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Short-term effects of PM₁, PM_{2.5}, and PM_{2.5} constituents on myocardial infarction mortality in qingdao, China: A time-stratified case-crossover analysis

Xiaoyun Ma^{a,1}, Haiping Duan^{b,1}, Hua Zhang^{c,1}, Xue Liu^{a,1}, Xiaohui Sun^c, Jing Wei^d, Min Zhao^e, Bo Xi^{a,*}

^a Department of Epidemiology, School of Public Health, Qilu Hospital, Cheeloo College of Medicine, Shandong University, Jinan, Shandong, China ^b Qingdao Municipal Center for Disease Control and Prevention, Qingdao, Shandong, China

^c Institute of Chronic Non-communicable Disease, Qingdao Municipal Center for Disease Control and Prevention, Qingdao, Shandong, China

^d Department of Atmospheric and Oceanic Science, Earth System Science Interdisciplinary Center, University of Maryland, College Park, MD, USA

^e Department of Nutrition and Food Hygiene, School of Public Health, Cheeloo College of Medicine, Shandong University, Jinan, Shandong, China

HIGHLIGHTS

• The effects of PM1 on MI mortality may be slightly higher than that of PM2.5.

• Sulfate, nitrate, ammonium, and BC exposures may raise the risks of MI mortality.

• Significant effects of PM and constituent exposures were observed in aging subgroup.

• Clean heating policies should cover the scope of PM2.5 constituents in Qingdao.

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ABSTRACT

Myocardial infarction (MI) is a major global contributor to disability and death. Few studies have investigated the impacts of short-term exposure to particulate matter with an aerodynamic diameter \leq 1 μ m (PM₁) and \leq 2.5 um (PM_{2.5}) constituents on MI mortality. We aimed to estimate the risks of MI mortality related to short-term exposures to PM1, PM2.5, and PM2.5 constituents. Daily MI deaths, daily concentrations of PM1, PM2.5, PM2.5 chemical compositions, and meteorological data in Qingdao during 2015-2019 were collected. We used a timestratified case-crossover design fitted with a conditional quasi-Poisson regression incorporated with distributed lag model to quantify the associations between PM1, PM2.5, and PM2.5 chemical compositions and MI mortality. Subgroup analyses were further implemented based on sex, age, and season. There were 36,235 MI deaths in Qingdao during 2015-2019. The results of single-day lagged effects showed that PM1 (lag0 and lag1 days), PM2.5 (lag0 and lag1 days), sulfate (lag0 and lag1 days), nitrate (lag0 day), ammonium (lag0 and lag1 days), and black carbon (BC, lag0 and lag1 days) were positively related to an elevated risk of MI death. The cumulative effects of per interquartile range (IQR) rise in PM1, PM2.5, sulfate, nitrate, ammonium, and BC concentrations lasting for three days (lag02 day) were related to excess risks of 2.5% (95% confidence interval [CI]: 0.2%, 4.9%), 2.0% (0.1%, 3.9%), 2.2% (0.3%, 4.1%), 2.0% (0.1%, 3.9%), 2.0% (0.1%, 4.0%), and 2.1% (0.2%, 3.9%) for MI mortality, respectively. Significant effects of PM1, PM2.5, sulfate, and BC exposures on excess risks of MI mortality were observed in aging subpopulations or during the cold season. Short-term exposures to PM1, PM2.5, sulfate, nitrate, ammonium, and BC were linked to elevated risks of MI mortality. These results strengthen the case for implementing policies to reduce air pollution.

* Corresponding author.

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E-mail address: xibo2010@sdu.edu.cn (B. Xi).

¹ Xiaoyun Ma, Haiping Duan, Hua Zhang, and Xue Liu contributed equally to this study.

1. Introduction

Myocardial infarction (MI), known as a typical clinical manifestation of cardiovascular disease (CVD), has risen to the top of the list of global causes of disability and mortality (Dai et al., 2014; McAloon et al., 2016; Murry et al., 2006). Above all, within seven days of a positive test for Corona Virus Disease 2019 (COVID-19), there is an almost double increased risk of MI mortality in Scotland (Ho et al., 2021a). Currently, heart transplantation is the only viable long-term treatment for MI, but it is limited by rare organ donors and surgical hazards (Alonaizan and Carr, 2022; Beliën et al., 2022). Therefore, early prevention and avoidance of exposure to risk factors (such as abnormal lipids, abdominal obesity, overnutrition, present smoking, particulate matter [PM], etc.) are particularly important (Anand et al., 2008; Mustafić et al., 2012).

