Health effects of $PM_{2.5}$ constituents and source contributions in major metropolitan cities, South Korea

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Abstract

South Korea is one of the East Asian countries suffering severe ambient $PM_{2.5}$ pollution which has been continuously reported to a risk factor driving both death and disability in many parts of the world. We investigated the associations of cause-specific mortality with both $PM_{2.5}$ chemical constituents and source contributions in four metropolitan cities, namely Seoul, Daejeon, Gwangju and Ulsan, by applying the generalized linear model (GLM) to the results of the positive matrix factorization (PMF) modeling. The cities represent each of four *air control zone* designated as of April 2020 by the special law for controlling air pollution on a regional basis in South Korea. We found that that the interquartile range (IQR) increase in the concentration $PM_{2.5}$ chemical constituents largely increased the relative risk (RR) of mortality, whereas the significance and strength of the associations were different among the cities. In addition, the effects of unit increase in source contributions also differed depending on regions. In conclusion, the results of this study suggest the $PM_{2.5}$ compositional characteristics may have influenced the heterogeneous health of $PM_{2.5}$ exposure among the cities. Therefore, the plans and policies based on the regional characteristics of $PM_{2.5}$ composition and health risk need to be implemented along with the reduction of total mass concentration of $PM_{2.5}$ for mitigating adverse health effects efficiently from a perspective of public health.

Introduction

Ambient particulate matters have become a one of the most concerning factors affecting human health in numerous large cities around the globe. Especially $PM_{2.5}$, defined as particle with aerodynamic diameter less than or equal to 2.5 ㎛, has been reported to have more severe impacts on human body than bigger particles due to physiochemical properties (Schins et al. 2004).

First, extremely small sizes lead $PM_{2.5}$ to both high mobility and difficulty of removal once inhaled into human body through breath. Unlike bigger airborne particulate matters, removed by mucociliary clearance, $PM_{2.5}$ can reach the deeper region of respiratory tract and retain in the lungs for longer periods of time. Thus $PM_{2.5}$ account for considerable proportions of particles observed in human pulmonary parenchyma and it is very difficult to be removed (Churg and Brauer 1997). Large surface area is another physical property of $PM_{2.5}$ explaining the relationship between the exposure to $PM_{2.5}$ and various health outcomes. The great specific surface area facilitates $PM_{2.5}$ to act as a carrier of highly toxic compounds such as polycyclic aromatic hydrocarbons (PAHs) and transition metals (Kong et al. 2010; Pandey et al. 2013). Furthermore, the chemical heterogeneity of $PM_{2.5}$ composition also plays an important role in affecting human health because each constituent of $PM_{2.5}$ has its own mechanisms and effects in terms of toxicity.

These properties of $PM_{2.5}$ mostly originate from the formation processes, especially secondary formation among various gaseous precursors in the atmosphere (Wang-Li 2015). Due to the complexity of formation processes among precursors, $PM_{2.5}$ is composed of various constituents including ionic compounds (e.g., sulfate, nitrate, ammonium), carbonaceous compounds (e.g., organic carbon, element carbon), trace elements (e.g., nicker, cadmium, arsenic) and other substances such as PAHs and volatile organic compounds (Ye et al. 2003; Dai et al. 2013; Amil et al. 2016). In addition, the proportions of each constituent vary considerably according to spatial and temporal conditions of the samples because $PM_{2.5}$ is highly source-dependent secondary aerosols (Cheng et al. 2012; He et al. 2012; Li et al. 2017).

In the similar context, the toxicity of $PM_{2.5}$ may differ spatiotemporally and it should be evaluated exactly considering chemical composition in the region of interest for developing relevant plans or policies to reduce health effects of $PM_{2.5}$. Nevertheless, many of administrative plans with regard to $PM_{2.5}$ have been mostly focused on the total mass of $PM_{2.5}$. The aims of this study are to investigate the effects of short-term exposure to $PM_{2.5}$ constituents on cause-specific mortality and find the association of source contributions with mortality in four major metropolitan cities, South Korea for controlling $PM_{2.5}$ from a perspective of public health.

Materials And Methods

Study population

As of April 2020, *Special act on the improvement of air quality in air control zone* became effective to manage the air quality efficiently and systemically on a regional basis in South Korea. According to the law, four *air control zone* of which air pollution is much severer than other region were designated for the country, namely Seoul metropolitan area, Middle area, Southern area, Southeastern area. The study populations were all residents in the four metropolitan cities during 2014-2018 shown in Figure 1 located in each *air control zone*.

Seoul (hereinafter SL) representing Seoul metropolitan area is located in the northwestern region of and is adjacent to Yellow Sea lying between mainland China on the west and north and the Korean peninsula on the east. Its area and population are 605 km^2 and around 9.6 million. Daejeon (hereinafter DJ), the largest city in Middle area, is located in the central region of the country and it had a population of 1.5 million in 2019. Gwangju (hereinafter GJ) of Southern area is a city in the southwest of the country and its population and area were approximately 1.5 million and 501 km^2 similar to those of Daejeon. Ulsan (hereinafter US), located in southeast part of the country and bordering the sea, had a population of 1.1 million in 2019 with total area of 1062 km^2. US where two huge industrial complexes exist is widely known as a city of heavy industry (Kim et al. 2018).

