




Characteristics of PM_{2.5} and its chemical constituents in Beijing, Seoul, and Nagasaki

Eun Ha Park¹ · Jongbae Heo²  · Setsuko Hirakura³ · Masahiro Hashizume³ · Furong Deng⁴ · Ho Kim^{2,5} · Seung-Muk Yi^{1,2}

Received: 16 May 2018 / Accepted: 15 August 2018 / Published online: 24 August 2018
© Springer Nature B.V. 2018

Abstract

Ambient fine particulate matter (PM_{2.5}) samples were collected from September 2013 to May 2015 in three cities in East Asian countries (Beijing, China; Seoul, South Korea; and Nagasaki, Japan) in order to analyze the spatiotemporal trends of PM_{2.5} chemical constituents including organic matter (OM), elemental carbon (EC), water-soluble inorganic ions (NO₃⁻, SO₄²⁻, and NH₄⁺), and trace elements. The average PM_{2.5} mass concentration were 125 ± 6.80 μg m⁻³, 44.6 ± 0.84 μg m⁻³, and 17.4 ± 0.37 μg m⁻³ in Beijing, Seoul, and Nagasaki, respectively. Higher carbonaceous concentrations were observed during winter in Beijing and Seoul, while higher concentrations were found during spring in Nagasaki. The highest seasonal averages of organic carbon (OC) to EC ratios were found during spring in Beijing, winter in Seoul, and fall in Nagasaki. The concentrations of secondary OC and its ratio to OC were high during fall and winter. For ion species, NO₃⁻ was dominant in Beijing and Seoul, while SO₄²⁻ was dominant in Nagasaki. Increased contributions of mobile sources in Beijing and Seoul were observed, with higher NO₃⁻/SO₄²⁻ ratios than those in Nagasaki. Three groups of air masses were found for the three cities using cluster analyses based on 72-h backward trajectories. The cluster from the Bohai economic zone had the highest concentration of PM_{2.5} for Beijing. For Seoul, a cluster that originated from the Yellow Sea near an industrial area in Liaoning Province and passed through a highly polluted industrial area in southwestern Seoul had high PM_{2.5} concentrations. A long-range transported cluster that originated in and crossed through heavily industrialized areas in China and South Korea for Nagasaki had higher ion species concentrations. The results of this study are useful to identify the current levels of PM_{2.5} and its chemical properties to establish a control plan for PM_{2.5} for Northeast Asia, including China, South Korea, and Japan.

Electronic supplementary material The online version of this article (<https://doi.org/10.1007/s11869-018-0616-y>) contains supplementary material, which is available to authorized users.

✉ Jongbae Heo
jongbae heo@gmail.com

✉ Seung-Muk Yi
yiseung@snu.ac.kr

¹ Department of Environmental Health Sciences, Graduate School of Public Health, Seoul National University, 1 Gwanak-ro, Gwanak-gu, Seoul 08826, Republic of Korea

² Institute of Health and Environment, Seoul National University, 1 Gwanak, Gwanak-ro, Gwanak-gu, Seoul 08826, Republic of Korea

³ Institute of Tropical Medicine, Nagasaki University, 1-12-4 Sakamoto, Nagasaki 852-8523, Japan

⁴ Department of Occupational and Environmental Health Sciences, Peking University School of Public Health, No. 38 Xueyuan Road, Beijing 100191, China

⁵ Department of Public Health Sciences, Graduate School of Public Health, Seoul National University, 1 Gwanak-ro, Gwanak-gu, Seoul 08826, Republic of Korea

Keywords PM_{2.5} · Organic matter · Inorganic ions · Cluster analysis · Northeast Asia

Introduction

Ambient PM_{2.5}, which is a major air pollutant, is mainly comprised of organic matter (OM), elemental carbon (EC), ion species, and trace elements. Exposure to PM_{2.5} is more associated with increases in cardiopulmonary health outcomes than exposure to larger particles (Dockery et al. 1993; Schwartz et al. 1993), and approximately 12% of premature deaths that are related to respiratory illnesses and cancers are attributed to PM_{2.5} exposure (Zhang et al. 2017a, b, c). Unusual high concentration events of PM_{2.5} have been frequently observed in East Asian countries, and their annual average PM_{2.5} levels are higher than the WHO's PM_{2.5} standards (25 μg m⁻³ for 24-h and 10 μg m⁻³ for annual average concentration) (Xie et al. 2016). The rapid development of China's economy has resulted in an increase in energy consumption, especially coal-dependent energy, and an increased

number of vehicles; this has led to an increase in anthropogenic emissions of $PM_{2.5}$ in megacities. Thus, $PM_{2.5}$ exposure is one of major public health problems in China (Chan and Yao 2008; Zhang et al. 2017a, b, c).

Long-range transported air pollutants from China have largely impacted $PM_{2.5}$ levels in many urban environments downwind of China, including South Korea and Japan. Several previous studies have demonstrated that health outcomes related to air pollution in South Korea and Japan, such as premature mortality, are influenced by both local emissions and long-range transported air pollution (Anenberg et al. 2014; Zhang et al. 2017a, b, c and the references there in).

To create a $PM_{2.5}$ management plan that is optimized for East Asia, the levels of $PM_{2.5}$ concentrations and the distribution characteristics of its chemical constituents should be identified. Several studies have shown results for chemical constituents at different sampling sites in different cities or countries. Some of these studies examined specific components such as organic matters or polycyclic aromatic hydrocarbons, (Jimenez et al. 2009; Hayakawa 2016) or measured $PM_{2.5}$ and compared results from other papers conducted $PM_{2.5}$ in different cities during the same period (Shimada et al. 2016). In this study, $PM_{2.5}$ and its major chemical constituents were measured in three East Asian cities and the results were compared to identify the current levels of $PM_{2.5}$ and its chemical properties between the cities. Finally, transportation pathways were identified through cluster analyses in order to better understand the characteristics of long-range transportation and contributing emission sources.

Materials and methods

Sampling

Ambient $PM_{2.5}$ and its chemical species were observed at the central location of Beijing and Seoul and at the suburban location of Nagasaki (Figure S1). The Beijing site was the roof (7th floor, ~21 m above ground) of the Department of Occupational and Environmental Health Sciences at Peking University School of Public Health (39.983° N, 116.355° E), which is surrounded by commercial buildings and is situated 100 m from the Xueyuan Road. The Seoul site was the roof (5th floor, ~17 m above ground) of the School of Public Health at Seoul National University (37.581° N, 127.001° E) and hence it was amenable to observe several types of pollutant emissions, such as residential, commercial, and traffic emissions. The Nagasaki site was the roof (4th floor, ~12 m above ground) of the Institute of Tropical Medicine at Nagasaki University (32.772° N, 129.868° E). Although Nagasaki is less urbanized than Beijing and Seoul, the sampling site is surrounded by residential and commercial areas, which makes the site to be suitable for representing the city.

The 24-h integrated $PM_{2.5}$ samples were collected on one in 3-day schedules from November 2014 to April 2015 in Beijing, from September 2013 to May 2015 in Seoul, and from February 2014 to May 2015 in Nagasaki. These samples were collected using a three-channel system, which was composed of three filter packs (URG Corp.), similar to the EPA Compendium Method IO-4.2 (Zhang et al. 2017a, b, c). Each channel was composed of a size-selective inlet, cyclone, vacuum pump, and dry gas meter. Based on the flow rate which was monitored for each channel using independent dry gas meters, cyclones (URG-2000-30EH and URG-2000-30EN, URG) provide a particle size cutoff. Flow rate for ionic species was 10 L/min and that of carbonaceous species and trace elements were 16.7 L/min.

Although the measurements were obtained under the same conditions in three cities, some limitations were inevitable. Unlike Seoul and Nagasaki, where the measurement periods covered all four seasons, measurements in Beijing were conducted only for 6 months. The “Comparisons of $PM_{2.5}$ between Beijing, Seoul, and Nagasaki” section describes our effects to overcome this limitation.

Chemical analysis

Filters were pre-treated at a laboratory in Seoul and sent to each of the sampling sites. The $PM_{2.5}$ mass was measured by weighing Teflon filters (47 mm Pall Life Sciences, 1 μ m pore size) with a microbalance (AND HM-202, precision 10^{-2} mg), and the filters were analyzed using X-ray fluorescence (XRF, XEPOS 03 Plus gas, SPECTRO) for trace elements. Water-soluble ion species (NO_3^- , SO_4^{2-} , and NH_4^+) were collected on Zefluor membrane filters (47 mm Pall Life Sciences, 1 μ m pore size) and analyzed using ion chromatography (Thermo Fisher Scientific, ICS-1100). Organic carbon (OC) and elemental carbon (EC) were collected on quartz microfiber filters (47 mm Pall Life Sciences, PALLFLEX Membrane Filters) and analyzed using thermal/optical transmittance with the National Institute of Occupational Safety and Health 5040 method (Thermal/Optical Carbon Aerosol Analyzer, Sunset Laboratory Inc.) (Winberry et al. 1999). All the filter samples and field blanks were safely sealed, frozen, and sent to the laboratory at the Seoul National University, where the chemical analyses were performed.