Globally, PM poses an enormous health threat and causes significant ambient pollution. In China, ambient PM pollution was reported to be one of the top four risk factors contributing to death in 2017 (Zhou et al., 2019). Over the past couple of decades, PM, mainly PM with an aerodynamic diameter $\leq 10 \ \mu m$ (PM₁₀) and $\leq 2.5 \ \mu m$ (PM_{2.5}), has been disclosed in several studies that were associated with elevated risks of CVD events (Brook et al., 2010; Zhou et al., 2021). Numerous studies revealed the links between PM₁₀ and PM_{2.5} exposures and stroke (Huang et al., 2019; Qiu et al., 2020), acute coronary syndrome (Kuzma et al., 2021; Vaudrey et al., 2020), congestive heart failure (Dominici et al., 2006; Qiu et al., 2020), and MI (Chen et al., 2018; Liang et al., 2018; Milojevic et al., 2014). However, authoritative studies have pointed out that smaller particles may pose stronger detrimental consequences on the cardiovascular system, leading to serious thrombosis and MI in mice (Chen et al., 2015; Farina et al., 2013; Kwon et al., 2020; Yang et al., 2019). In the absence of monitoring data for PM with an aerodynamic diameter $\leq 1 \mu m$ (PM₁), there is little evidence linking PM₁ exposure and MI mortality, particularly in developing countries. Therefore, studies investigating the association of PM1 with MI mortality are desired to fill the research gap.

Recently, $PM_{2.5}$ constituents have attracted considerable interest in cause-specific mortality and morbidity in epidemiological studies (Cao et al., 2012; Wang et al., 2019; Wang and Lin, 2016). Some previous studies suggested that the specific constituents of $PM_{2.5}$ might have varied health impacts via the toxicity of harmful substances and particles (Liao et al., 2015). However, the majority of studies emphasized the outcomes of respiratory illness (Ho et al., 2021b; Qiao et al., 2014; Wu et al., 2021), with little evidence of the impacts of $PM_{2.5}$ chemical compositions and MI death may contribute to connecting the $PM_{2.5}$ chemical compositions or emission sources with the health impacts and developing more targeted air pollution intervention programs.

In this study, we aimed to quantify the relationships of short-term exposures to PM_1 , $PM_{2.5}$, and $PM_{2.5}$ chemical compositions with the risks of MI mortality using a time-stratified case-crossover analysis, and to further evaluate the differences in effect estimates of PM_1 , $PM_{2.5}$, and $PM_{2.5}$ chemical compositions on MI mortality between the sex and age subgroups and seasons in Qingdao from January 1, 2015 to December 31, 2019.

2. Methods

2.1. Study area

Qingdao is a coastal city in Shandong Province, covering 11,293 km². It is situated on the eastern boundary of East China, which is flanked by China's Yellow Sea and the Bohai Sea. A permanent population of over 9.49 million lived in Qingdao by the end of 2019. Therefore, there are not only residential populations but also Qingdao Port, oil refineries, and chemical industries, which create an extremely

complex mix of PM_{2.5} chemical compositions (Bie et al., 2021).

2.2. Data collection

From January 1, 2015 to December 31, 2019, information on MI death in Qingdao was obtained from the Qingdao resident death monitoring system. The medical certificate or diagnosis of death is completed by the doctors or health workers from the hospital or health care institution, and then it is submitted to the Center for Disease Control and Prevention (CDC) of the local county within a prescribed time. Each county CDC will check the mortality registry form, and then report it to Qingdao CDC within 7 days. In this study, MI mortality (coded as I21 and I22) was the outcome of interest, based on the International Classification of Diseases version 10 (ICD–10). Information on the date of death, sex, and age was collected from the subjects. This study obtained ethical approval from the Ethics Review Committee of Public Health, Shandong University (No. LL20211203).

Daily mean PM₁, sulfur dioxide (SO₂), and ozone (O₃) concentrations from January 1, 2015 to December 31, 2019 were originated from the ChinaHighAirPollutants (CHAP) database (available at https://weij ing-rs.github.io/product.html), which combines the ground-based measurements, satellite remote sensing products, atmospheric reanalysis, and model simulations using artificial intelligence by considering the spatiotemporal heterogeneity of air pollution (Wei et al., 2019, 2021, 2022a,b). Daily mean PM2.5 and PM2.5 chemical compositions (including sulfate, nitrate, ammonium, organic matter [OM], and black carbon [BC]) for the same period were derived from the Tracking Air Pollution in China (TAP, available at http://tapdata.org.cn/), which combines information from multiple data sources, including ground observations, satellite aerosol optical depth, operational chemical transport model simulations, and other ancillary data (Geng et al., 2021; Xiao et al., 2021). Previous studies have elaborated on the descriptions of CHAP and TAP databases and the accuracy of the data (Geng et al., 2017; Wang et al., 2021; Zheng et al., 2021). With a spatial resolution of 10 km, the CHAP and TAP provide high-quality grid-level datasets for PM1, PM2.5, SO2, O3, and PM2.5 constituents. Daily mean concentrations of air pollutants for Qingdao City were calculated by aggregating the values of the grid cells within its boundary.

