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보건학석사 학위 논문

**Characteristics of PM<sub>2.5</sub> and its chemical  
constituents in Seoul, Taipei, and Kanazawa**

대한민국 서울, 대만 타이베이, 일본 카나자와의  
대기 중 PM<sub>2.5</sub> 성분 특성 파악

2014년 2월

서울대학교 보건대학원  
환경보건학과 대기환경전공  
박 은 하

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지도교수 이 승 목  
이 논문을 보건학석사 학위 논문으로 제출함

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서울대학교 보건대학원  
환경보건학과 대기환경전공  
박 은 하

박은하의 보건학석사 학위 논문을 인준함  
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위 원 장 김 호 (인)  
부 위 원 장 조 경 덕 (인)  
위 원 이 승 목 (인)

## **Abstract**

# **Characteristics of PM<sub>2.5</sub> and Its Chemical Constituents in Seoul, Taipei, Kanazawa**

**Park, Eun Ha**

**Department of Environmental Health**

**The Graduate School of Public Health**

**Seoul National University**

PM<sub>2.5</sub> samples were collected at two urban monitoring sites in Seoul, Taipei, and at one rural monitoring site in Kanazawa. The sampling site in Seoul was located on the roof of the School of Public Health building at Seoul National University (37.5° N, 127.0° E) in Seoul, Korea, and the site of Taipei was located on the roof of Gu-ting elementary school (25.0°N, 121.3° E) in Taipei, Taiwan. The measurements of Kanazawa were made on the roof of the School of Pharmaceutical and Health Science building at Kanazawa University (36.3° N, 136.4° E) in Kanazawa, Japan. Samples were collected from midnight to midnight (00:00 ~ 24:00) mostly every third day (Seoul: from April 2010 - May 2013, Taipei: December 2010 - May 2013, Kanazawa: November 2011 - May 2013). The PM<sub>2.5</sub> and chemical speciated samples were collected using a 3-channel system for 24-hour. The system had one channel using an Annular Denuder System (ADS) and two channels using filter packs (URG

Corp.).

The PM<sub>2.5</sub> mass concentrations were 37, 36, and 25  $\mu\text{g}/\text{m}^3$  in Seoul, Taipei, and Kanazawa, respectively. The measured chemical constituents were carbonaceous species such as organic carbon (OC) and elemental carbon (EC), ion species including nitrate ( $\text{NO}_3^-$ ), sulfate ( $\text{SO}_4^{2-}$ ), ammonium ( $\text{NH}_4^+$ ), and trace elements. Sulfate was the highest in both Taipei (9.90  $\mu\text{g}/\text{m}^3$ ) and Kanazawa (7.35  $\mu\text{g}/\text{m}^3$ ), while nitrate was highest in Seoul (9.31  $\mu\text{g}/\text{m}^3$ ).

Backward trajectories were calculated using Hybrid Single-particle Lagrangian Integrated Trajectory (HYSPLIT) 4 model and Potential Source Contribution Function (PSCF) model was applied to identify the possible source locations of carbonaceous and ionic species. PSCF results showed that the possible source locations of most chemical constituents measured in Seoul were the industrial areas in the eastern coast, middle regions of China, and the Gobi Desert. In Taipei, PSCF results identified the southern part of China and South-East Asia as the possible source areas. Kanazawa was mostly affected by eastern coastal region of China, southern part of Korea, and the Pacific Ocean.

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**Keywords : PM<sub>2.5</sub>, Chemical constituents, Backward trajectories, HYSPLIT 4, PSCF**

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# 1. Introduction

## 1.1 Backgrounds

Particulate matter (PM), especially fine particles defined as particles less than 2.5  $\mu\text{m}$  in aerodynamic diameter ( $\text{PM}_{2.5}$ ) have significant effects on human health. The major constituents of  $\text{PM}_{2.5}$  are ionic species ( $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ , and  $\text{NH}_4^+$ ), various organic compounds, elemental carbon, metal compounds and trace elements (Heo et al., 2009; Han et al., 2012). The primary particles are PM emitted to the atmosphere directly from emission sources, while secondary particles are PM formed in the atmosphere by chemical reactions involving PM precursor emissions (Chu, 2004).

Whereas larger particles are derived from soil and other crustal materials, fine particles are primarily from the combustion of fossil fuels, manufacturing, and power generation. Several studies indicated that fine particles can more easily penetrate into the lungs through statistical associations between mortality and morbidity and concentrations of particulate matter. Therefore, fine particles can increase the rate of respiratory and cardiovascular disease more than larger particles (Dockery et al., 1993; Schwartz et al., 2002; Heo, 2010). For this reason, the United States Environmental Protection Agency (US EPA) promulgated a national ambient air quality standard since 1997. On December 2012, EPA lowered the national ambient air quality standard for fine particle ( $\text{PM}_{2.5}$ ) to 12  $\mu\text{g}/\text{m}^3$  and retaining the 24-hour standard of 35  $\mu\text{g}/\text{m}^3$  since 2006.

Also, chemical compositions affect health risks. Particularly engine exhaust particles are associated with health risks much more than those from natural

sources (Forsberg et al., 2005; Hong et al., 2010). Accordingly, chemical composition and sources of PM<sub>2.5</sub> should be identified to establish pertinent strategies. Moreover, because of the very small particle size, PM<sub>2.5</sub> generally has a long lifetime in the atmosphere varying from a few days to weeks and tends to spread over a much larger geographic region (Wolff et al., 1985; Eldred and Cahill, 1994; Wilson and Suh, 1997; Chu, 2004).