Trajectories calculation and cluster analysis

The Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) 4 model (Draxler 2010; Wang et al. 2014) was used with one of the meteorological data Global Data Assimilation System (GDAS) 0.5° to calculate simple air parcel trajectories, which are defined as the paths of small particles of air at a certain point and time (Stohl et al. 2002; Draxler 2010). A 72-h backward trajectory was computed

as 6-h intervals from 00:00 UTC (Coordinated Universal Time; local time was 16:00, 22:00, 04:00, and 10:00 for Beijing and 15:00, 21:00, 03:00, and 09:00 for Seoul and Nagasaki) for each sampling day with an endpoint height of 1/2 mixing height over the planetary boundary layer. In total, 412, 1072, and 808 backward trajectories were acquired for Beijing, Seoul, and Nagasaki, respectively. Trajectories were statistically analyzed and combined into groups, which are called clusters. Applying the cluster analysis minimizes the trajectory differences within one cluster and maximizes the differences between the clusters (Stein et al. 2015).

Results and discussion

Overview of PM_{2.5} concentrations

A total of 103, 268, and 202 PM_{2.5} samples were collected in Beijing, Seoul, and Nagasaki, respectively. Average mass concentrations of PM_{2.5} and its chemical constituents in the three cities are summarized in Table 1. Figure 1 shows the seasonal averaged mass concentrations of PM_{2.5} in the three cities. The four seasons were divided into 3-month periods to easily compare the seasonal variations of PM_{2.5}. The conversion factor for OC to OM was used as one of the methods for PM_{2.5} mass closure (Srinivas and Sarin 2014). OM is usually estimated by multiplying the mass concentrations of OC by an appropriate conversion factor. The OM to OC conversion factors ranged from 1.1–3.7 in previous studies (Perrone et al. 2012; Xing et al. 2013; Srinivas and Sarin 2014 and the references therein). A conversion factor of 1.6 is usually used for rural areas. Persistent burning of lump coal and a high contribution of biomass burning in China led to high OC. More oxygenated OC resulted in a higher OM/OC ratio; thus, a factor of 1.7 was applied in this study (Kim et al. 2006; Xing et al. 2013). OM was the major contributor to PM_{2.5} with seasonally averaged values in the range of 19.4–63.8% across the sampling sites. Trace elements, which were analyzed by XRF, were classified as crustal (Al, Ca, Fe, K, Mn, Si, and Ti) and non-crustal (Cl, Cr, Cu, Mg, Na, Pb, V, Zn, Br, and Ni) by their origin, and a dust oxide model was applied for crustal species using the following equation (Gu et al. 2010).

$$[\text{Crustal}] = 1.889 \times [\text{Al}] + 1.4 \times [\text{Ca}] + 1.43 \times [\text{Fe}] + 1.205 \times [\text{K}] + 1.582 \times [\text{Mn}] + 2.139 \times [\text{Si}] + 1.668 \times [\text{Ti}]$$

The average PM_{2.5} mass concentration during the sampling period was $125 \pm 6.80 \mu\text{g m}^{-3}$, with a range of 7.1 to $743.8 \mu\text{g m}^{-3}$ in Beijing. The PM_{2.5} mass concentration level was 1.67 times higher than the class 2 standard for PM_{2.5} in China in 2012 (GB3095-2012, $75 \mu\text{g m}^{-3}$), which is applied to all areas of China, except for special regions, such as national parks. The seasonal change in PM_{2.5} were observed as: fall ($163 \pm 39.2 \mu\text{g m}^{-3}$) > spring ($125 \pm 34.4 \mu\text{g m}^{-3}$) > winter ($110 \pm 13.8 \mu\text{g m}^{-3}$), which could be explained by haze in

fall and Asian dust in spring. PM_{2.5} levels in fall were 1.48 and 1.30 times higher than those in winter and spring, respectively. Several previous studies have focused on the haze in winter, but the haze occurs frequently in fall (especially in China). Both local and regional transported sources are the major reasons of the haze with the meteorological conditions, such as low wind speed, and high humidity (Hua et al. 2015). Anthropogenic sources caused the haze in urban usually and the dominant constituents of PM_{2.5} during the haze are OM and ionic species (Tan et al. 2009). Even in the winter, with the lowest average concentration amounts, PM_{2.5} mass concentrations were 1.47 times higher than the class 2 standard in Beijing.

PM_{2.5} mass concentrations ranged from 3.8 to $236.2 \mu\text{g m}^{-3}$, with an average of $44.6 \pm 0.84 \mu\text{g m}^{-3}$ during the sampling period in Seoul. The seasonal variations in PM_{2.5} in this study were winter ($54.1 \pm 4.37 \mu\text{g m}^{-3}$) > spring ($45.0 \pm 2.19 \mu\text{g m}^{-3}$) > fall ($37.4 \pm 2.90 \mu\text{g m}^{-3}$) > summer ($32.5 \pm 3.87 \mu\text{g m}^{-3}$) and showed the same seasonal trends that were previously published in Kim et al. (2007). The high concentrations of PM_{2.5} in winter were probably due to the increase in emissions, such as those from heating and coal combustion, and also because of lowered mixing heights (Kim et al. 2007; Son et al. 2012). The high frequency of air masses from the Gobi Desert may be the most likely cause of a higher average PM_{2.5} mass concentration during spring in Seoul.

The average PM_{2.5} mass concentration during the sampling period was $17.4 \pm 0.37 \mu\text{g m}^{-3}$, with a range of 3.4 to $73.3 \mu\text{g m}^{-3}$ in Nagasaki. The seasonal average PM_{2.5} concentrations were $19.5 \pm 1.18 \mu\text{g m}^{-3}$, $16.6 \pm 1.50 \mu\text{g m}^{-3}$, $16.4 \pm 1.85 \mu\text{g m}^{-3}$, and $12.9 \pm 1.14 \mu\text{g m}^{-3}$ in spring, summer, winter, and fall, respectively. High PM_{2.5} concentrations in spring, with high mass concentrations of crustal constituents found in previous studies showed that long-range transport (LRT) is the major source of PM_{2.5} in Japan (Kaneyasu et al. 2014; Wang and Ogawa 2015).

Carbonaceous constituents

The average concentrations of OC and EC in Beijing were $37.9 \pm 3.72 \mu\text{g m}^{-3}$ and $3.21 \pm 0.26 \mu\text{g m}^{-3}$, accounting for 30.4% and 2.6% of the total PM_{2.5} concentration, respectively (Table 1 and Fig. 2). In Beijing, although the average PM_{2.5} concentration was highest in fall, the average OC and EC concentrations were highest in winter (Table S1). The ratios of OC/EC in winter were also high in Beijing, which could be closely related to higher coal consumption for heating in winter (Sun et al. 2004). The average concentrations of OC and EC in Seoul were $6.34 \pm 0.20 \mu\text{g m}^{-3}$ and $1.09 \pm 0.03 \mu\text{g m}^{-3}$, accounting for 14.2% and 2.44% of the total PM_{2.5} concentration, respectively. The average concentrations of OC and EC in Nagasaki were $3.70 \pm 0.14 \mu\text{g m}^{-3}$ and $0.47 \pm 0.02 \mu\text{g m}^{-3}$, accounting for 21.3% and 2.7% of the PM_{2.5}

Table 1 Concentrations of PM_{2.5} and its chemical constituents observed across the sampling sites

Species (average ± S.E.)	Beijing	Seoul	Nagasaki
Measurement periods	Nov. 2014~Apr. 2015	Sep. 2013~May 2015	Feb. 2014~May 2015
Number of samples	103	268	202
PM _{2.5} (μg m ⁻³)	125 ± 6.80	44.6 ± 0.84	17.4 ± 0.37
OM (μg m ⁻³)	64.5 ± 3.15	10.8 ± 0.17	6.29 ± 0.12
EC (μg m ⁻³)	3.21 ± 0.13	1.09 ± 0.02	0.47 ± 0.01
OC/EC	10.6 ± 0.16	6.22 ± 0.08	8.66 ± 0.15
NO ₃ ⁻ (μg m ⁻³)	8.31 ± 0.49	6.92 ± 0.21	1.55 ± 0.06
SO ₄ ²⁻ (μg m ⁻³)	5.11 ± 0.31	6.67 ± 0.19	2.24 ± 0.06
NH ₄ ⁺ (μg m ⁻³)	3.81 ± 0.26	4.21 ± 0.10	0.90 ± 0.03
NO ₃ ⁻ /SO ₄ ²⁻	2.14 ± 0.12	1.23 ± 0.03	1.00 ± 0.06
Cl (μg m ⁻³)	5.02 ± 0.28	0.51 ± 0.02	0.15 ± 0.01
Al (μg m ⁻³)	1.80 ± 0.08	1.13 ± 0.07	0.69 ± 0.04
Ca (μg m ⁻³)	1.68 ± 0.06	0.55 ± 0.03	0.29 ± 0.02
Cr (μg m ⁻³)	0.02 ± 0.00	0.01 ± 0.00	0.01 ± 0.00
Cu (μg m ⁻³)	0.16 ± 0.00	0.06 ± 0.00	0.05 ± 0.00
Fe (μg m ⁻³)	1.64 ± 0.05	0.70 ± 0.02	0.45 ± 0.01
K (μg m ⁻³)	2.34 ± 0.14	0.62 ± 0.02	0.29 ± 0.01
Mg (μg m ⁻³)	0.50 ± 0.03	0.22 ± 0.01	0.19 ± 0.01
Mn (μg m ⁻³)	0.11 ± 0.01	0.04 ± 0.00	0.02 ± 0.00
Na (μg m ⁻³)	1.48 ± 0.07	0.44 ± 0.01	0.60 ± 0.01
Pb (μg m ⁻³)	0.10 ± 0.01	0.02 ± 0.00	0.01 ± 0.00
Si (μg m ⁻³)	6.96 ± 0.29	5.18 ± 0.36	2.97 ± 0.21
Ti (μg m ⁻³)	0.22 ± 0.01	0.12 ± 0.01	0.06 ± 0.00
V (μg m ⁻³)	0.01 ± 0.00	0.01 ± 0.00	0.01 ± 0.00
Zn (μg m ⁻³)	0.41 ± 0.02	0.11 ± 0.00	0.05 ± 0.00
Br (μg m ⁻³)	0.08 ± 0.00	0.03 ± 0.00	0.01 ± 0.00
Ni (μg m ⁻³)	0.26 ± 0.01	0.08 ± 0.00	0.09 ± 0.00