Meanwhile, the hourly temperature and dewpoint temperature at $0.25^{\circ} \times 0.25^{\circ}$ (approximately 22.5 km \times 22.5 km) spatial resolution were collected from the ERA5-Land climate reanalysis product, which was supplied by the European Center for Medium-Range Weather Forecasts (Burkart et al., 2021; Urban et al., 2021). Then, the hourly relative humidity (*RH*) for each day was determined based on temperature and dewpoint temperature as follows (Alduchov and Eskridge, 1996):

$$RH = 100 \times \left[e^{(17.625 \times DT)/(243.04 + DT)} / e^{(17.625 \times T)/(243.04 + T)} \right]$$

where T and DT represent the temperature and dewpoint temperature, respectively. Finally, the daily mean T and RH were computed by averaging the hourly T and RH.

2.3. Statistical analysis

The traits of the MI deaths, air pollutant exposures, and meteorological factors during 2015–2019 were reported using descriptive statistics. The correlations among PM_1 , $PM_{2.5}$, and its chemical compositions were investigated by Spearman's rank correlation analyses.

A time-stratified case-crossover design was adopted in this study. The case periods for each death refer to the date of death. In addition, the same weekdays in the same month and year were used to match the control periods (Ma et al., 2011). We conducted conditional quasi-Poisson regression with distributed lag models to quantify the associations of short-term exposures to PM_{1} , $PM_{2.5}$, and $PM_{2.5}$ chemical

compositions with the risks of MI mortality. Cross-basis function was applied to fit the single-day lag (lag0–lag6) and cumulative lag (lag01–lag06) effects of air pollutants PM₁, PM_{2.5}, and PM_{2.5} constituents on MI mortality referring to previous studies (Chen et al., 2020c; Li et al., 2021; Yang et al., 2020b). The single-pollutant models were as follows (Armstrong et al., 2014):

$$log(E(Y_t)) = \alpha + cb(pollutants) + ns(T, df = 3) + ns(RH, df = 3)$$
$$+ factor(stratum) + factor(holiday)$$

where Y_t denotes the daily cases of MI mortality at daytime t; α is the intercept; cb(pollutants) represents the cross-basis function to fit the lagged effects of daily mean PM₁, PM_{2.5}, or one of the PM_{2.5} constituent concentrations in the single-pollutant model; natural cubic splines (*ns*) with degrees of freedom (*df*) of 3 were performed to control for daily mean *T* and *RH* based on previous literature (Qiao et al., 2014). The control periods known as "*stratum*" are used to account for the long-term trend, seasonal trend, and "weekday effects" (Zhang et al., 2019). We also adjusted the public holidays (*holiday*) in this model.

In addition, sensitivity analyses were carried out by 1) building twopollutant models to assess the robustness of the effects of PM_1 , $PM_{2.5}$, and $PM_{2.5}$ constituents on MI death when adding cross-basis functions for other air pollutants (SO₂ and O₃ for PM_1 and $PM_{2.5}$; SO₂, O₃, and $PM_{2.5}$ mass for $PM_{2.5}$ constituents); and 2) converting the *df* of *T* and *RH* from 3 to 4–6, respectively.

The subgroup analyses stratified by sex (male and female), age groups (<65 years and \geq 65 years), and season (warm season: April–September; cold season: October–March of next year) were conducted to identify potentially susceptible subpopulations. We included an interaction term in distributed lag models to test the interaction effects and statistical differences between effect estimates for sex, age, and season subgroups.

The excess risk was adopted to quantify the association, which referred to the percentage change ($PC\% = [e^{\beta}-1] \times 100\%$) and 95% confidence interval (CI) in the risks of MI death related to each interquartile range (IQR) rise in daily mean concentrations of PM₁, PM_{2.5}, and PM_{2.5} chemical compositions. A *P*-value of 0.05 was utilized to determine statistical significance for all two-tailed statistical tests. All analyses were conducted based on R version 4.0.3.

3. Results

Table 1 lists the characteristics of MI mortality in Qingdao. From 2015 to 2019, a total of 36,235 residents died from MI (19,463 males and 16,772 females). Additionally, deaths aged \geq 65 years accounted for 82.6%, and deaths occurring in the cold season accounted for 58.1%. On average, the daily mean [standard deviation (SD)] number of MI deaths was 19.8 (6.4), 10.7 (4.0), and 9.2 (3.8) for the overall population, men,

Table 1

Characteristics	of	myocardial	infarction	mortality	in	Qingdao,	China
(2015–2019).							