Concentrations of PM<sub>2.5</sub> have been increasing due to the increased local sources such as vehicle emissions. However, long-range transport from the industrial areas in China can also account for it. Air pollution has become a critical problem in China because industries in China have been developed so rapidly (Han et al., 2008). Also, air pollution in China can cause severe problems in adjacent countries such as Korea, Taiwan, and Japan. Especially during Asian Dust, also known as yellow-sand events, soil constituents and anthropogenic components from China have been transported to Seoul, Korea (Yi et al., 2001). Airborne particles are transported from the dry deserts of southern Mongolia and northern China by prevailing westerly winds during Asian Dust (Tsai and Chen, 2006). It affected middle latitude regions including some Chinese regions (Zhang and Iwasaka, 1999; Guo et al., 2004), Korea (Chung, 1992; Kim and Park, 2001; Ma et al., 2004), Japan (Ma et al., 2001, 2005; Nakamura et al., 2005), and Taiwan (Lin, 2001).

## 1.2 Objectives

The overall objectives of this study were to investigate the major chemical constituents of  $PM_{2.5}$  and to identify the possible source locations.

The specific objectives of this study were i) to investigate the major chemical constituents of  $PM_{2.5}$  in Seoul, Korea, in Taipei, Taiwan, and in Kanazawa, Japan, ii) to compare its chemical constituents among those three cities, and iii) to identify possible source locations of  $PM_{2.5}$  and its chemical species using Possible Source Contribution Function (PSCF).

## 2. Experimental Methods

### 2.1 Materials and Sampling Methods

PM<sub>2.5</sub> samples were collected at urban monitoring sites in Seoul, Taipei, and at one rural monitoring site in Kanazawa. Seoul is the capital and largest metropolis of Korea, and Taipei is the capital of Taiwan which is located on the northern tip of Taiwan. Kanazawa is located on the western margin of central Japan facing the East Sea. The sampling site in Seoul was located on the roof of the School of Public Health building at Seoul National University (37.5° N, 127.0° E) in Seoul, and the site of Taipei was located on the roof of Gu-ting elementary school (25.0° N, 121.3° E) in Taipei, Taiwan. One of the national ambient air monitoring sites of Taipei is located on the same sampling site about 4 meters away. The measurement site of Kanazawa was made on the roof of the School of Pharmaceutical and Health Science building at Kanazawa University (36.3° N, 136.4° E) in Kanazawa, Japan (Figure 1).

PM<sub>2.5</sub> samples were collected from midnight to midnight (00:00 ~ 24:00) mostly every third day from April 2010 to May 2013 in Seoul, December 2010 to May 2013 in Taipei, and November 2011 to May 2013 in Kanazawa. 24-hour PM<sub>2.5</sub> and chemical speciated samples were collected using a 3-channel system (Figure 2). The system had one channel using an Annular Denuder System (ADS) and two channels using filter packs (URG Corp.), similar to the EPA Compendium Method IO-4.2 (US EPA, 1999).

Every channel was composed of size-selective inlet, cyclone, vacuum pump, and dry gas meter. Cyclones (URG-2000-30EH and URG-2000-30EN, URG) provide a particle size cutoff based on the flow rate, and the flow rate was monitored for each channel by independent dry gas meters. Flow rates were 10 L/min for ionic species and 16.7 L/min for carbonaceous species and trace elements.

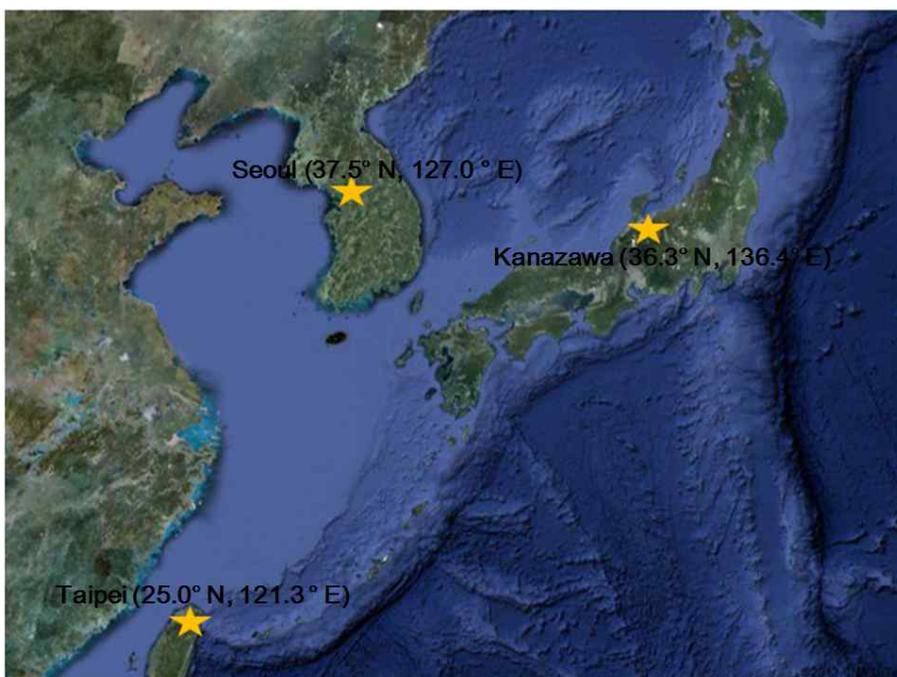


Figure 1. The locations of sampling sites in this study (Seoul, Taipei, and Kanazawa).



Figure 2. Three channel system of PM<sub>2.5</sub>.