Air pollution and meteorological data

National Institute of Environmental Research (NIER) under the ministry of environment, South Korea has been operating several air pollution intensive monitoring stations (APIMS) across the country to for $PM_{2.5}$ speciation along with monitoring gaseous pollutants including nitrogen dioxide, sulfur dioxide. Ambient air samples are collected every day at APIMS for 24 h period and analyzed for chemical constituents. We obtained the measured values of $PM_{2.5}$ speciation during 2014-2018 at SL (37.61°, 126.93°), DJ (36.32°, 127.41°), GJ (35.23°, 126.85°) and US (35.58°, 129.32°) sites from NIER. Chemical constituents were water-soluble ions (e.g., sulfate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+)), carbonaceous compounds (i.e., organic carbon (OC), elemental carbon (EC)) and trace elements (e.g., titanium (Ti), vanadium (V), nickel (Ni)).

Meteorological parameters are also so important factors affecting human health including morbidity and mortality that they should be included in the health effect model for control (Allen and Sheridan, 2014; Wang et al. 2019). Thus, we collected daily average temperature, relative humidity (RH) and barometric pressure (BP) data during 2014-2018 via Weather Data Service (https://data.kma.go.kr) operated by Korea Meteorological Administration.

Health outcomes data

Various health related parameters including mortality, hospital emergency room visits, out-of-hospital cardiac arrest (OHCA) have been widely used to estimate the adverse effects of exposure to $PM_{2.5}$ on health in various epidemiological studies (Atkinson et al. 2014; Qiao et al. 2014; Pradeau et al. 2015). In the present study, we selected cause-specific daily mortality data as health outcomes. Daily death counts in the four cities were obtained from MicroData Integrated Service (https://mdis.kostat.go.kr) operated by the Statistics Korea. We included death counts for residents in each city as mortality data after classifying them into all-cause (non-accidental, NA, A00-R99), cardiovascular disease (CVD, I00-I99) and respiratory disease (RD, J00-J99) mortality according to 10th version of International Classification of Disease (ICD-10). This study was approved by the Seoul National University Institutional Review Board (IRB No: E2101/003-001).

PMF modeling

Among various receptor models based on chemical analyses, positive matrix factorization (PMF) and chemical mass balance (CMB) have been frequently performed for identifying the sources and estimating their contributions. Many of previous studies showed the results based on both PMF and CMB were quite comparable (Begum et al. 2007; Ke et al. 2008; Teixeira et al. 2015). But in terms of applicability, PMF is more flexible than CMB because PMF doesn't need source profiles for model execution. It is difficult to obtain suitable source profiles established for the region of interest although it strongly affects the source apportionment results. Therefore, we selected PMF to investigate the sources and their contribution for $PM_{2.5}$ in the study cities. Principles and more details of PMF can be found elsewhere (Song et al. 2006; Watson et al. 2008; Manousakas et al. 2017).

As for PMF input data, we constructed the concentrations and their corresponding uncertainty data for the chemical species according to the method described in the guidebook of US EPA PMF 5.0 (Norris et al. 2008). Data values below the method detection limit (MDL) were replaced with half of the MDL and 5/6 × MDL were used as their corresponding uncertainties. For the measured values greater than the MDL, the uncertainties were calculated by the following Eq (1) and we used 10% as error fraction of each $PM_{2.5}$ constituents.

$$\text{Uncertainty} = [(\text{Error Fraction} \times \text{Concentration})^2 + (0.5 \times \text{MDL})^2]^{1/2} \quad (1)$$

Whole datasets on days when the mass balance and the ion balance was not satisfactory were excluded from the input data. After obtaining best solutions we also conducted error estimation using displacement method (DISP) for acquiring uncertainty estimates for each factor of the solutions (Paatero et al. 2014).

Health effects analyses

Health effects of exposure to $PM_{2.5}$ can be expressed as the strength of the associations between $PM_{2.5}$ constituents and health outcomes via time-series analyses using various statistical models. Previous studies have revealed the effects of $PM_{2.5}$ on health-related outcomes including mortality, morbidity using generalized linear model (GLM), generalized additive model (GAM) and other statistical models (Pascal et al. 2014; Phung et al. 2018; Cai et al. 2019).

In the present study, we selected 15 chemical species and adopted GLM with a natural spline for analyzing the adverse effects of $PM_{2.5}$ constituent and source contributions on daily mortality. Death counts were modeled with either Poisson distribution or over-dispersed Poisson distribution according to their distribution in the study cities. We applied natural spline (ns) function of time and temperature for controlling both long-term and seasonal trends of death counts. Furthermore, we added $PM_{2.5}$ as a covariate for adjustment of the effects by total $PM_{2.5}$ mass. With respect to degrees of freedom (df) of the natural spline, we conducted sensitivity analyses by changing df from 2 to 10 per year to find the df minimizing the Akaike Information Criterion (Akaike 1974). Based on the sensitivity results we selected df=2 per year for temperature in all cities and df=2 per year for time was used in DJ, GJ and US while df=6 per year was used for SL. Furthermore, we also examined the delayed effect, single-lag effect from on the day of exposure (lag 0) to seven days (lag 7) after exposure, because lagging effects of the exposure to $PM_{2.5}$ have been steadily observed (Janssen et al. 2013; Chai et al. 2019; Li et al. 2021). The final model we constructed is following Eq (2).