Group	Total		Daily	
	Count (n)	Percentage (%)	Mean	SD
All	36,235	100.0	19.8	6.4
Sex				
Male	19,463	53.7	10.7	4.0
Female	16,772	46.3	9.2	3.8
Age, years				
<65 years	6294	17.4	3.4	1.9
\geq 65 years	29,941	82.6	16.4	5.7
Season				
Warm season	15,172	41.9	16.6	4.6
Cold season	21,063	58.1	23.1	6.3

Abbreviations: SD, standard deviation; Warm season, April to September; Cold season, October to March of the next year.

and women, respectively. Moreover, the daily mean (SD) number of MI deaths for people <65 years (non-aging group) and ≥ 65 years (aging group) was 3.4 (1.9) and 16.4 (5.7), respectively.

As outlined in Table 2, the daily mean (SD) exposures of PM₁, PM_{2.5}, SO₂, O₃, sulfate, nitrate, ammonium, OM, and BC were 30.5 (14.3) $\mu g/m^3$, 50.9 (34.9) $\mu g/m^3$, 18.8 (13.3) $\mu g/m^3$, 94.2 (39.6) $\mu g/m^3$, 9.0 (6.1) $\mu g/m^3$, 12.4 (10.2) $\mu g/m^3$, 7.8 (6.2) $\mu g/m^3$, 10.7 (7.2) $\mu g/m^3$, and 2.1 (1.4) $\mu g/m^3$, respectively. Moreover, nitrate (24.4%) and OM (21.0%) made up the majority of the overall PM_{2.5} mass, followed by sulfate (17.7%), ammonium (15.3%), and BC (4.1%). Furthermore, the daily mean (SD) *T* and *RH* were 14.1 (9.9) °C and 65.1 (14.9) %, respectively.

Table 3 summarizes the correlations among PM₁, PM_{2.5}, and PM_{2.5} chemical compositions. PM_{2.5} was significantly correlated with PM₁ (r_s = 0.857), sulfate (r_s = 0.935), nitrate (r_s = 0.965), ammonium (r_s = 0.971), OM (r_s = 0.972), and BC (r_s = 0.928).

As presented in Fig. S1, the results of single-day lagged effects indicated that the risks of MI mortality were positively linked to exposures to PM₁ (lag0 and lag1 days), PM_{2.5} (lag0 and lag1 days), sulfate (lag0 and lag1 days), nitrate (lag0 day), ammonium (lag0 and lag1 days), and BC (lag0 and lag1 days) in single-pollutant models. Fig. 1 depicted detailed results of percentage change with 95% CI, and the results of cumulative lagged effects showed that the risks of MI mortality were positively related to exposures to PM1, PM2.5, nitrate, ammonium, and BC at lag01 and lag02 days, and sulfate at lag01-lag03 days. The largest estimate effects were observed at lag02 day except for PM1. For lag02 day, each IQR rise in PM1, PM2.5, sulfate, nitrate, ammonium, and BC concentrations were associated with excess risks of 2.5% (95% CI: 0.2%, 4.9%), 2.0% (0.1%, 3.9%), 2.2% (0.3%, 4.1%), 2.0% (0.1%, 3.9%), 2.0% (0.1%, 4.0%), and 2.1% (0.2%, 3.9%) for MI mortality, respectively. However, deaths from MI were not significantly associated with exposure to OM.

Fig. 2 illustrates the associations between PM1, PM2.5, and PM2.5 chemical composition exposures and MI mortality at lag02 day stratified by sex, age, and season. No significant effects of PM1, PM2.5, and PM2.5 chemical composition exposures on excess risks of MI deaths were observed in males and females. Besides, significant effects of PM1, PM2.5, sulfate, nitrate, ammonium, OM, and BC exposures on excess risks of MI mortality were observed in aging populations, and the corresponding excess risks of MI mortality were 3.0% (0.6%, 5.4%), 2.5% (0.6%, 4.4%), 2.7% (0.8%, 4.6%), 2.3% (0.4%, 4.3%), 2.4% (0.5%, 4.4%), 2.1% (0.3%, 3.9%), and 2.4% (0.6%, 4.3%), respectively. Furthermore, significant effects of PM1, PM2.5, sulfate, and BC exposures on excess risks of MI mortality were observed in the cold season, and the corresponding excess risks of MI mortality were 2.6% (0.1%, 5.1%), 2.1% (0.0%, 4.2%), 2.2% (0.2%, 4.3%), and 2.2% (0.3%, 4.2%), respectively. However, the differences between effect estimates for aging and nonaging subgroups and cold and hot seasons were not statistically significant (all *P* for heterogeneity >0.05).