## 2.2 Analytical Method

Mass concentrations of PM<sub>2.5</sub> were measured in each sampled cities after 24 hours from the sampling. Ion species (NO<sub>3</sub><sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup>) and carbonaceous species (OC, EC) were analyzed in Korea. They were stored at a refrigerator and sent to Korea every month. Teflon filters for trace elements which were weighed with a microbalance were sent to Clarkson University in New York, U.S. and analyzed using a X-ray Fluorescence (XRF). In this study, carbonaceous species, ion species, and trace elements of the three cities were analyzed in the same conditions and places. This singularity of this study can raise accuracy to compare chemical constituents of the three cities.

### 2.2.1 Mass Concentration of PM<sub>2.5</sub>

Channel 1 had Teflon filters (47 mm Gelman Teflo, 3 μm pore size) to collect particles for mass by gravimetry. The PM<sub>2.5</sub> mass was measured by weighing the Teflon filters with a microbalance (Sartorius, Japan, Precision : 10<sup>-2</sup> mg). A Teflon filter which has been stored at desiccator at least 24 hours was weighed before sampling. After sampling, it was dried in desiccator for 24 hours and then weighed again. Mass concentrations of PM<sub>2.5</sub> were calculated by the difference of them, and blank filter was always weighed to modify.

## 2.2.2 Ionic Constituents of PM<sub>2.5</sub>

Ionic constituents were measured in the second channel using 3-stage filter pack and the ADS. The ADS consists of annular denuders coated with sodium carbonate for anions and citric acid for cations. Filter pack contained Zeflour membrane filters (47 mm Pall Life Sciences, 2  $\mu\text{m}$  pore size) followed by Nylon membrane filters (47 mm Gelman Science, 1  $\mu\text{m}$  pore size) and Whatman paper filters (47 mm Whatman International Ltd.) coated with citric acid for anion and cation back-up filters.

Water soluble ion species ( $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^+$ ) were extracted and analyzed by the ion chromatography (IC) which is a liquid chromatography technique using ion exchange mechanisms.

Table 1. Operating conditions for Ion Chromatography

Species	Anion ( $\text{NO}_3^-$ , $\text{SO}_4^{2-}$ )	Cation ( $\text{NH}_4^+$ )
Instrument	DX-120 (Dionex)	
Column	Ionpac AS14A 4 × 250 mm	Ionpac CS12A 4 × 250 mm
Eluent	8.0 mM Sodium Carbonate + 1.0 mM Sodium Bicarbonate	20 mM Methane Sulfonic Acid
Flow rate	1 mL/min	
Suppressor	ASRS™ 300 4mm # 064554	CSRS™ 300 4mm # 064556
Standard solution	Seven Anion Standard II # 057590 (Dionex)	Six Cation Standard II # 046070 (Dionex)



Figure 3. Ion Chromatography (DX-120, Dionex).

### 2.2.3 Carbonaceous Species of PM<sub>2.5</sub>

Quartz microfiber filter (47 mm Whatman International Ltd.) which was pre-baked at 450 °C for 12-hour to lower their carbon blanks was used to collect carbonaceous species. Organic carbon (OC) and elemental carbon (EC) were analyzed by the National Institute of Occupational Safety and Health (NIOSH) 5040 method with Thermal/Optical Transmittance (TOT) (Birch and Cary, 1996). Collected Quartz filter samples were punched to a standard size (1.5 cm × 1.0 cm) and placed in a quartz oven. Once the oven is purged with helium gas, a stepped temperature ramp increased the oven temperature to 870 °C in 4 stages for OC and 6 stages for EC.

Table 2. OC and EC operation program used in this study

Step	Carrier Gas	Ramp Time (seconds)	Program Temperature (°C)
1	Helium	60	315
2	Helium	60	475
3	Helium	60	615
4	Helium	90	870
	Helium	oven heaters are turned off to cool oven	
5	2% Ox in He	45	550
6	2% Ox in He	45	625
7	2% Ox in He	45	700
8	2% Ox in He	45	775
9	2% Ox in He	45	850
10	2% Ox in He	120	910
	Cal Gas + Helium/Ox	External Std. Calibration and cool-down	



Figure 4. Thermal / Optical Transmittance (TOT) (OC/EC Carbon Aerosol Analyzer Model 3, Sunset Laboratory Inc.).

## 2.2.4 Quality Assurance and Quality Control (QA/QC)

QA/QC was conducted out to curtail error during procedures for data certification. During the sampling periods, the flow rate was monitored for each channel by independent dry gas meters. Cyclones provided a particle size cutoff based on the flow rate, so the flow rate was modified before every sampling. Flow rates were 10 L/min for ionic species and 16.7 L/min for mass concentration, carbonaceous species and trace elements. To reduce the errors caused by static, static eliminator was used during analysis of the PM<sub>2.5</sub> mass concentration which was measured by weighing the Teflon filters. In addition, the blank filters were stored and weighed together to be modified. In case of ion species, standard materials (Seven Anion Standard II and Six Cation Standard II, Dionex) were analyzed routinely to examine the accuracy. The standard curve was used when the coefficient of determination ( $r^2$ ) was greater than 0.999 (linear) over a standard solution concentration ranging from 0.1 to 20 ppm. Carbon standard solution was made up of  $10.00 \pm 0.01$  g of sucrose and distilled water, and was analyzed on the blank filter. For ionic species, recovery efficiencies were determined for every 20 samples by spiking standard solution of 2.5 ppm, and those were ranged from 93% to 104%, 90% to 104%, 94% to 110%, for  $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^+$  respectively. For OC and EC, reproducibility test had acceptable results, and those were ranged from 97% and 103%, and 99% and 102%, OC, and EC, respectively.