concentrations, respectively. The highest concentrations of OC and EC were found in winter for Beijing and Seoul, but in spring for Nagasaki.

The relationships between OC and EC in the three cities, as demonstrated by linear regressions, are shown in Figure S3. Strong OC and EC correlations were obtained in Beijing, with a coefficient of determination (R^2) of 0.87, 0.98, and 0.89 for spring, fall, and winter, respectively. The correlation between OC and EC concentrations would be strong if the major sources of OC and EC were emissions from primary sources. This suggested that similar emissions and transportation processes affected the correlation of OC and EC (Zhang et al. 2011; Zhang et al. 2017a, b, c and the references there in). Strong OC and EC correlations during fall in Beijing indicated influences from common sources, such as coal combustion, biomass burning, and mobile sources (Cao et al. 2005; Zhang et al. 2011). OC and EC concentrations were less strongly correlated in Seoul ($R^2 = 0.14$ – 0.62) and Nagasaki ($R^2 = 0.11$ – 0.63) than in Beijing. In order to identify the sources of carbon species, the OC/EC ratio was also used (Kong et al. 2010). In Beijing, the average OC/EC ratio in spring

(11.0 ± 0.71) was higher than in other seasons. The highest average OC/EC ratios were found in winter in Seoul (7.45 ± 0.38) and in fall in Nagasaki (10.1 ± 0.54). The ranges of OC/EC ratios were 2.70–18.0 (average 10.6) for Beijing, 1.41–18.4 (average 6.23) for Seoul, and 2.14–29.3 (average 8.65) for Nagasaki. The significantly high OC/EC ratios (OC/EC > 2.0) indicated that secondary organic carbon (SOC) might have formed (Zhao et al. 2015; Zhang et al. 2017a, b, c and the references there in).

The concentrations of SOC could be calculated using EC as a tracer of primary emission sources because EC originates only from primary emissions and resists chemical reactions (Kim et al. 2007; Lin et al. 2009). Two equations were applied to calculate SOC (Lim and Turpin 2002).

$$\begin{aligned} \text{OC}_{\text{pri}} &= (\text{OC}/\text{EC})_{\text{pri}} \times \text{EC} + Z \\ \text{OC}_{\text{sec}} &= \text{OC}_{\text{tot}} - \text{OC}_{\text{pri}} \end{aligned}$$

where OC_{pri} is the primary OC (POC), $(\text{OC}/\text{EC})_{\text{pri}}$ is an estimated value of the primary OC/EC ratio, OC_{sec} is the SOC, and OC_{tot} is the total mass concentration of OC. POC from

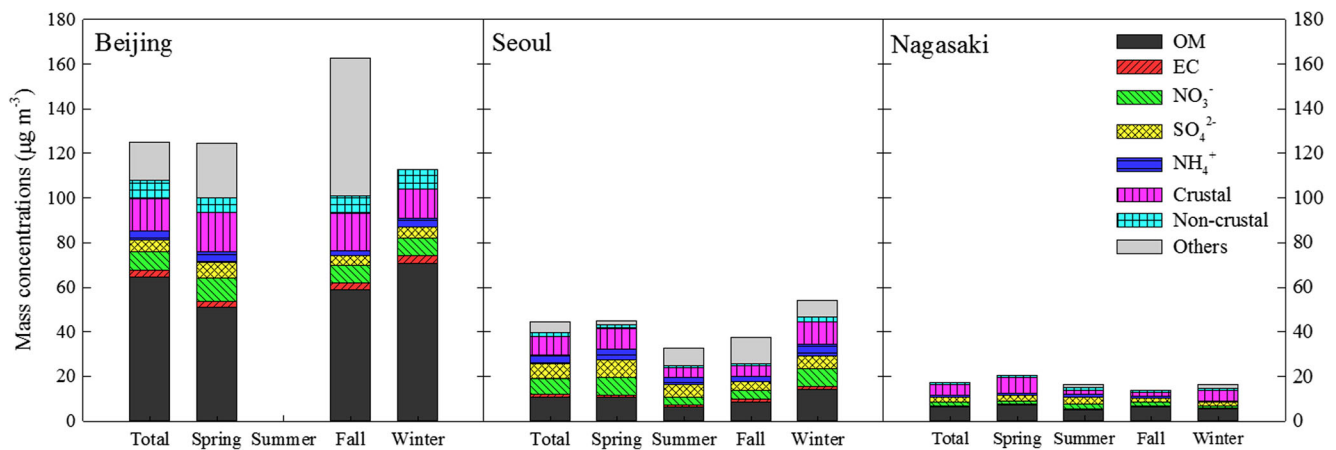


Fig. 1 Seasonal $PM_{2.5}$ mass concentrations and its chemical constituents (Beijing: Nov. 2014–Apr. 2015, Seoul: Sep. 2013–May 2015, and Nagasaki: Feb. 2014–May 2015)

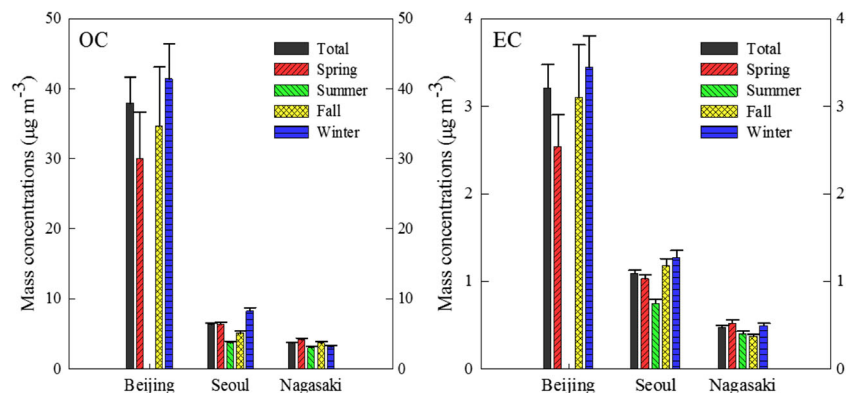
non-combustion sources is represented as Z in the equation (Lim and Turpin 2002; Snyder et al. 2009). To calculate $(OC/EC)_{pri}$ and POC from non-combustion sources, a Deming regression was used (Saylor et al. 2006; Lin et al. 2009).

The calculated $(OC/EC)_{pri}$ values in Beijing were 7.41, 7.04, and 7.05 for spring, fall, and winter, respectively. The values were 2.76, 4.31, 3.02, and 3.24 for Seoul in spring, summer, fall, and winter. In the same order, the values were 3.45, 3.51, 7.46, and 4.81 for Nagasaki. According to the previous studies (Cao et al. 2005 and the references therein), OC/EC ratios are 1.1, 2.7, and 9.0 for motor vehicles, coal combustion, and biomass burning, respectively. Higher $(OC/EC)_{pri}$ values of three seasons in Beijing and fall in Nagasaki than that of other seasons in Nagasaki and whole seasons in Seoul may indicate the influence of biomass burning. Relatively lower $(OC/EC)_{pri}$ may be resulted from the contribution of motor vehicle and coal combustion.