$$\ln(E[Y_t]) = \beta_0 + \beta_1 \cdot X_{t-l} + \beta_2 \cdot PM_{2.5} + \beta_3 \cdot RH + \beta_4 \cdot BP + \beta_5 \cdot DOW$$

$$+ ns(time, df) + ns(temperature, df) + \varepsilon \quad (2)$$

where, Yt is the observed daily death counts on the day t, X_{t-l} represents the daily mean concentration of $PM_{2.5}$ chemical constituents at day t to l (l=0, 1 ,2 ..., 7); RH, BP, DOW stands for daily average relative humidity and barometric pressure, day of week respectively; β_1-β_5 is the coefficient of Xt, $PM_{2.5}$, RH, BP, DOW. After drawing β_1 from Eq (2), the relative risk (RR) was calculated by the following Eq (3).

$RR = e^{\beta 1}$ (3)

All analyses were performed by using 'mgcv' package in statistical software R version 4.0.1 (http://www.R-project.org)

Results And Discussion

Descriptive statistics of Death, Meteorological data and $PM_{2.5}$ composition

Tables 1 and 2 shows the descriptive statistics of explanatory variables for the four cities. Due to the largest population in SL, daily death counts were much larger in SL while those in the other cities were quite similar to each other. The proportions of CVD and RD death in total death in SL and DJ were comparable to those of the whole country (21.4% and 10.8%) whereas the CVD and RD death in GJ (26.3%, 15.8%) and US (30.8% and 15.4%) accounted for higher proportions of total death.

Table 1
Summary of daily mortality data and meteorological conditions in SL, DJ, GJ and US during 2014-2018.

SL	Avg. (S.D.)	Min.	P_{25}	Median	P_{75}	Max.	Unit	DJ	Avg. (S.D.)	Min.	P_{25}	Median	P_{75}	Max.
NA	108 (14)	69	98	107	115	182	counts	NA	17 (5)	5	14	17	20	33
CVD	24 (6)	8	20	23	27	44		CVD	4 (2)	0	2	3	5	11
RD	11 (4)	2	8	10	13	29		RD	2 (2)	0	1	2	3	8
Temp	13.3 (10.8)	-14.8	3.8	14.8	22.9	33.7	°C	Temp	13.7 (10.2)	-12.7	4.7	14.9	22.5	33.4
RH	59.4 (14.8)	21.8	48.4	59.5	69.5	99.8	%	RH	69.8 (13.8)	27.5	60.0	70.9	79.3	99.3
BP	1006.2 (7.9)	981	999.8	1006.5	1012.6	1026.8	hPa	BP	1008.6 (7.9)	985.7	1002.3	1008.9	1015.0	1029.1
GJ								US						
NA	19 (5)	6	16	18	22	36	counts	NA	13 (4)	2	9.25	12	14	28
CVD	5 (3)	0	3	4	6	13		CVD	4 (2)	0	2	3	4	13
RD	3 (2)	0	1	2	3	11		RD	2 (2)	0	0	1	2	8
Temp	14.7 (9.4)	-9.5	6.3	15.75	22.7	32.0	°C	Temp	14.7 (8.7)	-8.2	7.2	15.7	21.8	31.8
RH	69.0 (15.2)	23.9	58.2	69.8	79.5	99.0	%	RH	65.9 (19.4)	16.8	50.9	68.7	81.3	99.8
BP	1008.2 (7.8)	985.3	1001.9	1008.4	1014.4	1027.6	hPa	BP	1008.7 (8.1)	982.5	1002.6	1008.6	1014.6	1030

Table 2
Summary of $PM_{2.5}$ mass and chemical constituents concentration of study cities during 2014-2018.