## 2.3 Model Overview

### 2.3.1 Hybrid Single-Particle Lagrangian Integrated Trajectory (HYSPLIT) 4 Model

Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model is a complete system to calculate simple air parcel trajectories which are defined as the paths of small particles of air at a certain point and time to complicated dispersion and deposition simulation (Stohl et al., 2002; Draxler and Hess, 2005; Seo, 2013). In this study, the 120-hour backward trajectories were calculated using the National Oceanic and Atmospheric Administration (NOAA) HYSPLIT4 Model and Global Data Assimilation System (GDAS) meteorological data.

### 2.3.2 Potential Source Contribution Function (PSCF) Model

Potential Source Contribution Function (PSCF) is a hybrid receptor model which uses chemical data of the receptor sites and combined backward trajectories which are originated from receptor sites so as to locate source areas (Han et al., 2008). The PSCF model was developed by Ashbaugh et al. (1985) and Malm et al. (1986). Both chemical and meteorological data for each sample are required for PSCF. The areas around the receptor site are divided into equal sized grid cells. The PSCF model counts each trajectory segments of an endpoint that terminates within a given grid cell. At the

receptor site, the possibility is associated to the number of endpoints in that cell related with the total number of endpoints for all sampling dates (Han et al., 2004; Seo, 2013).

The PSCF value,  $PSCF_{ij}$ , can be interpreted as the conditional possibility that an air parcel which passed the  $ij$ -th cell were in a high concentration on the monitoring site and was defined as:

$$PSCF_{ij} = \frac{m_{ij}}{n_{ij}}$$

where,  $n_{ij}$  is the total number of end points that passed through the  $ij$ -th cell, and  $m_{ij}$  is the number of end points of the  $ij$ -th cell which are associated with sample over the threshold criteria (Heo et al., 2009).

### 3. Results and Discussion

#### 3.1 Chemical Compositions of PM<sub>2.5</sub> in Seoul, Taipei, and Kanazawa

Figure 5 shows time series for PM<sub>2.5</sub> concentrations of three cities. Figures 6, 7, and 8 show the concentrations of chemical species, and the fraction of chemical species in PM<sub>2.5</sub>. Trace elements were classified as crustal (Al, Si, Ti, Fe, Mg, Na), mixed (Cl, Ca, K, Mn), and anthropogenic (Cr, Ni, Cu, Zn, Br, Pb, S) elements.

The average mass concentration of PM<sub>2.5</sub> was 37  $\mu\text{g}/\text{m}^3$  in Seoul, and its standard deviation was 82  $\mu\text{g}/\text{m}^3$ . This result indicates that the large variation of PM<sub>2.5</sub> concentration in Seoul was possibly due to the long-range transport. Nitrate was the highest in Seoul (9.31  $\mu\text{g}/\text{m}^3$ )  $\text{SO}_4^{2-}$  (7.30  $\mu\text{g}/\text{m}^3$ ) >  $\text{NH}_4^+$  (6.59  $\mu\text{g}/\text{m}^3$ ) > OC (4.96  $\mu\text{g}/\text{m}^3$ ) > EC (4.26  $\mu\text{g}/\text{m}^3$ ) > crustal (3.50  $\mu\text{g}/\text{m}^3$ ) > anthropogenic (2.96  $\mu\text{g}/\text{m}^3$ ) > mixed (1.24  $\mu\text{g}/\text{m}^3$ ).

The average mass concentration of PM<sub>2.5</sub> measured in Taipei was 36 ± 35  $\mu\text{g}/\text{m}^3$ . Sulfate was the highest constituent in Taipei (9.90  $\mu\text{g}/\text{m}^3$ ) followed by  $\text{NO}_3^-$  (9.31  $\mu\text{g}/\text{m}^3$ ) >  $\text{NH}_4^+$  (6.59  $\mu\text{g}/\text{m}^3$ ) > OC (3.18  $\mu\text{g}/\text{m}^3$ ) > crustal (3.10  $\mu\text{g}/\text{m}^3$ ) > anthropogenic (2.95  $\mu\text{g}/\text{m}^3$ ) > EC (2.11  $\mu\text{g}/\text{m}^3$ ) > mixed (1.17  $\mu\text{g}/\text{m}^3$ ). Concentrations of ion species were high (25.8  $\mu\text{g}/\text{m}^3$ ) while carbonaceous species were low (5.29  $\mu\text{g}/\text{m}^3$ ) in Taipei.

The average mass concentration of PM<sub>2.5</sub> in Kanazawa was 25  $\mu\text{g}/\text{m}^3$  and its standard deviation was 23  $\mu\text{g}/\text{m}^3$ . Sulfate was the highest in Kanazawa (7.35

$\mu\text{g}/\text{m}^3$ ) followed by OC ( $3.41 \mu\text{g}/\text{m}^3$ ) > crustal ( $2.74 \mu\text{g}/\text{m}^3$ ) >  $\text{NO}_3^-$  ( $2.65 \mu\text{g}/\text{m}^3$ ) > anthropogenic ( $2.49 \mu\text{g}/\text{m}^3$ ) >  $\text{NH}_4^+$  ( $1.71 \mu\text{g}/\text{m}^3$ ) > EC ( $1.11 \mu\text{g}/\text{m}^3$ ) > mixed ( $0.67 \mu\text{g}/\text{m}^3$ ).