Seasonal mass concentrations and the ratios of POC and SOC to OC are presented in Table 2. SOC concentrations in Beijing were $10.5 \pm 4.22 \mu\text{g m}^{-3}$, $12.8 \pm 4.28 \mu\text{g m}^{-3}$, and $17.2 \pm 2.69 \mu\text{g m}^{-3}$, contributing 35.0%, 37.0%, and 41.4% of OC in spring, fall, and winter, respectively. For Seoul, the

estimated SOC were $2.39 \pm 0.20 \mu\text{g m}^{-3}$, $2.15 \pm 0.23 \mu\text{g m}^{-3}$, $1.53 \pm 0.19 \mu\text{g m}^{-3}$, and $4.13 \pm 0.45 \mu\text{g m}^{-3}$, contributing 37.7%, 57.9%, 30.2%, and 50.1% of OC in spring, summer, fall, and winter, respectively. Estimated SOC concentrations in Nagasaki were $2.24 \pm 0.17 \mu\text{g m}^{-3}$, $1.49 \pm 0.21 \mu\text{g m}^{-3}$, $0.90 \pm 0.19 \mu\text{g m}^{-3}$, and $0.76 \pm 0.16 \mu\text{g m}^{-3}$, contributing 53.7%, 49.4%, 24.5%, and 24.4% of OC during spring, summer, fall, and winter, respectively. OC was dominated by POC in Beijing while POC was comparable with SOC in Seoul and Nagasaki. Calculated ratios of SOC/OC in Beijing showed slightly lower than those in Xi'an (33.4%) (Zhang et al. 2015) and higher than Lin'an (28.1%) (Zhang et al. 2017a, b, c) and Beijing (16.2–32.3%) (Zhao et al. 2013). Compared with the results of the estimated SOC in other cities in South Korea, the estimated SOC in this study was lower than those in Incheon (58.2%) (Choi et al. 2013) and Chuncheon (Han et al. 2011) (35–63%). Higher mass concentrations of SOC and the portion to OC were shown during winter in Beijing. For Seoul, mass concentration of SOC was higher during winter while the percentage of SOC to OC was higher during summer. As high temperatures and sunlight are favorable for SOC formation, SOC usually has a higher percentage in

Fig. 2 Seasonal mass concentrations of OC and EC in $PM_{2.5}$ (Beijing: Nov. 2014–Apr. 2015, Seoul: Sep. 2013–May 2015, and Nagasaki: Feb. 2014–May 2015)



summer. However, stagnant air with low temperature favored the partitioning of semi-volatile organic compounds (SVOC) (Han et al. 2011; Zhang et al. 2017a, b, c). In contrast, mass concentration and the percentage were both higher during winter in Nagasaki. The higher concentrations of PM_{2.5} with long-range transportation through westerlies and hot and humid climate conditions even in spring may explain the unusual seasonal concentrations of SOC in Nagasaki.

Water-soluble inorganic ions

Secondary pollutants were the major sources of aerosols, as SO₄²⁻, NO₃⁻, and NH₄⁺ accounted for much higher percentages of the total mass concentration. In Beijing, the highest concentrations of NO₃⁻ (10.3 ± 2.80 μg m⁻³), SO₄²⁻ (7.11 ± 2.07 μg m⁻³), and NH₄⁺ (5.07 ± 1.60 μg m⁻³) were found in spring (Fig. 3). Nitrate would be favor to form with cooler temperatures if ammonium is sufficiently present in the atmosphere. For the reason, many previous studies reported that NO₃⁻ concentration is higher during winter than summer with sufficient NH₄⁺. Seasonal mass concentrations of NH₄⁺ were also high during winter and spring (4.87 ± 0.42 μg m⁻³ and 4.71 ± 0.31 μg m⁻³, respectively), similar to results from previous studies (Seinfeld and Pandis 1998; Kim et al. 2007). Average mass concentrations of NO₃⁻ in Beijing were approximately 20% higher than those in Seoul, and the mean SO₄²⁻ concentrations were lower than those in Seoul. The lower mass concentrations of SO₄²⁻ in Beijing were attributed to the absence of summer during the study period.

The seasonal average concentrations of SO₄²⁻ (7.82 ± 0.58 μg m⁻³ for spring, 5.88 ± 0.66 μg m⁻³ for winter, 5.51 ± 1.60 μg m⁻³ for summer, and 3.94 ± 0.42 μg m⁻³ for fall) were lower than those of NO₃⁻ (8.05 ± 0.85 μg m⁻³ for winter, 7.58 ± 0.60 μg m⁻³ for spring, 3.84 ± 0.63 μg m⁻³ for fall, and 3.67 ± 0.72 μg m⁻³ for summer) in Seoul (Fig. 3). The possible reason for this is that Seoul may receive polluted air, including NO₃⁻, from increased mobile sources transported from China, and it also has a large nitrogen oxide (NO_x) contribution from local mobile sources (He et al. 2003). NO₃⁻ and NH₄⁺ concentrations showed typical seasonal variations, with higher concentrations during winter than that of in summer; this may be because NO_x and NH₃ would be difficult to convert to NO₃⁻ and NH₄⁺, respectively given high temperatures in summer (Zhou et al. 2016; Zhang et al. 2017a, b, c and the references there in).

In Nagasaki, the mass concentrations of the three ion species were high in summer (2.30 ± 0.37 μg m⁻³ for NO₃⁻, 2.83 ± 0.40 μg m⁻³ for SO₄²⁻, and 1.24 ± 0.18 μg m⁻³ for NH₄⁺) (Fig. 3). This result was caused by the higher intensity of sunlight and the high temperatures in summer, which promoted photochemical reactions in the atmosphere (Sun et al. 2004). The formation of SO₄²⁻ and NO₃⁻ is highly dependent on temperature, and usually, SO₄²⁻ shows high concentration

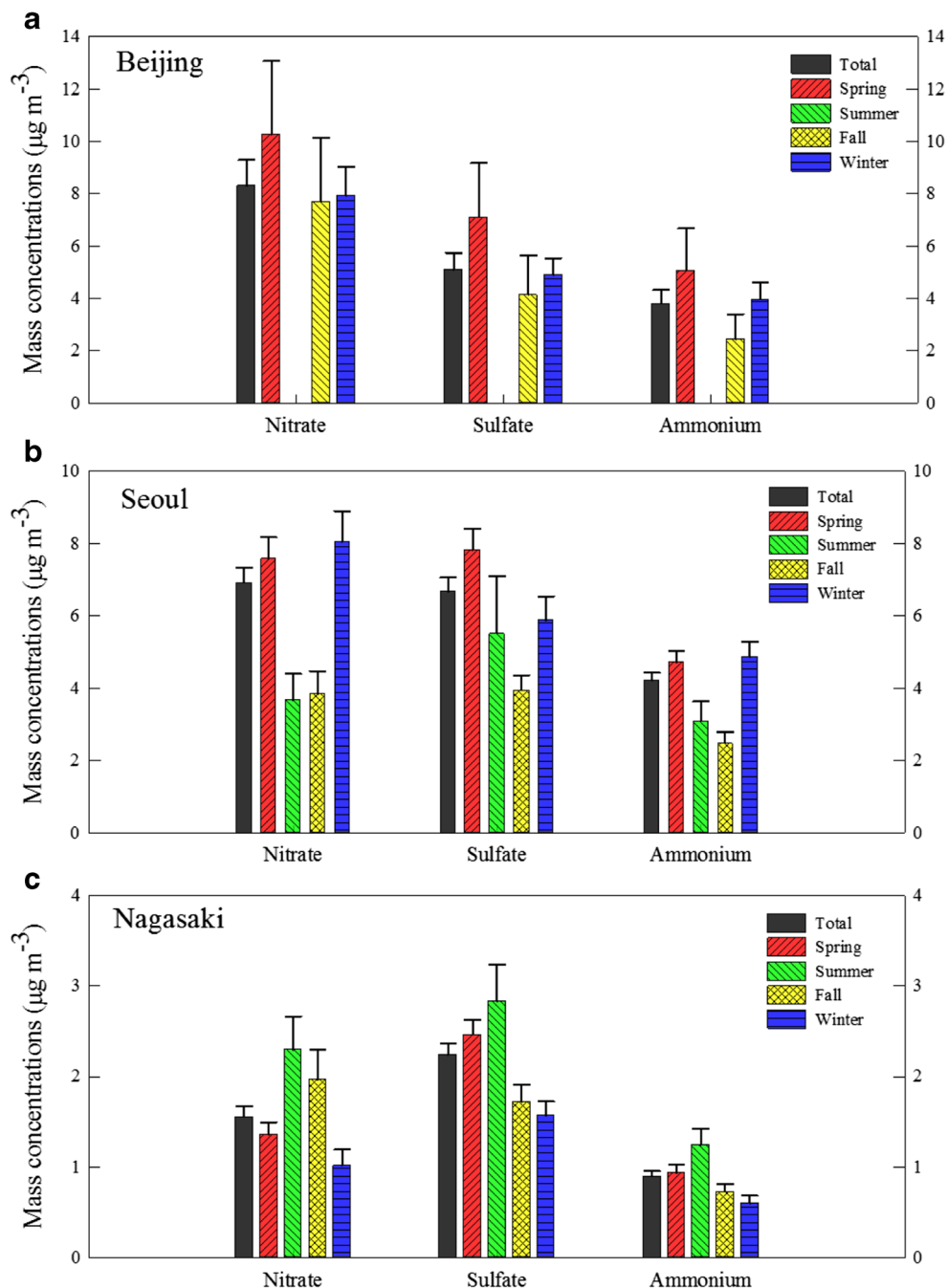
Table 2 Seasonal mass concentrations of POC and SOC and the ratios of POC and SOC to OC

		POC		SOC	
		μg m ⁻³	%	μg m ⁻³	%
Beijing	Spring	19.5 ± 2.69	65.0	10.5 ± 4.22	35.0
	Fall	21.8 ± 4.24	63.0	12.8 ± 4.28	37.0
	Winter	24.3 ± 2.55	58.6	17.2 ± 2.69	41.4
Seoul	Spring	3.94 ± 0.12	62.3	2.39 ± 0.20	37.7
	Summer	1.56 ± 0.10	42.1	2.15 ± 0.23	57.9
	Fall	3.55 ± 0.25	69.8	1.53 ± 0.19	30.2
Nagasaki	Spring	4.12 ± 0.26	49.9	4.13 ± 0.45	50.1
	Summer	1.93 ± 0.13	46.3	2.24 ± 0.17	53.7
	Summer	1.53 ± 0.10	50.6	1.49 ± 0.21	49.4
	Fall	2.77 ± 0.16	75.5	0.90 ± 0.19	24.5
	Winter	2.36 ± 0.13	75.6	0.76 ± 0.16	24.4

at high temperature, whereas NO₃⁻ shows higher concentration at low temperature (Wang et al. 2005). However in Nagasaki, there were no typical trends for all three ion species since the temperature does not drop below freezing even in winter and also a very high temperature and humidity in summer.