Fig. S2 manifests the effect estimates of PM₁, PM_{2.5}, and PM_{2.5} chemical compositions in two-pollutant models. We found the estimated effects were robust for PM₁, PM_{2.5}, sulfate, ammonium, OM, and BC when adjusted for SO₂, O₃, or PM_{2.5} mass, respectively. However, the effect estimates of nitrate on MI mortality tended to be not statistically significant when adjusted for O₃. In addition, results from Fig. S3 indicated that altering the *df* for *T* and *RH* into 4–6 had little impact on the findings.

4. Discussion

This study simultaneously evaluated the acute effects of PM_1 , $PM_{2.5}$, and $PM_{2.5}$ constituents on MI mortality, filling the gap regarding the association of PM_1 and $PM_{2.5}$ constituents with the risks of MI mortality. We observed that short-term exposure to PM_1 , $PM_{2.5}$, sulfate, nitrate, ammonium, and BC at lag0 or lag1 day significantly raised the risks of MI mortality. Additionally, PM_1 had a slightly stronger association with MI mortality than $PM_{2.5}$. Significant effects of PM_1 , $PM_{2.5}$, sulfate, and

Table 2

Distribution of daily air pollutants, PM_{2.5} constituents, and meteorological factors in Qingdao, China (2015–2019).

Variables	$\text{Mean}\pm\text{SD}$	Minimum	Median	Maximum	IQR	Percent PM _{2.5} mass (%)
Air pollutants						
$PM_1 (\mu g/m^3)$	30.5 ± 14.3	1.0	26.5	105.4	15.9	-
$PM_{2.5} (\mu g/m^3)$	50.9 ± 34.9	2.5	40.9	256.3	35.2	-
SO ₂ (μg/m ³)	18.8 ± 13.3	5.2	15.0	115.3	13.0	-
$O_3 (\mu g/m^3)$	94.2 ± 39.6	16.8	92.7	249.9	62.1	-
PM _{2.5} constituents						
Sulfate ($\mu g/m^3$)	9.0 ± 6.1	0.6	7.5	45.7	6.4	17.7
Nitrate ($\mu g/m^3$)	12.4 ± 10.2	0.3	9.4	70.5	10.4	24.4
Ammonium ($\mu g/m^3$)	$\textbf{7.8} \pm \textbf{6.2}$	0.3	6.0	46.0	6.5	15.3
OM $(\mu g/m^3)$	10.7 ± 7.2	0.4	8.8	55.8	6.9	21.0
BC (μg/m ³)	2.1 ± 1.4	0.1	1.8	10.6	1.3	4.1
Meteorological factors						
Temperature (°C)	14.1 ± 9.9	-12.2	15.1	31.1	18.1	-
Relative humidity (%)	65.1 ± 14.9	25.0	66.2	97.6	23.6	-

Abbreviations: SD, standard deviation; IQR, interquartile range; PM₁, particulate matter with an aerodynamic diameter \leq 1 µm; PM_{2.5}, particulate matter with an aerodynamic diameter \leq 2.5 µm; SO₂, sulfur dioxide; O₃, ozone; OM, organic matter; BC: black carbon.

Table 3

Correlations among PM1, PM2.5, and PM2.5 constituents in Qingdao, China (2015-2019).

Variables	PM_1	PM _{2.5}	Sulfate	Nitrate	Ammonium	OM	BC
PM ₁	1.000						
PM _{2.5}	0.857*	1.000					
Sulfate	0.744*	0.935*	1.000				
Nitrate	0.850*	0.965*	0.885*	1.000			
Ammonium	0.834*	0.971*	0.937*	0.983*	1.000		
OM	0.834*	0.972*	0.883*	0.897*	0.902*	1.000	
BC	0.794*	0.928*	0.855*	0.827*	0.851*	0.963*	1.000

Abbreviations: PM₁, particulate matter with an aerodynamic diameter $\leq 1 \mu m$; PM_{2.5}, particulate matter with an aerodynamic diameter $\leq 2.5 \mu m$; OM, organic matter; BC: black carbon; **P* < 0.001.



Fig. 1. Percentage change (with 95% confidence interval) in myocardial infarction mortality associated with an IQR increase in PM₁, PM_{2.5}, and PM_{2.5} constituents with different lag days in single-pollutant model. Abbreviations: IQR, interquartile range; PM₁, particulate matter with an aerodynamic diameter of 1 µm or less; PM_{2.5}, particulate matter with an aerodynamic diameter of 2.5 µm or less.