Figure 9 shows the comparisons of  $\text{PM}_{2.5}$  and its chemical constituents between three cities Seoul, Taipei, and Kanazawa. Average  $\text{PM}_{2.5}$  mass concentration measured in Kanazawa ( $25 \pm 23 \mu\text{g}/\text{m}^3$ ) was much lower than Taipei ( $36 \pm 35 \mu\text{g}/\text{m}^3$ ) and Seoul ( $37 \pm 82 \mu\text{g}/\text{m}^3$ ). OC was the highest in Seoul ( $4.96 \mu\text{g}/\text{m}^3$ ) and EC was high in Seoul ( $4.26 \mu\text{g}/\text{m}^3$ ) followed by Taipei ( $2.11 \mu\text{g}/\text{m}^3$ ) and Kanazawa ( $1.11 \mu\text{g}/\text{m}^3$ ).  $\text{NO}_3^-$  and  $\text{NH}_4^+$  were high in Seoul ( $9.31 \mu\text{g}/\text{m}^3$  and  $6.59 \mu\text{g}/\text{m}^3$ ), whereas  $\text{SO}_4^{2-}$  was high in Taipei ( $9.90 \mu\text{g}/\text{m}^3$ ). Seoul and Taipei showed similar concentrations of trace elements and Kanazawa was lower than two cities (Seoul and Taipei). Average concentration of crustal elements was the highest among the trace elements, while average concentration of mixed elements was the lowest.

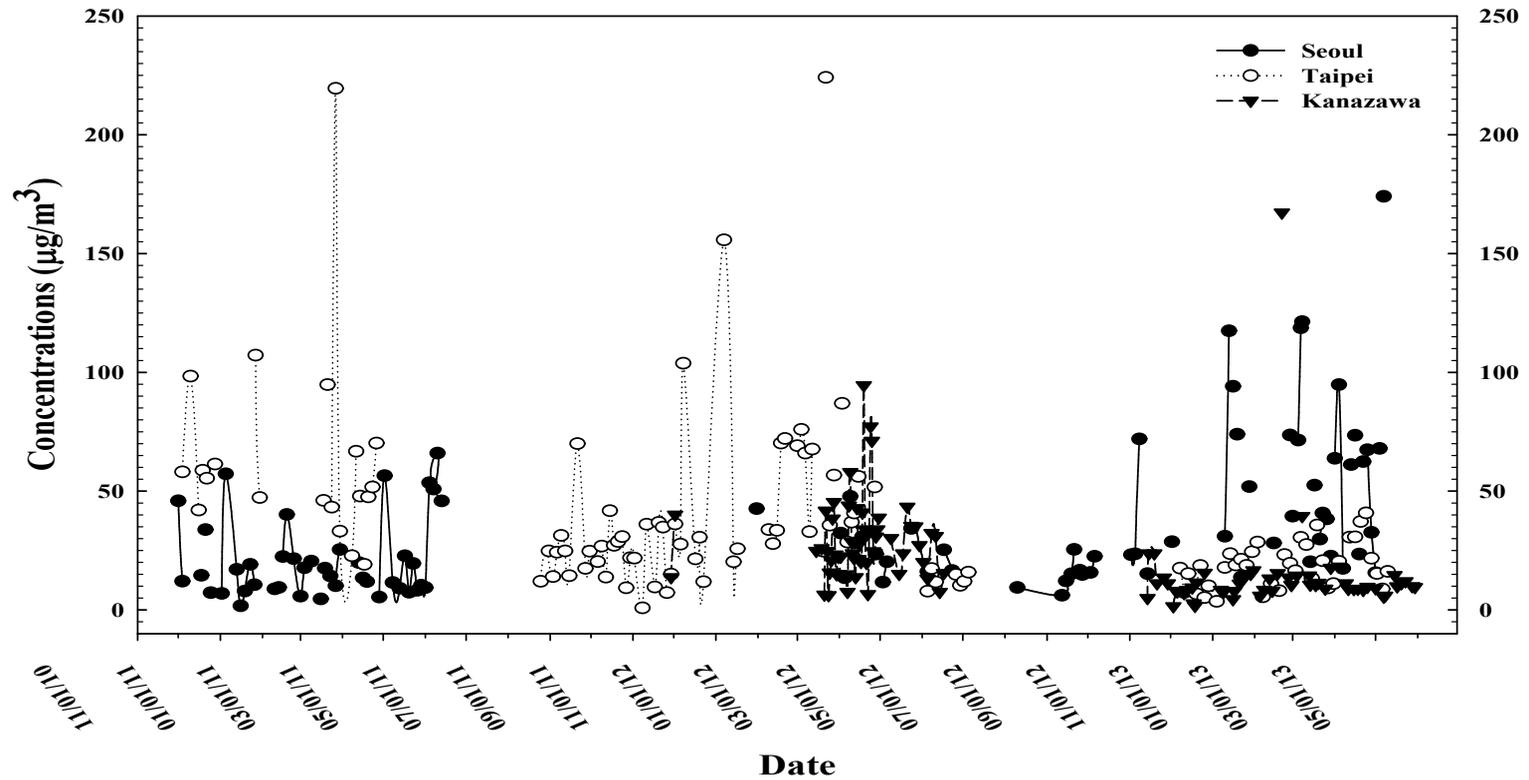


Figure 5. Time series of  $\text{PM}_{2.5}$  concentrations measured in three cities (Seoul, Taipei, and Kanazawa). Note that sampling site of Seoul was under construction from June 2011 to March 2012. Taipei (June 2011 to August 2011 and August 2012) and Kanazawa (December 2011 to February 2012 and July 2012 to October 2012) had no samples due to rainy season and heavy typhoons.