The mass ratio of NO₃⁻/SO₄²⁻ has been used to estimate the relative importance of mobile and stationary sources (Kong et al. 2010; Gao et al. 2015; Xu et al. 2016). High NO₃⁻/SO₄²⁻ values implied stronger influences from mobile sources in the case of decreased SO₄²⁻ concentrations, because the ratios of NO_x/SO_x from gasoline and diesel emissions were estimated to be 13:1 and 8:1, respectively (Kong et al. 2010; Xu et al. 2016). The average mass ratios of NO₃⁻/SO₄²⁻ during the study period were 2.14 ± 0.24 (range 0.26–16.0), 1.24 ± 0.07 (range 0.003–8.02), and 1.00 ± 0.12 (range 0.04–13.6) for Beijing, Seoul, and Nagasaki, respectively. The mass ratio of NO₃⁻/SO₄²⁻ in Beijing was two times higher than that in Seoul. This may have been caused by the increase in mobile sources of emissions due to vehicle growth in Beijing, which increased from 2.1 million in 2003 to 5.4 million in 2013 (Gao et al. 2015). The average ratio of NO₃⁻/SO₄²⁻ in Seoul was 1.24 ± 0.07, and the average concentration of NO₃⁻ was 24% higher than that of SO₄²⁻. The higher proportion of NO₃⁻ in Seoul may be due to the production of a large amount of NO_x from high population density and traffic volume. The mass ratio of NO₃⁻/SO₄²⁻ was 1:1 in Nagasaki, which was relatively small compared to that in Beijing. Unlike Beijing and Seoul, which have high rates of motor vehicle and public transportation use, trams are very common along the major roads in Nagasaki, which results in less contribution from mobile sources. In addition, the lack of direct impacts from local industrial sources may imply that the major emission sources that affect NO₃⁻ and SO₄²⁻ are from other cities

Fig. 3 Seasonal mass concentrations of ion species in PM_{2.5} (Beijing: Nov. 2014–Apr. 2015, Seoul: Sep. 2013–May 2015, and Nagasaki: Feb. 2014–May 2015)



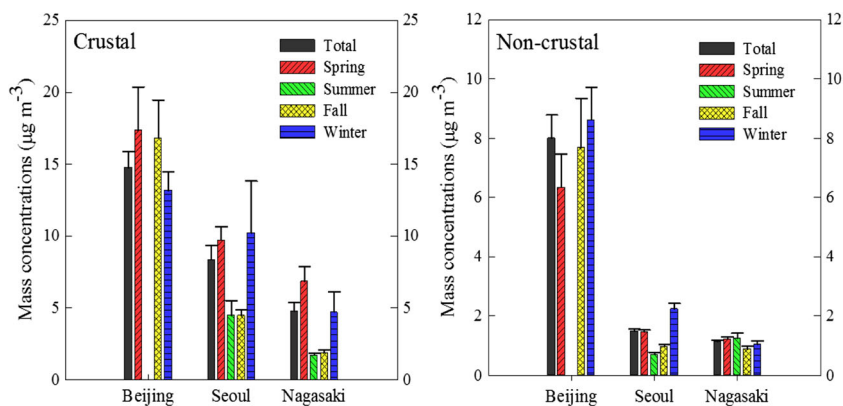
or countries through long-range transportation. Through long-range transportation overseas, HNO₃ is usually neutralized with cation compounds, such as Ca, which leads to a larger contribution of H₂SO₄ formed by the oxidation of SO₂. The descending order of NO₃⁻/SO₄²⁻ ratios in Beijing, Seoul, and Nagasaki may confirm the effect of long-range transportation.

Trace elements

As mentioned in the “[Overview of PM_{2.5} concentrations](#)” section, trace elements were grouped as crustal and non-crustal

and their seasonal mass concentrations were shown in Fig. 4. The average concentrations of crustal and non-crustal species in Beijing were $14.8 \pm 1.12 \mu\text{g m}^{-3}$ and $8.01 \pm 0.78 \mu\text{g m}^{-3}$, accounting for 11.8% and 6.40% of the total PM_{2.5} concentration, respectively. A higher concentration of crustal species was shown in spring ($17.4 \pm 3.02 \mu\text{g m}^{-3}$, accounting for 13.9% of total PM_{2.5}). The non-crustal species concentration in winter was considerably higher ($8.63 \pm 1.10 \mu\text{g m}^{-3}$) accounting for 7.82% of PM_{2.5} in Beijing. In Seoul, the average concentrations of crustal and non-crustal species were $8.36 \pm 1.00 \mu\text{g m}^{-3}$ and $1.49 \pm 0.06 \mu\text{g m}^{-3}$, accounting for 18.7%

Fig. 4 Seasonal mass concentrations of crustal and non-crustal species in PM_{2.5} (Beijing: Nov. 2014–Apr. 2015, Seoul: Sep. 2013–May 2015, and Nagasaki: Feb. 2014–May 2015)



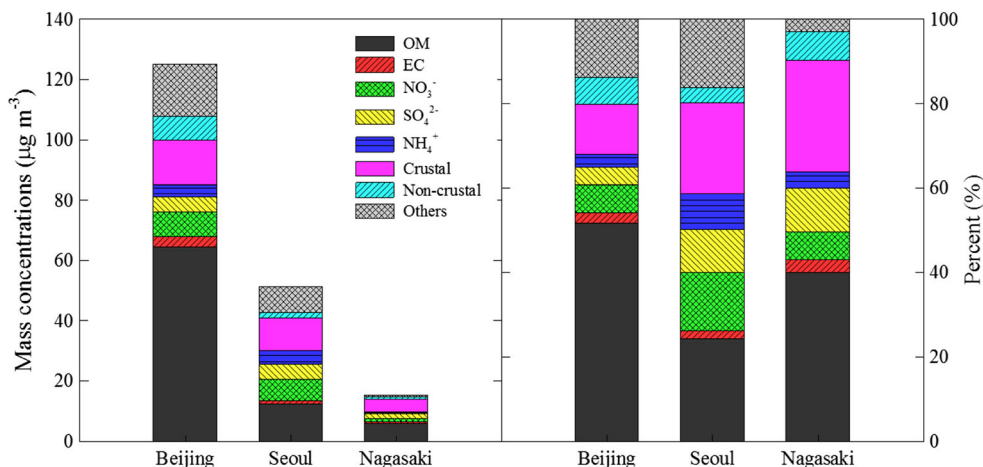
and 3.33% of the total PM_{2.5} concentration, respectively. Seasonal variations in crustal and non-crustal species were higher in winter ($10.2 \pm 3.60 \mu\text{g m}^{-3}$ and $2.25 \pm 0.17 \mu\text{g m}^{-3}$) and spring ($9.72 \pm 0.92 \mu\text{g m}^{-3}$ and $1.45 \pm 0.07 \mu\text{g m}^{-3}$) in Seoul. Concentrations of crustal and non-crustal species in Nagasaki were $4.77 \pm 0.58 \mu\text{g m}^{-3}$ and $1.14 \pm 0.05 \mu\text{g m}^{-3}$, respectively, during the study period. In Nagasaki, higher seasonal average concentrations of crustal species were found during spring and winter ($6.84 \pm 1.03 \mu\text{g m}^{-3}$ and $4.68 \pm 1.44 \mu\text{g m}^{-3}$), which was similar to Beijing and Seoul, while average concentrations of non-crustal species were higher during summer and spring ($1.25 \pm 0.17 \mu\text{g m}^{-3}$ and $1.22 \pm 0.07 \mu\text{g m}^{-3}$). Asian dust was identified as a major contributor of PM_{2.5} and its species, such as crustal species, in many previous studies (Kim et al. 2007; Han et al. 2011 and the references there in). In Korea and Japan, over 60% and 80% of total Asian dust events issued by the Korean Meteorological Agency and Japan Meteorological Agency, respectively, occurred during the spring season of this study period. Therefore, the highest seasonal mass fractions of crustal species to PM_{2.5} were shown during spring in the three cities, which accounted for 13.9%, 21.6%, and 35.0% in Beijing, Seoul, and Nagasaki, respectively.