SL	Avg. (S.D.)	Min.	P_{25}	Median	P_{75}	Max.	unit	DJ	Avg. (S.D.)	Min.	P_{25}	Median	P_{75}	Max.
$PM_{2.5}$	27.7 (17.2)	1.0	16.5	23.6	33.8	153.4	□/□	$PM_{2.5}$	29.2 (16)	1.71	18.1	26.5	36.7	117.2
SO_4^{2-}	5.4 (4.7)	0.0	2.3	4.1	6.8	39.1		SO_4^{2-}	5.2 (3.7)	0.2	2.5	4.2	6.9	28.3
NO_3^-	6.1 (5.9)	0.0	1.7	4.3	8.4	35.6		NO_3^-	5.4 (5.2)	0.1	1.5	3.6	7.8	31.2
Cl^-	0.3 (0.4)	0.0	0.1	0.2	0.5	2.6		Cl^-	0.4 (0.4)	0.0	0.1	0.2	0.5	2.8
Na^+	0.1 (0.1)	0.0	-	0.1	0.2	1.0		Na^+	0.2 (0.3)	0.0	0.1	0.2	0.3	2.5
NH_4^+	3.9 (3.2)	0.0	1.7	3.0	5.1	24.0		NH_4^+	3.6 (2.6)	0.1	1.8	3.1	4.8	18.0
K^+	0.2 (0.2)	0.0	-	0.1	0.2	1.0		K^+	0.2 (0.2)	0.0	0.1	0.1	0.2	1.5
OC	3.8 (1.9)	0.3	2.5	3.4	4.7	16.8		OC	4.5 (2.1)	0.6	3.0	4.2	5.7	16.5
EC	1.3 (0.6)	0.1	0.8	1.1	1.6	4.5		EC	1.4 (0.7)	0.3	0.8	1.2	1.7	4.9
S	3.1 (2.3)	0.1	1.5	2.4	3.9	23.3		S	3.9 (3.9)	0.1	1.5	2.6	4.8	40.1
Ca	65.8 (69.6)	1.5	29.4	48.5	80.9	1105.0	ng/□	Ca	58 (73.8)	1.5	16.5	36.4	75.5	1113
Ti	9.8 (7.8)	0.5	5.4	8.3	12.1	106.5		Ti	7.7 (8)	0.5	3.3	5.8	9.5	78.6
V	3.7 (4.6)	0.3	0.3	1.8	5.0	31.6		V	3.0 (2.6)	0.3	1.1	2.3	4.2	16.9
Cr	1.3 (1)	0.3	0.3	1.1	1.7	7.5		Cr	1.2 (1.2)	0.3	0.3	0.9	1.6	10.1
Mn	12.3 (8.5)	0.3	6.5	10.1	15.7	56.1		Mn	10 (6.6)	0.3	5.2	8.6	13.4	39.9
Fe	195 (112.2)	20.9	121	172.8	242.9	1074.5		Fe	183 (113.9)	16.6	103.7	160	234.3	1002.7
Ni	1.6 (1.6)	0.2	0.4	1	2.2	10.1		Ni	1.6 (1.4)	0.2	0.7	1.3	2.0	16.8
Cu	7.5 (5.2)	1.0	3.8	6.6	9.8	35.0		Cu	7 (4.4)	1.0	3.8	6.1	9.3	33.6
Zn	65.5 (43.6)	2.5	35.4	54	82.7	302.3		Zn	50.7 (32)	2.2	27.2	43.9	66	254.0
As	4.0 (3.3)	0.3	1.7	3.4	5.6	25.7		As	2.7 (2.6)	0.3	0.6	1.9	4.0	20.0
Se	1.2 (1.2)	0.3	0.3	0.9	1.6	8.6		Se	1.8 (1.4)	0.3	0.8	1.5	2.5	8.5
Br	8.9 (7)	0.1	4.2	6.9	11.0	63.1		Br	7.3 (5.8)	0.1	3.2	5.9	9.8	48.6
Pb	21.8 (15.7)	0.7	11.9	18.2	27.7	132.9		Pb	18.8 (13.9)	0.7	8.7	15.3	24.7	115.3

Table 2
(continued)

GJ	Avg. (S.D.)	Min.	P_{25}	Median	P_{75}	Max.	unit	US	Avg. (S.D.)	Min.	P_{25}	Median	P_{75}	Max.
$PM_{2.5}$	26.9 (15.8)	3.3	16.2	23.5	33.5	132.9	□/□	$PM_{2.5}$	20.4 (11.8)	2	11.7	17.8	26.1	88.3
SO_4^{2-}	5.4 (3.9)	0.3	2.6	4.3	7.0	32.2		SO_4^{2-}	4.1 (3.1)	0.4	1.9	3.1	5.3	24.8
NO_3^-	4.6 (4.8)	0.1	1.4	3.0	6.3	41.1		NO_3^-	3.0 (3.0)	0.1	0.9	2.0	3.9	21.8
Cl^-	0.6 (0.5)	0.0	0.2	0.5	0.8	3.0		Cl^-	0.2 (0.2)	0.0	0.1	0.2	0.3	1.4
Na^+	0.2 (0.1)	0.0	0.1	0.1	0.2	1.3		Na^+	0.1 (0.1)	0.0	0.1	0.1	0.2	0.8
NH_4^+	3.7 (2.5)	0.2	1.9	3.1	4.7	19.3		NH_4^+	2.5 (1.8)	0.2	1.2	2.1	3.3	14.5
K^+	0.2 (0.2)	0.0	0.1	0.1	0.2	3.1		K^+	0.1 (0.2)	0.0	0.0	0.1	0.1	2.0
OC	4.1 (2.2)	0.2	2.5	3.7	5.3	17.5		OC	3.1 (1.6)	0.1	1.9	2.8	4.1	9.5
EC	1.1 (0.6)	0.1	0.8	1.0	1.4	3.7		EC	0.8 (0.4)	0.1	0.4	0.7	1.0	3.4
S	2.3 (1.7)	0.1	1.2	1.9	3.0	13.0		S	5.2 (3.9)	0.5	2.5	4.0	6.9	27.6
Ca	58.1 (81.8)	1.5	18.9	33.0	63.2	912.6	ng/□	Ca	51.9 (51)	1.5	23.5	36.3	61.4	581.4
Ti	8.5 (9.7)	0.5	3.6	5.6	9.7	103.9		Ti	9.1 (8.8)	0.5	4.0	6.8	11.2	89.8
V	3.5 (3.1)	0.3	1.3	2.8	4.8	23.5		V	7.7 (11)	0.3	1.1	3.1	9.1	86.6
Cr	0.8 (0.7)	0.3	0.3	0.3	1.1	7.1		Cr	2.0 (1.3)	0.3	1.0	1.7	2.8	9.0
Mn	11.9 (7.9)	0.3	6.4	10.3	15.5	52.6		Mn	18.6 (14.7)	0.3	7.9	15.3	25.3	131.0
Fe	177 (118.3)	12.1	103.4	149.1	216.9	1129.8		Fe	197 (128.4)	10.3	109	169.8	255.9	1177.6
Ni	1.3 (1.1)	0.2	0.5	1.0	1.7	9.8		Ni	3.0 (3.6)	0.2	0.8	1.6	3.6	25.8
Cu	4.0 (3.6)	1.0	2.1	3.3	5.1	46.3		Cu	6.7 (4.6)	1.0	3.8	5.7	8.5	56.0
Zn	57.1 (35.3)	2.4	32.2	48.7	73.2	230.4		Zn	64.3 (53.1)	0.6	29.9	52	81.6	487.5
As	3.7 (2.2)	0.3	2.1	3.3	4.7	16.0		As	5.9 (7.5)	0.3	1.5	3.2	7.1	64.9
Se	1.3 (1.1)	0.3	0.3	1.0	1.8	6.6		Se	1.5 (1.6)	0.3	0.3	1.0	2.1	10.8
Br	8.8 (6.1)	0.3	4.4	7.3	11.9	43.5		Br	8.0 (5.7)	0.4	4.1	6.5	10.6	55.4
Pb	19.7 (17.3)	0.7	7.5	15.0	26.9	115.4		Pb	17.7 (15.4)	0.7	7.1	13.4	23.6	121.7