BC exposures on excess risks of MI mortality were observed in aging populations or during the cold season, although the difference between effect estimates for the aging and non-aging subgroups and cold and hot seasons were not statistically different.

The association of acute exposure to $PM_{2.5}$ with MI mortality has been reported in some previous studies in recent years (Chen et al., 2018; Lin et al., 2018). Two studies from 112 to 75 U.S. cities disclosed that a 10 µg/m³ rise in PM_{2.5} exposure at lag01 day was linked to elevated risks of 1.18% (0.48%, 1.89%) and 1.22% (0.62%, 1.82%) for MI deaths, respectively (Dai et al., 2014; Zanobetti and Schwartz, 2009). A study from Hong Kong, China also reported that each 10 µg/m³ increase in PM_{2.5} concentration at lag01 day was in association with a 2.35% (0.38%, 4.36%) increase in daily deaths from acute MI (Liang et al., 2018). However, most existing studies do not take PM₁, a smaller PM, into account. In comparison to PM_{2.5}, PM₁ is thought to pose more negative effects on human health on account of more toxin adsorption, increased surface-to-volume ratios, and higher vascular penetration (Chen et al., 2017b; Chen et al., 2020b; Fang et al., 2022; Wu et al., 2020; Yin et al., 2020). For example, Yin et al. reported that PM₁-related excess risks of CVD and stroke were 21.0% and 32.0% higher than those for PM_{2.5}, respectively (Yin et al., 2020). Chen et al. also reported the increased risks of ischemic stroke were higher related to a 10 μ g/m³ rise

	Subgrou	n	Percentage change (9	5%CI)				z	Р
		-P-	r oroontaige ontailige (o						
'M ₁	Gender	Male	2.7 (-0.1, 5.6)		· · · · ·	-		0.316	0.752
		Female	2.4 (-0.7, 5.6)	H	•	-			
	Age	<65	0.7 (-4.0, 5.6)		•	-		0.257	0.797
		≥65	3.0 (0.6, 5.4)			-			
:	Season	Cold	2.6 (0.1, 5.1)		·•	•		0.102	0.919
		Hot	5.5 (-2.9, 14.7)			•			
M _{2.5}	Gender	Male	1.9 (-0.3, 4.2)					0.107	0.915
		Female	1.9 (-0.6, 4.5)		—				
	Age	<65	-0.6 (-4.4, 3.3)					0.350	0.727
		≥65	2.5 (0.6, 4.4)		—				
)	Season	Cold	2.1 (0.0, 4.2)					0.066	0.947
		Hot	4.2 (-1.9, 10.6)		•		-		
ulfate	Gender	Male	2.2 (-0.1, 4.4)					0 044	0.965
unate	Centuer	Female	1.9 (-0.6, 4.3)					0.011	0.000
	Age	<65	-1.0 (-4.6, 2.8)					0.587	0.557
	Age	≥65	2.7 (0.8, 4.6)					0.007	0.001
1	Season		2.2 (0.2, 4.3)					0.006	0.995
	ocuson	Hot	3.3 (-1.6, 8.4)		•			0.000	0.000
litrate	O	Mala	00/00 4 0					0.004	0.000
Inate	Gender		2.0 (-0.2, 4.4)					0.064	0.933
	A	Female <65	1.7 (-0.8, 4.3)					0.004	0.000
	Age		-0.2 (-4.0, 3.8)					0.021	0.983
	0	≥65	2.3 (0.4, 4.3)					0.004	0.000
,	Season	Hot	1.9 (-0.1, 4.0) 4.7 (-1.4, 11.3)					0.091	0.928
mmonium	Gender		2.1 (-0.2, 4.5)		••••			0.108	0.914
		Female	1.7 (-0.9, 4.2)	F	• •				
	Age	<65	-0.6 (-4.4, 3.4)	•				0.211	0.833
		≥65	2.4 (0.5, 4.4)						
-	Season		2.0 (-0.1, 4.1)		••••			0.010	0.992
		Hot	4.8 (-1.0, 10.9)		•				
organic matter	Gender	Male	1.1 (-1.0, 3.3)	H				0.508	0.612
		Female	2.0 (-0.3, 4.4)		• • · · ·				
	Age	<65	-1.3 (-4.8, 2.4)	• • •				0.653	0.514
		≥65	2.1 (0.3, 3.9)		—				
	Season	Cold	1.7 (-0.3, 3.6)					0.025	0.980
		Hot	4.2 (-1.9, 10.7)		•		-		
Black carbon Ge	Gender	Male	1.8 (-0.3, 4.1)					0.507	0.612
		Female	2.2 (-0.2, 4.6)		—				
	Age	<65	-0.0 (-3.7, 3.8)		••			0.592	0.554
	-	≥65	2.4 (0.6, 4.3)		—				
	Season	Cold	2.2 (0.3, 4.2)		—			0.450	0.653
		Hot	4.3 (-2.2, 11.2)		•				
				-5	0 5	5 1	0 1	1 15	_