Comparisons of PM_{2.5} between Beijing, Seoul, and Nagasaki

As mentioned above (“Sampling” section), the sampling periods of Beijing, Seoul, and Nagasaki were different. To compare the results of PM_{2.5} measured in the three cities on equal terms, results from the same time period (November 2014 to April 2015) were comparatively analyzed (Fig. 5). Despite comparisons among the same time periods, the chemical compositions of PM_{2.5} were different among the three cities. In addition, different sources and the regions were affected by long-range transport (e.g., Seoul by northeast China, and Nagasaki by from eastern coastal regions in China through southern parts of South Korea).

During the period, a total of 104, 109, and 88 PM_{2.5} samples were measured and the average mass concentrations of PM_{2.5} were $125 \pm 13.6 \mu\text{g m}^{-3}$, $51.0 \pm 3.07 \mu\text{g m}^{-3}$, and $15.3 \pm 1.01 \mu\text{g m}^{-3}$ in Beijing, Seoul, and Nagasaki, respectively (For Beijing, the results of mass concentrations and the percentage of chemical constituents to PM_{2.5} were the same as the results described in the “Overview of PM_{2.5} concentrations” section). Average mass concentrations of

Fig. 5 Mass concentrations of chemical constituents of PM_{2.5} and its percentage to the PM_{2.5} in three cities during Nov. 2014 to Apr. 2015



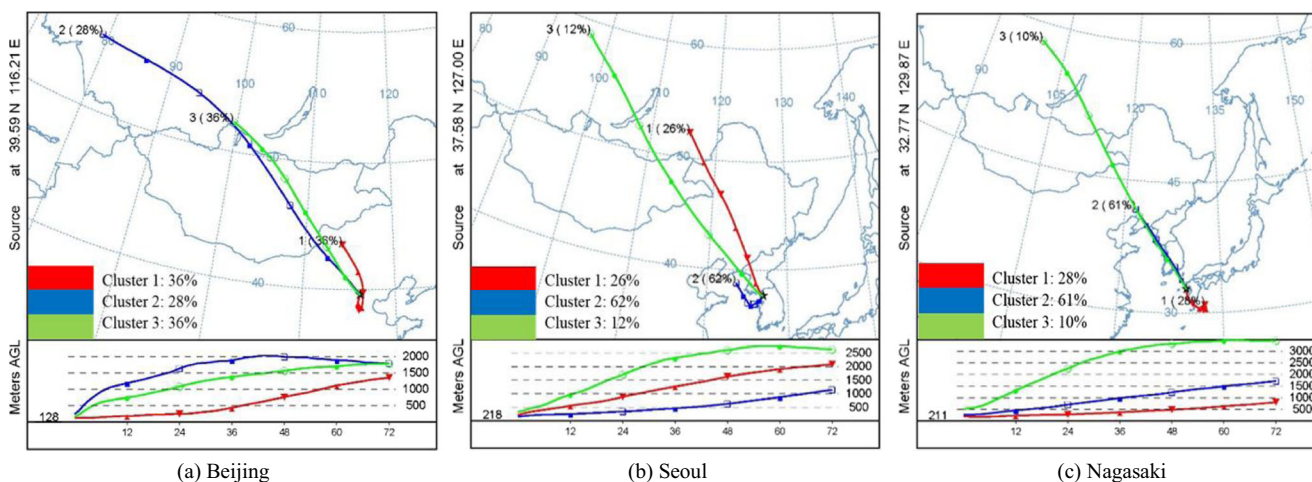


Fig. 6 Mean backward trajectory clusters of **a** Beijing, **b** Seoul, and **c** Nagasaki (72-h air parcel backward trajectories every 6 h in each day)

PM_{2.5} measured in Beijing were 2.5 and 8.2 times higher than those in Seoul and Nagasaki. OM was the highest constituent in Beijing (64.5 $\mu\text{g m}^{-3}$, 51.6% to PM_{2.5}) and Nagasaki (6.29 $\mu\text{g m}^{-3}$, 39.9% to PM_{2.5}), while ionic species were higher (17.8 $\mu\text{g m}^{-3}$, 32.4% to PM_{2.5}) in Seoul. Among the ionic species, NO₃⁻ was higher in Beijing and Seoul (8.31 and 6.97 $\mu\text{g m}^{-3}$, respectively), while SO₄²⁻ was higher in Nagasaki (1.58 $\mu\text{g m}^{-3}$). PM_{2.5} measured in Seoul showed higher fraction of crustal species (21.5%) than the total period of the study (18.7%) because the period includes spring (except May). For Nagasaki, crustal constituents accounted for 26.4% during the period and 27.4% during total study period.

A total 32 samples were collected on the same day in three cities during the study period. They were divided into three groups (Nov–Dec, Jan–Feb, and Mar–Apr) and the chemical compositions of PM_{2.5} were presented in Figure S4. OM was the major component of PM_{2.5} and EC was higher in order to Mar–Apr > Jan–Feb > Nov–Dec in all three cities. NO₃⁻ was the highest component among the ion species in Beijing and Seoul. In Nagasaki, NO₃⁻ was higher during Nov–Dec, while SO₄²⁻ was higher from January. Crustal species was high in order to Nagasaki > Seoul > Beijing during Mar–Apr.

Backward trajectory cluster analysis

Three groups of backward trajectories were obtained for each city based on their origins, and transportation paths of each group were expressed by one cluster mean backward trajectory (Fig. 6). The mass concentrations and the ratios of PM_{2.5} mass concentrations which are on average explained by the species for each cluster are given in Figures S5–S7.

For Beijing, cluster 1 had higher PM_{2.5} mass concentrations (194 ± 14.5 $\mu\text{g m}^{-3}$) than cluster 2 (72.0 ± 7.23 $\mu\text{g m}^{-3}$) and cluster 3 (98.0 ± 8.03 $\mu\text{g m}^{-3}$) (Fig. 6a and Table S2). Cluster 1, which was the shortest, showed a typical local transportation

pattern and accounted for 36% of total trajectories. Cluster 1 for Beijing had high concentrations of OM (90.5 ± 6.72 $\mu\text{g m}^{-3}$, accounting for 46.7%) and ion species (28.8 ± 2.23 $\mu\text{g m}^{-3}$, accounting for 14.8%). The local cluster for Beijing spent much more time passing over the Bohai economic zone, which is a highly industrialized and densely populated zone in China (Zhang et al. 2009; Gao et al. 2011). The higher mass concentrations of PM_{2.5} and its constituents for cluster 1 seems to be due to accumulated primary particles generated from industrial areas (Cao et al. 2011; Zhang et al. 2017a, b, c). Cluster 2 (28%) originated in Russia and moved quickly through industrial cities in Siberia, Mongolia, and Inner Mongolia before arriving in Beijing. Cluster 3, which occupied 36% of the total backward trajectories, originated near the border between Russia and Mongolia and moved northwest to Beijing through Mongolia and Inner Mongolia. Clusters 2 and 3 were northwest clusters representing long-range transportation, and they had higher mass ratios of crustal species to PM_{2.5} (17.2% and 11.7% for cluster 2 and cluster 3, respectively) than cluster 1 (10.2%).

Figure 6b shows the cluster mean trajectory plot of Seoul. Cluster 1 (26%) originated in the southeastern area of Russia and crossed through North China and North Korea to Seoul. Low concentrations of ion species and crustal species occurred in cluster 1, which was transported over a long range but did not cross Mongolia or the highly industrialized zone. The shortest and most frequent cluster for Seoul (cluster 2) accounted for 62%. Cluster 2 originated from the Yellow Sea near Dalian, which is a port city of Liaoning province with a well-known, large industrial area. The air masses moved to the southwest part of Seoul, which includes Ansan, Dangiin, and Taeon and contains industrial complexes. Long residence times over industrial zones with high emissions from point sources caused the high concentrations of PM_{2.5}, OM, and ion species. High concentrations of dominant ion components

(NO₃⁻, SO₄²⁻, and NH₄⁺) were associated with the local cluster of Seoul (43.5%) (He et al. 2003). Cluster 3 originated in Russia and passed through Mongolia; it was transported over a long distance and was associated with high concentrations of crustal species (33.0%).