During the study period, the daily average temperature (℃) ranged from -14.8 to 33.7 (SL), -12.7 to 33.4 (DJ), -9.5 to 32.0 (GJ), -8.2 to 31.8 (US), which shows substantial temporal variation in all cities while the five-year average temperatures (℃) were 13.3, 13.7, 14.7 and 14.7 in SL, DJ, GJ and US respectively. The daily average relative humidity (%) ranged 21.8-99.8, 27.5-99.3, 23.9-99.0 and 16.8-99.8 in SL, DJ, GJ and US respectively.

The average concentrations of $PM_{2.5}$ mass at all sites exceeded both World Health Organization guidelines (5 μg/m^3 of annual mean concentration) and U.S. Environment Protection Agency standards (15 μg/m^3 of annual mean concentration). This suggests the population in four cities have been exposed to potential health risk during 2014-2018. It is notable that Ulsan showed approximately 25-30% lower $PM_{2.5}$ concentration than the other cities.

$PM_{2.5}$ chemical compositions showed considerable spatial variability. Secondary inorganic aerosols (SIA; i.e., SO_4^{2-}, NO_3^-, NH_4^+) constructed the total mass at a range of 46.9% (US)-55.6% (SL), which indicates secondary aerosol formation were a important factor in $PM_{2.5}$. In addition, the SO_4^{2-}/NO_3^- ratio was much lower in SL (0.88) than those in DJ (0.97), GJ (1.16) and US (1.38), which implies the dominant gas-phase pollutants were different across the cities. Carbonaceous species, reported to have second largest proportions in $PM_{2.5}$ mass in South Korea (Son et al. 2012; Bae et al. 2020), accounted for 18.2% (SL), 20.1% (DJ), 19.6% (GJ) and 18.4% (US) respectively. The proportions of trace elements were much higher in US (27.4%) than in SL (12.5%), DJ (14.5%) and GJ (10.0%), considered to reflect the industrial characteristics of US.

Source apportionment $PM_{2.5}$

A total of 1,438, 1,278, 1,419 and 1,211 $PM_{2.5}$ speciation datasets were used for identifying $PM_{2.5}$ sources and their contributions for SL, DJ, GJ and US respectively. We chose the number of factors based on the evaluation of model results including Q-value, residual distribution and the coefficient of determination and we confirmed source profiles in the study cities were physically meaningful and understandable.

Nine-factor solution for DJ and ten-factor solution for SL, GJ and US solution were drawn while source contributions were quite different among cities although the resolved factors were identical. Sources and their contributions to $PM_{2.5}$ are listed in Table 3 and source profiles for SL, DJ, GJ and US obtained from PMF model are displayed in Figures S1-S4 respectively.