Fig. 2. Percentage change (with 95% confidence interval) in myocardial infarction mortality associated with an IQR increase in PM₁, PM_{2.5}, and PM_{2.5} constituents at lag02 day stratified by sex, age, and season in single-pollutant model. *Abbreviations: IQR, interquartile range; PM₁, particulate matter with an aerodynamic diameter of* 1 µm or less; PM_{2.5}, particulate matter with an aerodynamic diameter of 2.5 µm or less.

in PM₁ (1.4%, 0.5%–2.3%) than in PM_{2.5} (0.7%, 0.0%–1.4%) in China (Chen et al., 2020b). In this study, our results also indicated that PM₁ had slightly stronger detrimental impacts on MI mortality than PM_{2.5}. Specifically, each IQR increment in PM₁ and PM_{2.5} concentrations at lag02 day was linked to excess risks of 2.5% (95% CI: 0.2%, 4.9%) and 2.0% (0.1%, 3.9%) for MI mortality, respectively.

In addition to its particle size, the toxicity of PM is also attributed to its variability in constituents and emission sources of diverse PM profiles (Hassanvand et al., 2017; Kelly and Fussell, 2012; Lu et al., 2015). Among the water-soluble inorganic ions, sulfate, nitrate, and ammonium are the most abundant in PM_{2.5} mass. In this study, we found nitrate and ammonium made up 24.4% and 15.3% of the overall PM_{2.5} mass, respectively. Several studies from China reported lower percentages of nitrate and ammonium in Xi'an (nitrate: 8.4%; ammonium: 4.8%), Shanghai (nitrate: 15.9%; ammonium: 10.3%), Guangzhou (nitrate: 18.6%; ammonium: 12.6%), and nationwide level (nitrate: 22.3%; ammonium: 14.7%) (Cao et al., 2012; Lin et al., 2016; Qiao et al., 2014; Yang et al., 2020a). Higher levels of nitrate and ammonium in Qingdao may be due to residential coal combustion during the heating period and traffic and transportation during the non-heating period (Wu et al.,

2017; Xu et al., 2015). Numerous studies observed significant impacts of sulfate, nitrate, and ammonium on deaths from CVD. Yang et al. reported an IQR increase in sulfate, nitrate, and ammonium at lag03 day was related to increased risks of 0.8% (0.2%, 1.4%), 1.0% (0.2%, 1.8%), and 1.2% (0.4%, 1.9%) for cardiovascular mortality in China, respectively (Yang et al., 2020a). Lin et al. also reported per IQR increase in sulfate, nitrate, and ammonium exposures at lag03 day were significantly associated with elevated risks of 2.2% (1.1%, 3.4%), 2.0% (0.5%, 3.4%), and 3.4% (1.6%, 5.2%) for cardiovascular mortality in Guangzhou, China, respectively (Lin et al., 2016). However, the impacts of sulfate, nitrate, and ammonium exposures on MI death were scarce and inconsistent. Yang et al. reported that only ammonium was positively associated with MI mortality. For per IQR increase in ammonium exposure at lag03 day, the elevated risk of MI death was 1.3% (0.2%, 2.5%) in China (Yang et al., 2020a). According to a study from Guangzhou, China, there was no significant relationship between nitrate and MI death (Li et al., 2021). In this study, we observed positive relationships of MI mortality with sulfate, nitrate, and ammonium exposures at lag02 day, and the corresponding excess risks were 2.2% (0.3%, 4.1%), 2.0% (0.1%, 3.9%), 2.0% (0.1%, 4.0%) for MI mortality,

respectively. In addition, significant effects of sulfate exposure on excess risks of MI mortality were observed during the cold season, probably mainly due to increased sulfate exposure derived from residential coal used for heating in the winter (Bie et al., 2021).