Figure 6c shows the three cluster mean trajectories from local (cluster 1, accounting for 28%) and long-range transportation, with clusters 2 and 3 accounting for 61% and 10%, respectively. The local air masses (cluster 1), originating from the Pacific Ocean near Miyazaki, crossed to Kagoshima in Kyushu. The total mass ratio of ion species was high at 32.6% compared to the average PM_{2.5} mass concentration (14.0 ± 0.46 μg m⁻³) for the local cluster. In a previous study (Kaneyasu et al. 2014), NO₃⁻ in Kyushu was considered to originate from locally emitted air pollutants rather than from those transported over a long range. A similar result was shown in the present study, with the highest concentrations of NO₃⁻ (1.92 ± 0.12 μg m⁻³) in cluster 1. Cluster 2, which originated in the northern part of China near Beijing and passed through North Korea and South Korea to Nagasaki, had the highest concentrations of PM_{2.5}, OM, and SO₄²⁻. PM_{2.5}, particularly SO₄²⁻ (2.51 ± 0.08 μg m⁻³) and OM (6.90 ± 0.17 μg m⁻³), were largely dominated by long-range transported aerosols (Kaneyasu et al. 2014). In addition to SO₄²⁻, cluster 2 also had high NO₃⁻ and NH₄⁺ concentrations because it originated in and crossed through highly industrialized zones in China and South Korea. Cluster 3 originated in Russia, moved northwest through Mongolia, and quickly moved through Inner Mongolia. The directions of clusters 2 and 3 for Nagasaki were almost the same, but because of their origins and residence times in industrialized areas, PM_{2.5} concentrations were much higher in cluster 2 (19.5 ± 0.53 μg m⁻³) than in cluster 3 (14.0 ± 0.98 μg m⁻³). Results of seasonal cluster analysis and that of PM_{2.5} chemical constituents for each clusters in three cities were added in Figure S8–S10.

Conclusion

The characteristics of PM_{2.5} and its chemical constituents in Beijing, Seoul, and Nagasaki were identified in this study. The average PM_{2.5} concentration was 125 ± 6.80 μg m⁻³, and the highest seasonal concentration was found in fall (163 ± 39.2 μg m⁻³). In Seoul, the average concentration of PM_{2.5} was 44.6 ± 0.84 μg m⁻³, and higher and lower concentrations were found in winter (54.1 ± 4.37 μg m⁻³) and summer (32.5 ± 3.87 μg m⁻³), respectively. The average PM_{2.5} concentration in Nagasaki was 17.4 ± 0.37 μg m⁻³, which was 7.2 and 2.6 times lower than that in Beijing and Seoul, respectively. High PM_{2.5} concentrations in Nagasaki were found during spring, with high concentrations of crustal species owing to the effect of long-range transportation, especially on Asian dust. The average ratios of carbonaceous species to PM_{2.5}

were 32.9%, 16.6%, and 24.0% in Beijing, Seoul, and Nagasaki, respectively. OC and EC were weakly correlated in Seoul ($R^2 = 0.14–0.62$) and Nagasaki ($R^2 = 0.11–0.63$), while they were strongly correlated in Beijing ($R^2 = 0.87–0.98$). The average NO₃⁻ concentration was higher than that of SO₄²⁻ in Beijing, owing to the rapid increase in mobile emissions. Similar to Beijing, the NO₃⁻ concentration was slightly higher than that of SO₄²⁻ in Seoul and Nagasaki. High NO₃⁻/SO₄²⁻ values, which were found in Beijing and Seoul, implied strong effects from mobile sources. Identification of three transportation pathways of air masses were conducted for each city. The shortest cluster (36%) in Beijing which represents local emissions showed the highest concentrations of PM_{2.5}. The most frequent cluster for Seoul, which originated at the Yellow Sea near one of the port cities in Liaoning Province and passed through industrial complexes located in the southwest of Seoul, had high PM_{2.5} concentrations. A cluster that originated near a highly industrialized zone in the northern part of China and passed through North Korea and South Korea to Nagasaki had the highest PM_{2.5}, OM, and SO₄²⁻ concentrations. These results verified that local emissions and long-range transported emissions should be considered mutually in order to control PM_{2.5} in Northeast Asia. The findings of this study signify that long-range transportation sources and local sources of PM_{2.5} should be considered simultaneously to implement efficient mitigation plans of PM_{2.5} in East Asia.

Funding information This work was supported by the Global Research Lab (#K21004000001-10A0500-00710) and the Basic Science Research Program (2014R1A2A2A04007801) through the National Research Foundation of Korea (NRF) which is funded by the Ministry of Science, ICT (Information and Communication Technologies).

References

- Anenberg SC, West JJ, Yu H, Chin M, Schulz M, Bergmann D, Bey I, Bian H, Diehl T, Fiore A, Hess P, Marmer E, Montanaro V, Park R, Shindell D, Takemura T, Dentener F (2014) Impacts of intercontinental transport of anthropogenic fine particulate matter on human mortality. *Air Qual Atmos Health* 7:369–379. <https://doi.org/10.1007/s11869-014-0248-9>
- Cao J, Wu F, Chow J, Lee S, Li Y, Chen S, An Z, Fung K, Watson J, Zhu C (2005) Characterization and source apportionment of atmospheric organic and elemental carbon during fall and winter of 2003 in Xi'an, China. *Atmos Chem Phys* 5: 3127–3137
- Cao G, Zhang X, Gong S, An X, Wang Y (2011) Emission inventories of primary particles and pollutant gases for China. *Chin Sci Bull* 56: 781–788. <https://doi.org/10.1007/s11434-011-4373-7>
- Chan CK, Yao X (2008) Air pollution in mega cities in China. *Atmospheric Environment* 42:1–42. DOI: <https://doi.org/10.1016/j.atmosenv.2007.09.003>
- Choi Jk, Heo JB, Ban SJ, Yi SM, Zoh KD (2013) Source apportionment of PM_{2.5} at the coastal area in Korea. *Sci Total Environ* 447:370–380