Table 3
Sources with their contributions to $PM_{2.5}$ in SL, DJ, GJ and US during 2014-2018

Source	SL		DJ		GJ		US	
	μg/m^3	%	μg/m^3	%	μg/m^3	%	μg/m^3	%
Secondary nitrate	6.4	20.3	5.7	19.5	5.2	19.5	3.6	18.9
Secondary sulfate	6.9	21.8	5.7	19.7	5.7	21.3	4.5	23.9
Mobile	7.1	22.3	6.3	21.7	5.5	20.5	4.7	24.9
Biomass burning	1.4	4.3	0.8	2.7	0.9	3.2	0.7	3.6
District heating	2.8	8.7	2.6	8.9	3.3	12.2	0.7	3.7
Soil	0.8	2.6	1.2	4.2	1.1	4.2	1.0	5.3
Industry	1.4	4.4	1.9	6.6	1.2	4.3	1.7	8.8
Coal combustion	3.4	10.7	3.1	10.7	2.6	9.7	0.7	3.5
Oil combustion	0.9	2.9	1.8	6.1	0.6	2.2	0.6	3.4
Aged sea salt	0.7	2.1	-	-	0.8	2.9	0.7	3.9

In SL, secondary nitrate, secondary sulfate and mobile source made up the majority (64.4%) of the $PM_{2.5}$ mass concentration, consistent with the past studies conducted for SL (Heo et al. 2009; Moon et al., 2010). Mobile was the largest contributor followed by secondary sulfate, secondary nitrate and coal combustion, which implies sources related to secondary formation were dominant for $PM_{2.5}$. Especially sulfur related sources, secondary sulfate and coal combustion, were thought to be affected by several huge coal-fired power plants (e.g., Taean 6100 MW, Dangjin 6040 MW, Yeongheung 5080 MW) located in the southwest part of the city. These results are supported by previous studies which also identified those coal power stations as attributable factors to $PM_{2.5}$ in SL by analyzing polycyclic aromatic hydrocarbons (Kang et. al. 2020) and executing dispersion modelling (Kim et. al. 2016).

Similar to SL, $PM_{2.5}$ mass concentration in DJ was mostly contributed by four factors; mobile (21.7%), secondary sulfate (19.7%), secondary nitrate (19.5%) and coal combustion (10.7%) while aged sea salt source was not solely resolved due to the location in deep land. In GJ secondary sulfate (21.3%) was the largest contributor followed by mobile (20.5%) and secondary nitrate (19.5%) source and those major factors in total explained 61.3% of $PM_{2.5}$ mass concentration.

In US, the two largest factors were mobile (24.9%) and secondary sulfate (23.9%) sources which explained $PM_{2.5}$ mass bit more than in other cities, assumed to reflect the characteristics of the city where both huge heavy industrial complex and industrial ports exist. The annual emission of SOx in the city during 2014-2017 was 48,433 ton on average more than two times of the national average annual emission (19,516 ton). In the same context the contribution of industry (8.8%) was much bigger than that in the other cities and it was fourth-largest factor in the city.

Association of $PM_{2.5}$ chemical constituents and source contributions with mortality

Using PMF-modeled source contributions as well as $PM_{2.5}$ speciation data, we analyzed the effects of the $PM_{2.5}$ chemical constituents of $PM_{2.5}$ and sources on cause-specific mortality. Overall, the IQR increase in the concentration of each constituents influenced all cause-specific mortality whereas only a few constituents were significantly associated with the mortality with different strength depending on city and cause of death, which shows risk heterogeneity among various $PM_{2.5}$ constituents. In addition, the impacts of source contributions on mortality also varied among the cities. The associations of cause-specific mortality with both $PM_{2.5}$ constituents and sources are summarized in Tables S1-S2 respectively and Table 4 shows the highest RR of mortality significantly associated with chemical constituents.

Table 4
The highest RR and 95% CI of the significant associations of cause-specific mortality with $PM_{2.5}$ constituents in SL, DJ, GJ and US.

City	Cause	Constituents	Lags	Constituents	Lags	Constituents	Lags	Constituents	Lags
SL	NA	K^+ (1.016) (1.002, 1.031)	7	Ti (1.011) (1.003, 1.019)	7	EC (1.015) (1.000, 1.031)	2	-	-
	CVD	Ti (1.016) (1.001, 1.032)	7	Fe (1.023) (1.003, 1.044)	7	As (1.030) (1.003, 1.058)	1	-	-
	RD	Pb (1.071) (1.013, 1.128)	0	-	-	-	-	-	-
DJ	NA	EC (1.051) (1.004, 1.100)	2	Fe (1.039) (1.009, 1.069)	2	-	-	-	-
	CVD	OC (1.146) (1.057, 1.239)	4	V (1.080) (1.007, 1.154)	1	Mn (1.097) (1.027, 1.167)	1	Ni (1.042) (1.005, 1.080)	1
		Zn (1.076) (1.005, 1.148)	1	Pb (1.091) (1.006, 1.177)	1	-		-	-
	RD	OC (1.219) (1.090, 1.354)	5	As (1.190) (1.007, 1.384)	0	-		-	-
GJ	NA	Ti (1.015) (1.001, 1.029)	3	-	-	-	-	-	-
	RD	SO_4^{2-} (1.085) (1.009, 1.163)	1	NH_4^+ (1.071) (1.000, 1.143)	1	Ti (1.048) (1.009, 1.088)	5	V (1.109) (1.040, 1.179)	1
		Fe (1.061) (1.001, 1.120)	5	Ni (1.100) (1.033 1.170)	1	Zn (1.078) (1.010, 1.146)	1	-	-
US	NA	SO_4^{2-} (1.047) (1.004, 1.091)	4	OC (1.056) (1.001, 1.113)	4	Mn (1.060) (1.018, 1.103)	0	-	-
	CVD	OC (1.126) (1.009, 1.248)	4	-	-	-	-	-	-

