is often dominated by resuspended dusts and crustal material from paved or unpaved roads or from construction, farming, and mining activities (Kan et al., 2007). Some nature-generated particles which predominately comprise coarse particles in Beijing (Sun et al., 2004; Hu et al., 2011; Liu et al., 2013), such as resuspended dusts, crustal material from paved or unpaved roads or from farming, are less toxic to human health (Longueville et al., 2013). But fine particles are typically generated from the combustion of coal, biomass, motor vehicle, waste and oil (Song et al., 2002; Tie et al., 2006; Kan et al., 2007; Chen et al., 2011; Liu et al., 2013). The health effect is significantly modified by aluminium, bromine, chromium, arsenic, iron, potassium, silicon, sulphate, nickel, sodium, elemental carbon and zinc, which are high in locations with higher residual oil-burning and traffic-related pollution (Bell et al., 2009; Zanobetti and Schwartz, 2009; Zhou et al., 2011). This fact reflected different characteristics of source from airborne particulate matter size fractions in the study area. On the other hand, owing to low specific surface area, coarse particles could not absorb more toxic substances. Previous study also found that the in vivo pulmonary toxicity of urban particles varied with size, with the greatest toxicity from particles <1.7 μm and the lowest toxicity from those >3.5 μm (Costa and Dreher, 1997). Moreover, it is well known that airborne particles in order to have an effect on human health have to come into contact with cells and tissues of the human body, and the amount and site of deposition depends on the aerodynamic and thermodynamic properties of the particle inhaled, particularly on their size and shape (Zereini and Wiseman, 2010). Coarse particles cannot reach the periphery of the lung where they might not settle at the surface of the small bronchi, the respiratory bronchiole and the alveoli (Heyder et al., 1986). Besides, small particles have been associated with plasma viscosity, sequestration of red cells in the circulation, and indicators of cardiac autonomic dysfunction including increased heart rate, decreased heart rate variability, and increased cardiac arrhythmias (Peters et al., 1997; Dockery, 2001).

In many locations, patterns of air pollution are driven by weather. The effects of particles on human health are smaller than the one caused by temperature, especially during extreme conditions. Previous study also found that marked changes in ambient temperature could cause physiological stress and alter a person's physiological response to air pollutants, perhaps making them more susceptible to the adverse effects of $PM_{2.5}$ (Stephen et al., 2012). Another major finding of this study is that the modifying effect of temperature and season on the association between PM and mortality is significant, which is consistent with previous studies conducted in Hong Kong (Wong et al., 1999, 2001; Hong et al., 2002, 2002; Ko et al., 2007a,b). However some studies using several parallel models have shown that high temperature could enhance the PM_{10} effect on daily mortality (Katsouyanni et al., 1993, 2001; Li et al., 2011; Ren and Tong, 2006; Qian et al., 2008). These studies found that temperature modified the association between PM_{10} and mortality, but the results were sensitive to the number of degrees of freedom (Roberts, 2004; Li et al., 2013). Our study use the degrees provided by smoothness estimated automatically to reduce the human error. On the other hand, examining the modifying effect of $PM_{2.5}$ instead of PM_{10} will also reduce some errors. In all other cases, the weather conditions in Beijing differ from other countries. Beijing has extremely hot summers, and most people spend more time indoors with air conditioning, decreasing the opportunities for human exposure outdoors. Therefore, the modifying effect of temperature or season and particles on mortality may be different in areas with different weather patterns, latitudes, air pollution levels and prevalence of air-conditioning systems. Thus, further research is needed to examine the interactive effects between air pollutants and temperature or season on mortality, especially in different spatial settings.

5. Conclusions

In summary, we find significant associations of daily mortality with $PM_{2.5}$ and PM_{10}, but not with $PM_{2.5-10}$, in Beijing. Compared with other developed countries and regions, the level of adverse effects between $PM_{2.5}$ and mortality in Beijing is lower, but the growth trend of relative risk will become increasingly more obvious in the future. The effects of particles on human health are smaller than the one caused by temperature, especially during extreme conditions. We also find it is important to avoid exposing outdoor particularly in cool seasons in Beijing. Moreover, controlling anthropogenic sources of emissions is crucial to avoid the risks associated with exposure to the different particle fractions.

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