Table 3. The concentrations of chemical constituents in PM<sub>2.5</sub> measured in Seoul, Taipei, and Kanazawa (unit in  $\mu\text{g}/\text{m}^3$ )

		Seoul	Taipei	Kanazawa
PM <sub>2.5</sub>	N	141	120	106
	Mean	36.6	36.0	25.1
	SD	82.4	35.3	22.7
	min	0.844	0.850	1.55
	max	712	224	167
OC	N	133	118	111
	Mean	4.96	3.18	3.41
	SD	1.97	1.48	2.16
	min	1.79	0.320	0.729
	max	13.7	8.91	17.8
EC	N	133	117	108
	Mean	4.26	2.11	1.11
	SD	2.82	1.57	0.925
	min	0.574	0.0227	0.0500
	max	18.6	7.77	6.18
NO <sub>3</sub> <sup>-</sup>	N	127	117	120
	Mean	9.31	4.94	2.65
	SD	7.14	2.81	2.64
	min	1.30	1.65	0.832
	max	39.4	18.1	18.3
SO <sub>4</sub> <sup>2-</sup>	N	127	111	120
	Mean	7.30	9.90	7.35
	SD	5.82	7.66	7.94
	min	1.35	1.36	0.193
	max	36.9	38.2	36.1
NH <sub>4</sub> <sup>+</sup>	N	99	106	120
	Mean	6.59	4.69	1.71
	SD	5.01	3.19	1.85
	min	0.706	0.243	0.251
	max	23.9	17.4	16.8
Crustal	N	140	123	111
	Mean	3.50	3.10	2.74
	SD	3.95	1.72	2.04
	min	0.804	1.19	0.750
	max	41.1	13.0	11.3
Mixed	N	140	123	111
	Mean	1.24	1.17	0.670
	SD	1.67	1.01	0.670
	min	0.0863	0.179	0.107
	max	11.7	6.13	3.63
Anthropogenic	N	140	123	111
	Mean	2.96	2.95	2.49
	SD	2.16	1.74	1.89
	min	0.273	0.592	0.370
	max	11.1	8.70	11.3

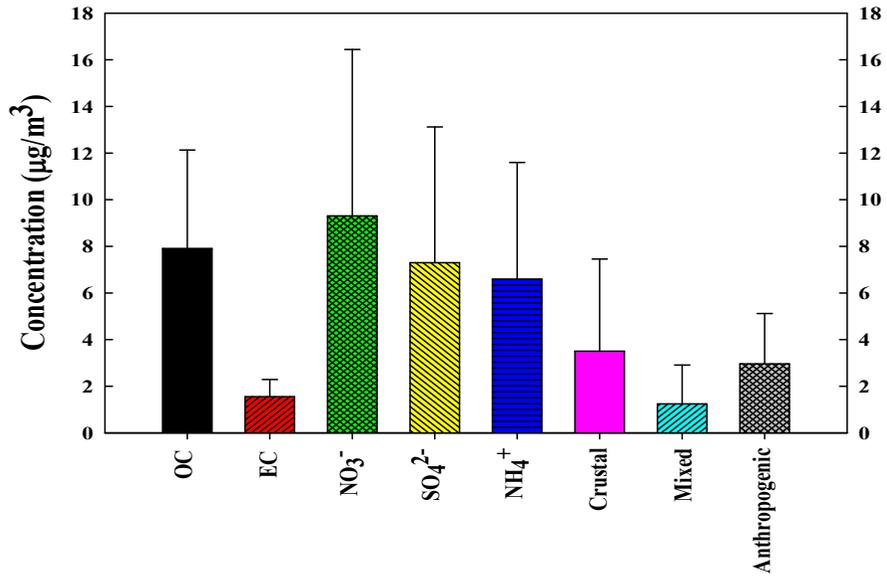


Figure 6. Average concentrations of chemical constituents in PM<sub>2.5</sub> measured in Seoul (error bars represent standard deviations).

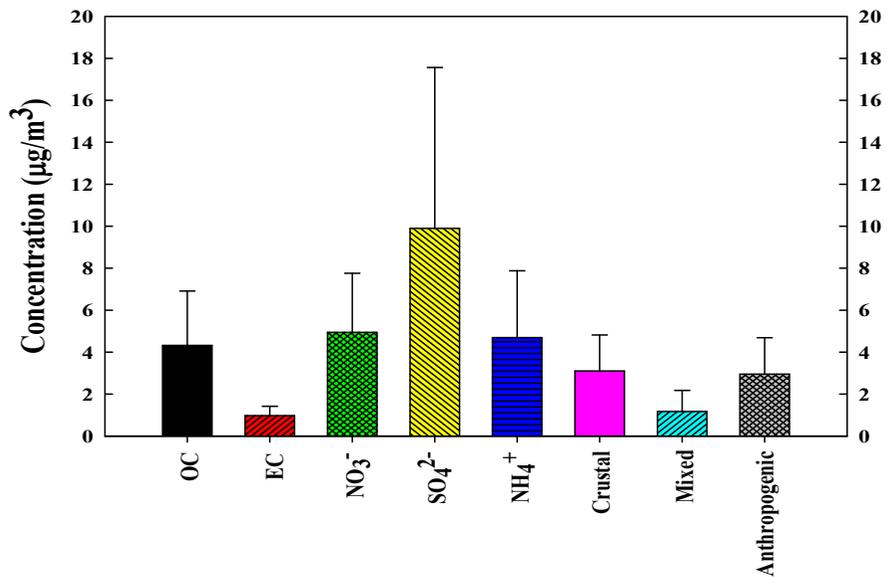


Figure 7. Average concentrations of chemical constituents in PM<sub>2.5</sub> measured in Taipei (error bars represent standard deviations).