In SL, the chemical constituents showing significant associations were as follows: K^+ (RR 1.016, 95% CI 1.002-1.032), EC (1.015, 1.000-1.031) and Ti (1.011, 1.003-1.019) with NA mortality, Ti (1.016, 1.001-1.032), Fe (1.023, 1.003-1.044) and As (1.030, 1.003-1.058) with CVD mortality and Pb (1.071, 1.013-1.128) with RD mortality. Ti and Fe, mostly originated from soil dust, are known to generate reactive oxygen species which lead to cell damage (Moreno et al. 2019). As and Pb, markers of coal combustion, are toxic even at low levels and they are known as carcinogen to human by International Agency for Research on Cancer. Significant associations of mortality with sources were mostly found for CVD mortality: district heating (1.032, 1.007-1.057), coal combustion (1.034, 1.003-1.065), soil (1.016, 1.000-1.032). Overall, the results in SL showed that the health effects were closely linked to transition metals from both natural and combustion-derived sources. Transition metals have impacts on cardiovascular via both direct and indirect pathways inducing oxidative stress and inflammation (Mills et al. 2009). In addition, respiratory system is subject to damage by those metals which trigger oxidative stress in lung alveoli (Donaldson et al. 1996).

In DJ we found $PM_{2.5}$ constituents increased the RR of all types of mortality but most of the significant associations were related to CVD mortality. Constituents showing significant associations were EC (1.051, 1.004-1.100) and Fe (1.039, 1.009-1.069) with NA mortality, OC (1.146, 1.057-1.239), V (1.080, 1.007-1.154), Mn (1.097, 1.027-1.167), Ni (1.042, 1.005-1.080), Zn (1.076, 1.005-1.148) and Pb (1.091, 1.006-1.177) with CVD mortality and OC (1.219, 1.090-1.354) and As (1.190, 1.007-1.384) with RD mortality. Beside transition metals mentioned earlier, OC and EC have been consistently reported to influence on various indicators of cardiovascular disease such as blood pressure, heart rate variability (Huang et al. 2012; Schneider et al. 2012; Wu et al. 2013) which can lead to cardiovascular health outcomes (Bell et al. 2009; Ito et al. 2011). Accordingly, sources with carbonaceous species and transition metals were thought to be major factors associated with mortality in DJ.

Significant associations in GJ were mostly found for RD mortality, which was uniquely observed among the four cities. Chemical species significantly associated with RD mortality were ionic species, SO_4^{2-} (1.085, 1.009-1.163), NH_4^+ (1.071, 1.000-1.143), as well as transition metals including Ti (1.048, 1.009-1.088), V (1.109, 1.040-1.179), Fe (1.061, 1.001-1.120), Ni (1.100, 1.033-1.170), Zn (1.078, 1.010-1.146). While several previous studies revealed the significant association SO_4^{2-} with the risk of mortality, there are not explicit biological mechanisms of SO_4^{2-} (Ueda et al. 2016). Nevertheless, some plausible theories have been suggested to explain the positive association of SO_4^{2-} with adverse health effects. First, particle acidity by SO_4^{2-} may change the pulmonary toxicity of other $PM_{2.5}$ constituents or physical properties from their own toxicity (Dreher 2000). Second, catalyzation of metals into more bioavailable forms is another possible explanation for the high associations of SO_4^{2-} with health effects (Lay et al. 2001). Regarding source contributions, sources of the species with significant association (e.g., coal combustion, mobile) were revealed to increase the RD mortality in GJ.

In US, SO_4^{2-} (1.047, 1.004-1.091), OC (1.056, 1.001-1.113) and Mn (1.060, 1.018-1.103) for NA mortality and OC (1.126, 1.009-1.248) for CVD mortality increased the RR significantly. In addition, secondary sulfate (1.052, 1.007- 1.098), mobile (1.053, 1.010-1.098) and industry (1.059, 1.017-1.102) sources were strongly associated with NA or RD mortality. These results were in consistent with the source profiles from PMF modelling which identified higher contribution of mobile and secondary sulfate and the composition of $PM_{2.5}$ compositional characteristic with the highest SO_4^{2-}/NO_3^- ratio among four cities.

Conclusion

In the present study, we investigated the associations of cause-specific mortality with the IQR increase in the concentration of $PM_{2.5}$ constituents and source contributions in four metropolitan cities in South Korea. The significance and strength of associations varied across the study cities, which implied the adverse health effects of short-term exposure to $PM_{2.5}$ were spatially heterogeneous. In SL significant associations were found for both NA and CVD mortality mostly related to transition metals and relevant sources including soil and coal combustion. In DJ most of the significant associations were found for CVD mortality with $PM_{2.5}$ constituents from combustion related sources including mobile, oil combustion and industry. However, many of significant associations in GJ were found for RD mortality with constituents related to secondary formation (e.g., SO_4^{2-}, NH_4^+) and heavy metals. Lastly, we found significant associations in US were closely related to SO_4^{2-}, OC and Mn in line with the characteristic of the city where large heavy and chemical industry complexes are located in.