- Dockery DW, Pope CA, Xu X, Spengler JD, Ware JH, Fay ME, Ferris BGJ, Speizer FE (1993) An association between air pollution and mortality in six U.S. cities. *N Engl J Med* 329:1753–1759. <https://doi.org/10.1056/nejm199312093292401>
- Draxler R (2010) HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) model access via NOAA ARL READY website. <http://ready.arl.noaa.gov/HYSPLIT.php>
- Gao X, Yang L, Cheng S, Gao R, Zhou Y, Xue L, Shou Y, Wang J, Wang X, Nie W, Xu P, Wang W (2011) Semi-continuous measurement of water-soluble ions in PM_{2.5} in Jinan, China: temporal variations and source apportionments. *Atmos Environ* 45:6048–6056. <https://doi.org/10.1016/j.atmosenv.2011.07.041>
- Gao J, Tian H, Cheng K, Lu L, Zheng M, Wang S, Hao J, Wang K, Hua S, Zhu C (2015) The variation of chemical characteristics of PM_{2.5} and PM₁₀ and formation causes during two haze pollution events in urban Beijing, China. *Atmos Environ* 107:1–8
- Gu J, Bai Z, Liu A, Wu L, Xie Y, Li W, Dong H, Zhang X (2010) Characterization of atmospheric organic carbon and element carbon of PM_{2.5} and PM₁₀ at Tianjin, China. *Aerosol Air Qual Res* 10:167–176. <https://doi.org/10.4209/aaqr.2009.12.0080>
- Han YJ, Kim SR, Jung JH (2011) Long-term measurements of atmospheric PM_{2.5} and its chemical composition in rural Korea. *J Atmos Chem* 68:281–298. <https://doi.org/10.1007/s10874-012-9225-6>
- Hayakawa K (2016) Environmental behaviors and toxicities of polycyclic aromatic hydrocarbons and nitropolycyclic aromatic hydrocarbons. *Chem Pharm Bull* 64:83–94
- He Z, Kim YJ, Ogunjobi KO, Hong CS (2003) Characteristics of PM_{2.5} species and long-range transport of air masses at Taean background station, South Korea. *Atmos Environ* 37:219–230. [https://doi.org/10.1016/S1352-2310\(02\)00834-8](https://doi.org/10.1016/S1352-2310(02)00834-8)
- Hua Y, Cheng Z, Wang S, Jiang J, Chen D, Cai S, Fu X, Fu Q, Chen C, Xu B (2015) Characteristics and source apportionment of PM_{2.5} during a fall heavy haze episode in the Yangtze River Delta of China. *Atmos Environ* 123:380–391
- Jimenez JL, Canagaratna M, Donahue N, Prevot A, Zhang Q, Kroll JH, DeCarlo PF, Allan JD, Coe H, Ng N (2009) Evolution of organic aerosols in the atmosphere. *Science* 326:1525–1529
- Kaneyasu N, Yamamoto S, Sato K, Takami A, Hayashi M, Hara K, Kawamoto K, Okuda T, Hatakeyama S (2014) Impact of long-range transport of aerosols on the PM_{2.5} composition at a major metropolitan area in the northern Kyushu area of Japan. *Atmos Environ* 97:416–425. <https://doi.org/10.1016/j.atmosenv.2014.01.029>
- Kim YJ, Kim KW, Kim SD, Lee BK, Han JS (2006) Fine particulate matter characteristics and its impact on visibility impairment at two urban sites in Korea: Seoul and Incheon. *Atmos Environ* 40(Supplement 2):593–605. <https://doi.org/10.1016/j.atmosenv.2005.11.076>
- Kim HS, Huh JB, Hopke PK, Holsen TM, Yi SM (2007) Characteristics of the major chemical constituents of PM_{2.5} and smog events in Seoul, Korea in 2003 and 2004. *Atmos Environ* 41:6762–6770
- Kong S, Han B, Bai Z, Chen L, Shi J, Xu Z (2010) Receptor modeling of PM_{2.5}, PM₁₀ and TSP in different seasons and long-range transport analysis at a coastal site of Tianjin, China. *Sci Total Environ* 408:4681–4694. <https://doi.org/10.1016/j.scitotenv.2010.06.005>
- Lim HJ, Turpin BJ (2002) Origins of primary and secondary organic aerosol in Atlanta: results of time-resolved measurements during the Atlanta supersite experiment. *Environ Sci Technol* 36:4489–4496. <https://doi.org/10.1021/es0206487>
- Lin P, Hu M, Deng Z, Slanina J, Han S, Kondo Y, Takegawa N, Miyazaki Y, Zhao Y, Sugimoto N (2009) Seasonal and diurnal variations of organic carbon in PM_{2.5} in Beijing and the estimation of secondary organic carbon. *J Geophys Res-Atmos* 114:n/a-n/a. DOI: <https://doi.org/10.1029/2008JD010902>
- Perrone MG, Larsen BR, Ferrero L, Sangiorgi G, De Gennaro G, Udisti R, Zangrando R, Gambaro A, Bolzacchini E (2012) Sources of high PM_{2.5} concentrations in Milan, Northern Italy: molecular marker data and CMB modelling. *Sci Total Environ* 414:343–355. <https://doi.org/10.1016/j.scitotenv.2011.11.026>
- Saylor RD, Edgerton ES, Hartsell BE (2006) Linear regression techniques for use in the EC tracer method of secondary organic aerosol estimation. *Atmos Environ* 40:7546–7556. <https://doi.org/10.1016/j.atmosenv.2006.07.018>
- Schwartz J, Slater D, Larson TV, Pierson WE, Koehn JQ (1993) Particulate air pollution and hospital emergency room visits for asthma in Seattle. *Am Rev Respir Dis* 147:826–831
- Seinfeld J, Pandis S (1998) Atmospheric physics and chemistry. From air pollution to climate change, John Wiley & Sons
- Shimada K, Takami A, Kato S, Kajii Y, Hasegawa S, Fushimi A, Shimizu A, Sugimoto N, Chan C, Kim Y (2016) Characteristics of carbonaceous aerosols in large-scale Asian wintertime outflows at Cape Hedo, Okinawa, Japan. *J Aerosol Sci* 100:97–107
- Snyder DC, Rutter AP, Collins R, Worley C, Schauer JJ (2009) Insights into the origin of water soluble organic carbon in atmospheric fine particulate matter. *Aerosol Sci Technol* 43:1099–1107
- Son JY, Lee JT, Kim KH, Jung K, Bell ML (2012) Characterization of fine particulate matter and associations between particulate chemical constituents and mortality in Seoul, Korea. *Environ Health Perspect* 120:872–878. <https://doi.org/10.1289/ehp.1104316>
- Srinivas B, Sarin MM (2014) PM_{2.5}, EC and OC in atmospheric outflow from the Indo-Gangetic Plain: temporal variability and aerosol organic carbon-to-organic mass conversion factor. *Sci Total Environ* 487:196–205. <https://doi.org/10.1016/j.scitotenv.2014.04.002>
- Stein A, Draxler RR, Rolph GD, Stunder BJ, Cohen M, Ngan F (2015) NOAA's HYSPLIT atmospheric transport and dispersion modeling system. *Bull Am Meteorol Soc* 96:2059–2077
- Stohl A, Eckhardt S, Forster C, James P, Spichtinger N, Seibert P (2002) A replacement for simple back trajectory calculations in the interpretation of atmospheric trace substance measurements. *Atmos Environ* 36:4635–4648
- Sun Y, Zhuang G, Wang Y, Han L, Guo J, Dan M, Zhang W, Wang Z, Hao Z (2004) The air-borne particulate pollution in Beijing—concentration, composition, distribution and sources. *Atmos Environ* 38:5991–6004. <https://doi.org/10.1016/j.atmosenv.2004.07.009>
- Tan J, Duan J, He K, Ma Y, Duan F, Chen Y, Fu J (2009) Chemical characteristics of PM_{2.5} during a typical haze episode in Guangzhou. *J Environ Sci* 21:774–781
- Wang J, Ogawa S (2015) Effects of meteorological conditions on PM_{2.5} concentrations in Nagasaki, Japan. *Int J Environ Res Public Health* 12:9089–9101. <https://doi.org/10.3390/ijerph120809089>
- Wang Y, Zhuang G, Tang A, Yuan H, Sun Y, Chen S, Zheng A (2005) The ion chemistry and the source of PM_{2.5} aerosol in Beijing. *Atmos Environ* 39:3771–3784
- Wang H, Xu J, Zhang M, Yang Y, Shen X, Wang Y, Chen D, Guo J (2014) A study of the meteorological causes of a prolonged and severe haze episode in January 2013 over central-eastern China. *Atmos Environ* 98:146–157
- Winberry W Jr, Ellestad T, Stevens R (1999) Compendium method for the determination of inorganic compounds in ambient air: compendium method IO-4.2: determination of reactive acidic and basic gases and strong acidity of atmospheric fine particles (< 2.5 μm), EPA/625/R-96/010a. US Environmental Protection Agency, Cincinnati, OH
- Xie Y, Dai H, Dong H, Hanaoka T, Masui T (2016) Economic impacts from PM_{2.5} pollution-related health effects in China: a provincial-level analysis. *Environ Sci Technol* 50:4836–4843. <https://doi.org/10.1021/acs.est.5b05576>
- Xing L, Fu TM, Cao J, Lee S, Wang G, Ho K, Cheng MC, You CF, Wang T (2013) Seasonal and spatial variability of the OM/OC mass ratios

- and high regional correlation between oxalic acid and zinc in Chinese urban organic aerosols. *Atmos Chem Phys* 13:4307–4318
- Xu H, Cao J, Chow JC, Huang RJ, Shen Z, Chen LWA, Ho KF, Watson JG (2016) Inter-annual variability of wintertime PM_{2.5} chemical composition in Xi'an, China: evidences of changing source emissions. *Sci Total Environ*:545–546–546–555. <https://doi.org/10.1016/j.scitotenv.2015.12.070>
- Zhang Q, Streets DG, Carmichael GR, He KB, Huo H, Kannari A, Klimont Z, Park IS, Reddy S, Fu JS, Chen D, Duan L, Lei Y, Wang LT, Yao ZL (2009) Asian emissions in 2006 for the NASA INTEX-B mission. *Atmos Chem Phys* 9:5131–5153. <https://doi.org/10.5194/acp-9-5131-2009>
- Zhang F, Zhao J, Chen J, Xu Y, Xu L (2011) Pollution characteristics of organic and elemental carbon in PM_{2.5} in Xiamen, China. *J Environ Sci* 23:1342–1349
- Zhang Q, Shen Z, Cao J, Zhang R, Zhang L, Huang RJ, Zheng C, Wang L, Liu S, Xu H (2015) Variations in PM_{2.5}, TSP, BC, and trace gases (NO₂, SO₂, and O₃) between haze and non-haze episodes in winter over Xi'an, China. *Atmos Environ* 112:64–71
- Zhang Q, Jiang X, Tong D, Davis SJ, Zhao H, Geng G, Feng T, Zheng B, Lu Z, Streets DG, Ni R, Brauer M, van Donkelaar A, Martin RV, Huo H, Liu Z, Pan D, Kan H, Yan Y, Lin J, He K, Guan D (2017a) Transboundary health impacts of transported global air pollution and international trade. *Nature* 543:705–709. <https://doi.org/10.1038/nature21712>
- Zhang Y, Wei J, Tang A, Zheng A, Shao Z, Liu X (2017b) Chemical characteristics of PM_{2.5} during 2015 spring festival in Beijing, China. *Aerosol Air Qual Res* 17:1169–1180
- Zhang Y, Zhang H, Deng J, Du W, Hong Y, Xu L, Qiu Y, Hong Z, Wu X, Ma Q, Yao J, Chen J (2017c) Source regions and transport pathways of PM_{2.5} at a regional background site in East China. *Atmos Environ* 167:202–211. <https://doi.org/10.1016/j.atmosenv.2017.08.031>
- Zhao P, Dong F, He D, Zhao X, Zhang X, Zhang W, Yao Q, Liu H (2013) Characteristics of concentrations and chemical compositions for PM_{2.5} in the region of Beijing, Tianjin, and Hebei, China. *Atmos Chem Phys* 13:4631–4644
- Zhao M, Huang Z, Qiao T, Zhang Y, Xiu G, Yu J (2015) Chemical characterization, the transport pathways and potential sources of PM_{2.5} in Shanghai: seasonal variations. *Atmos Res* 158–159:66–78. <https://doi.org/10.1016/j.atmosres.2015.02.003>
- Zhou J, Xing Z, Deng J, Du K (2016) Characterizing and sourcing ambient PM_{2.5} over key emission regions in China I: water-soluble ions and carbonaceous fractions. *Atmos Environ* 135:20–30. <https://doi.org/10.1016/j.atmosenv.2016.03.054>