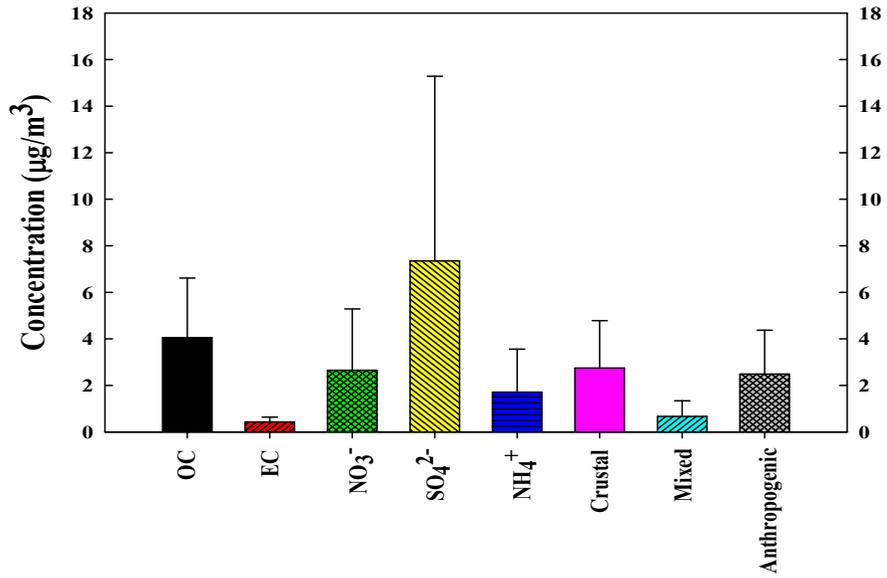


Figure 8. Average concentrations of chemical constituents in PM<sub>2.5</sub> measured in Kanazawa (error bars represent standard deviations).

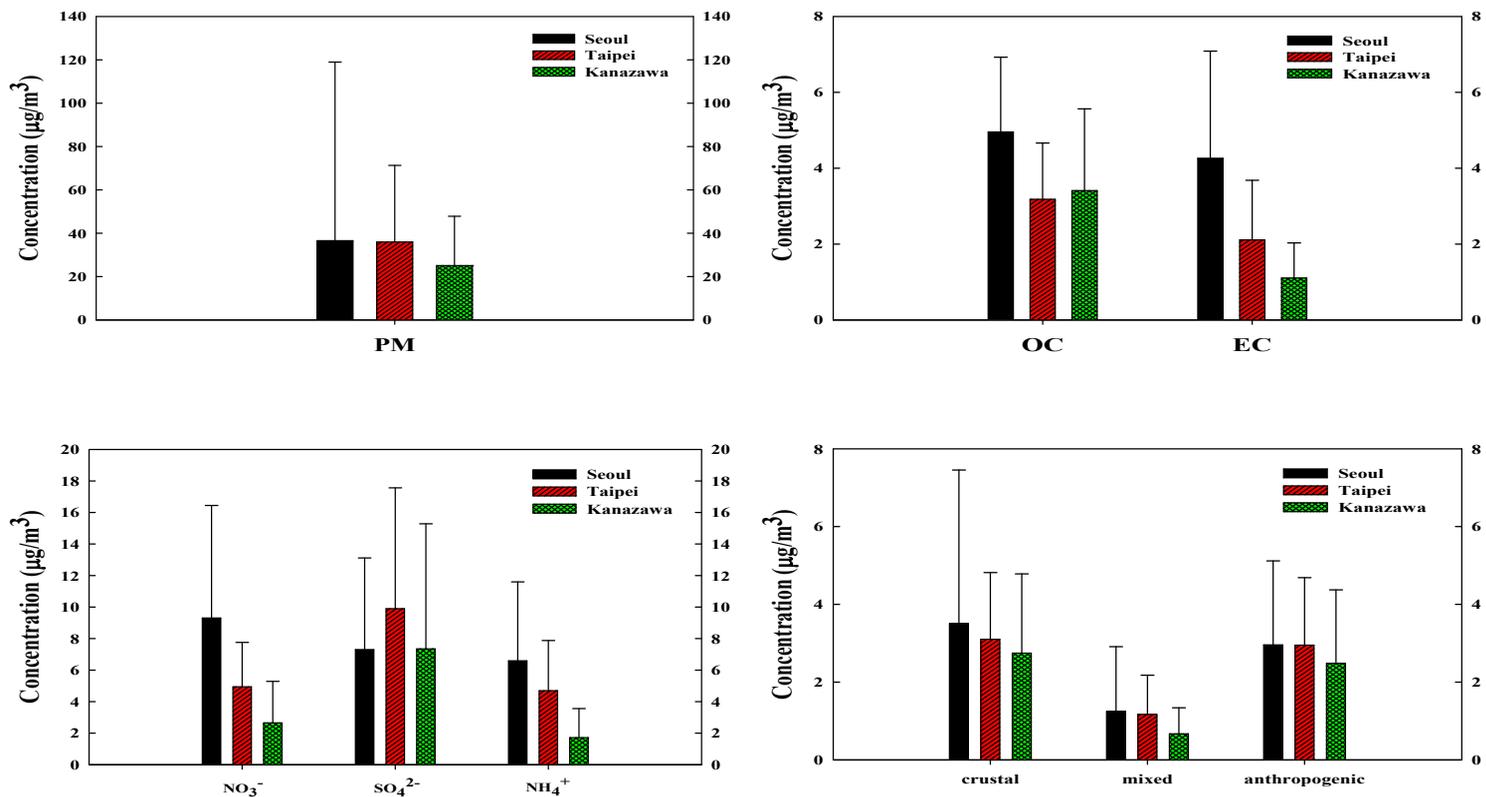
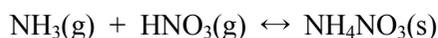
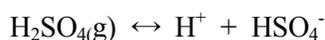


Figure 9. Average concentrations of chemical constituents in PM<sub>2.5</sub> measured in Seoul, Taipei, and Kanazawa (error bars represent standard deviations).