BC, a pro-oxidant chemical component of PM2.5, is able to carry noxious species like polycyclic aromatic hydrocarbons, which induce oxidative stress and inflammation and may further lead to respiratory and cardiovascular diseases (Chowdhury et al., 2022; Zanobetti et al., 2014). BC contains a large fraction of elemental carbon (EC), and the terms of BC and EC are often used interchangeably (Smith et al., 2009). Little evidence has assessed the effect of short-term exposure to BC on MI death. In this study, we observed for the first time that per IQR increase in BC exposure at lag02 day resulted in an excess risk of 2.1% (0.2%, 3.9%) for MI mortality in Qingdao, China. Moreover, significant effects of BC exposure on excess risks of MI mortality were observed in the cold season. As previously reported, the highest concentrations of BC were noted during the winter in Qingdao and the Northern Yellow Sea, which may be on account of the monsoon climate in the coastal areas and increased domestic use of coal for heating throughout the winter (Bie et al., 2021; Wang et al., 2013). Furthermore, BC emissions are mainly derived from residential coal and biomass combustion, regional transport, and ship emissions in Qingdao (Bie et al., 2021; Chen et al., 2017a; Cui et al., 2021). Thus, more effective clean heating policies should be implemented to reduce BC pollution in Qingdao.

Previous evidence regarding the effects of PM and PM constituents among different sexes and age groups was inconsistent. Yang et al. found similar estimates of the effects of EC, organic carbon (OC), sulfate, nitrate, and ammonium at lag03 day between men and women in China (Yang et al., 2020a). A study from Shenyang, China reported females had higher relative risks of cardiovascular mortality related to PM1 and PM_{2.5} exposures than males (Ma et al., 2011). However, higher risk estimates of respiratory disease were observed in males than in females in terms of short-term exposures to EC, sulfate, and ammonium (Jo et al., 2018; Zhou et al., 2022). For age groups, many studies have reported the elderly are vulnerable subpopulations to PM2.5, sulfate, nitrate, and ammonium exposures (Liang et al., 2018; Tian et al., 2019; Yang et al., 2020a; Zhou et al., 2022). Nevertheless, a study from Guangzhou, China suggested that different age groups had similar risks of emergency department visits attributed to PM1 and PM2.5 exposures (Liu et al., 2021). According to our findings, significant effects of PM1, PM2.5, sulfate, nitrate, ammonium, OM, and BC exposures on excess risks of MI mortality were observed in aging populations. However, the difference between effect estimates for male and female and aging and non-aging subgroups were not statistically significant. Future studies incorporating larger sample sizes or more regions were called for verify our findings.

Furthermore, the potential biological mechanisms regarding the relationships of PM and its chemical compositions with deaths from MI have been proposed but not fully established. Numerous factors, such as oxidative stress, systemic inflammation, endothelial dysfunction, and thrombogenicity, may result in these negative consequences of PM and its constituents (Chen et al., 2020a; Lederer et al., 2021; Lin et al., 2016; Yang et al., 2018). Further investigations into the mechanisms are imperative to pinpoint the precise biological process underlying the relationship between PM and acute illness episodes, in particular for smaller particles and $PM_{2.5}$ chemical compositions.

This study first quantified the relationship of short-term exposures to PM_1 and $PM_{2.5}$ constituents with deaths from MI in China. The large sample size from the Qingdao resident death monitoring system, combined with Qingdao's relatively high level of air pollutants, enhanced our capacity to identify the acute effects of PM_1 , $PM_{2.5}$, and $PM_{2.5}$ constituents on MI mortality. Meanwhile, these findings were further proved to be reliable using two-pollutant models. However, there were several limitations in our research. Firstly, we used geographically mean daily ambient air pollutants data instead of personal air pollutant exposures in this study, which may lead to an underestimation of health

impacts linked to air pollutants. However, using a personal dosimeter was not feasible in this retrospective study. Secondly, it was unable to distinguish the associations from various size fractions and constituents because of the high correlations among PM_1 , $PM_{2.5}$, and its components. The experimental research needs to provide additional evidence for these findings. Thirdly, due to the lack of data, other $PM_{2.5}$ components linked to cardiovascular events, such as transition metals, were not taken into account in our research (Weichenthal et al., 2021). Finally, this study was conducted based on ecological design, which prevents us from concluding the causal connection.

5. Conclusions

Short-term exposures to PM_1 , $PM_{2.5}$, sulfate, nitrate, ammonium, and BC were positively linked to the risks of MI mortality. Significant effects of PM_1 , $PM_{2.5}$, sulfate, and BC exposures on excess risks of MI mortality were observed in aging subpopulations or during the cold season. Larger prospective multicenter studies are imperative to mine more rigorous evidence and clarify the underlying mechanisms.

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Credit author contribution statement

Xiaoyun Ma: Methodology, Writing – original draft. Haiping Duan: Resources, Data curation, Writing – review & editing. Hua Zhang: Formal analysis, Validation, Writing – review & editing. Xue Liu: Visualization, Writing – original draft. Xiaohui Sun: Formal analysis, Writing – review & editing. Jing Wei: Resources, Data curation. Min Zhao: Validation, Writing – review & editing. Bo Xi: Supervision, Funding acquisition, Writing – review & editing.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.atmosenv.2022.119478.

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