From the results, we drew a conclusion that the risk of mortality increased by short-term exposure to $PM_{2.5}$ were quite heterogeneous. Therefore, identifying $PM_{2.5}$ constituents and source contributions affecting health outcomes significantly in the region of interest should takes priority over designing policies for controlling $PM_{2.5}$ efficiently. In addition, the policies, currently focused on $PM_{2.5}$ mass concentration, need to be shifted to health risk-based ones for protecting public health from air pollution.

There is still limitation of the present study to be improved in further studies. Although ambient $PM_{2.5}$ compositional data was employed as a proxy of personal exposure to $PM_{2.5}$ and we acquired the data from one site for each city due to the insufficient number of APIMS. As the area one APIMS cover is broader, the exactness of estimating the level of $PM_{2.5}$ exposure decrease, which may result in inevitable errors in evaluating the health effects. Accordingly, more sites for $PM_{2.5}$ speciation need to be established considering relevant indexes such as population, administrative area for a better assessment of $PM_{2.5}$ exposure.

Declarations

Ethics approval and consent to participate Not applicable.

Consent for publication Not applicable.

Data Availability All data generated or analyzed during this study are included in this published article and its supplementary information files.

Competing interests The authors declare that they have no competing interests

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Authors' contributions Ho Kim and Seung-Muk Yi supervised study design; Inho Song, Dae-Gon Kim and Kwonho Jeon contributed to data collection and verification; Sangcheol Kim, Juyeon Yang performed data analyses and wrote the first draft of the manuscript; Sangcheol Kim, Juyeon Yang, Jieun Park, Inho Song, Dae-Gon Kim, Kwonho Jeon, Ho Kim and Seung-Muk Yi contributed to interpretation of the data. All authors read and approved the final manuscript.

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Figures

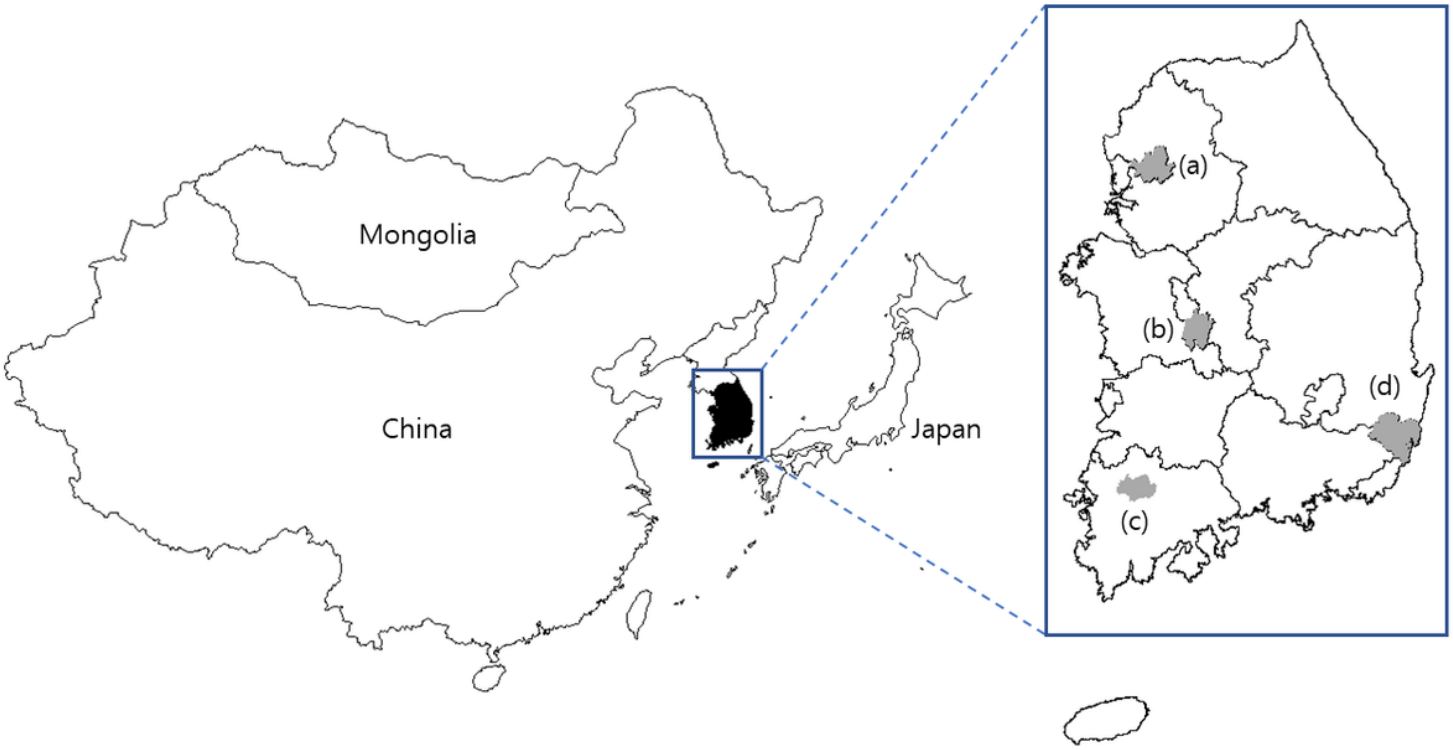

Figure 1

Geographical locations of the study cities; (a) SL, (b) DJ, (c) GJ and (d) US

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