Ammonia is the only base in the ambient air with adequate quantities to neutralize the nitric and sulfuric acids. On top of that, ammonia is the important pathway to the formation of secondary nitrate and secondary sulfate (Seinfeld and Pandis, 1998). Therefore, the aerosol acidity of nitrate and sulfate particles can be evaluated using the aerosol electro-neutrality relationship between ammonium and sulfate and nitrate ions (Chu, 2004). Each mole of sulfate will remove 2 mole of ammonia from the gas phase, and it will be available for the reaction with nitrate if there is excess ammonia (Han et al., 2008).



The ammonium availability index,  $J$ , which is defined as the molar ratio of an observed 24-hour ammonium cation concentration to the amount needed to fully neutralize observed 24-hour sulfate and nitrate anion concentrations as follows:

$$J = \frac{[\text{NH}_4^+]}{2[\text{SO}_4^{2-}] + [\text{NO}_3^-]} \times 100(\%)$$

If  $J > 100\%$ , the environment is ammonia rich to fully neutralize the sulfate

and nitrate; if  $J = 100\%$ , the aerosols are neutral, indicating sufficient neutralization of sulfate and nitrate; and when  $J < 100\%$ , there is an ammonium deficit which indicates that sulfate and nitrate are acidic (Chu, 2004; Kim et al., 2007).

The mean values of  $J$  during the sampling period were 127% for Seoul, 112% for Taipei, and 74% for Kanazawa. There were enough ammonium present to neutralize sulfate and nitrate in Seoul and Taipei, and also  $(\text{NH}_4)_2\text{SO}_4$  and  $\text{NH}_4\text{NO}_3$  were generally present in the atmosphere in Seoul and Taipei, while nitrate and sulfate are acidic in Kanazawa.

It is important that index  $J$  only indicates the acidity of the nitrate and sulfate particles, it does not accurately fit to the pH value of the monitored ambient aerosols. Calcium, sodium, organic acids, and many other species can also make for pH value of the aerosol. Furthermore, the index  $J$  is a daily averaged value which is calculated from the 24-hour filter based data, but the actual acidity of the nitrate and sulfate particles fluctuates during the day (Chu, 2004).

Carbonaceous particles in the atmosphere consist of two major components, OC and EC. EC is directly emitted from sources mostly during combustion, while OC either originates from direct emissions of particles or is formed in the atmosphere by chemical reactions. Secondary OC, formed during photo-oxidation of hydrocarbons by condensation of low vapor pressure products, is associated with smog and low visibility (Seinfeld, 1986; Chow et al., 1996; Lee and Kang, 2001; Kim et al., 2007).

There are no direct analytical techniques to separately quantify primary OC (POC) and secondary OC (SOC), which made it hard to distinguish. Higher OC/EC ratio could be an indicator of secondary formation. The ratio of OC/EC exceeding 2.0 has been used to identify the existence of secondary organic aerosols. (Turpin and Huntzicker, 1995; Lee and Kang, 2001; Russel et al., 2004).

Identification of POC and SOC is fairly important in further analysis (Yu et al., 2009). Measured or monitored EC data is used in the EC tracer method because POC ( $OC_{pri}$ ) and EC are usually emitted from the same combustion sources. The EC tracer method has been widely used to estimate the SOC ( $OC_{sec}$ ) aerosol (Turpin and Huntzicker 1995; Castro et al., 1999; Lin and Tai 2001; Cao et al., 2004).

The total measured OC could be written by:

$$OC_{measured} = OC_{pri} + OC_{sec}$$

POC could be distinguished in two types; OC emitted with EC which is referred to as  $OC_{combustion}$ , and OC emitted without EC which is referred to as

OC<sub>non-combustion</sub>. Therefore, OC<sub>pri</sub> was calculated as:

$$OC_{pri} = OC_{combustion} + OC_{non-combustion}$$

Considering that in all the samples the ratio of primary OC/EC remained at a fixed level over space and time,

$$OC_{combustion} = \left(\frac{OC}{EC}\right)_{pri} \times EC_{measured}$$

Then, OC<sub>sec</sub> could be calculated as:

$$OC_{sec} = OC_{measured} - [OC_{non-combustion} + \left(\frac{OC}{EC}\right)_{pri} \times EC_{measured}]$$

The parameters of primary OC/EC ratio and OC<sub>non-combustion</sub> were estimated by the linear regression model (Lin et al., 2009). The Deming regression method was used rather than normal least-squares linear regression (Lee et al., 2010). Deming regression is often used in the EC tracer method to figure out POC and SOC. Deming fit is better when the biomass burning contribution is high, and when limited information is available on the comparative uncertainties of OC and EC (Chu, 2005; Saylor et al., 2006).

Table 4 and Figure 10 show the seasonal variations on average concentrations of POC, SOC, and EC, and Figure 11 shows the fractions of POC and SOC in Seoul, Taipei, and Kanazawa. Total concentrations of carbonaceous species in Seoul, Taipei, and Kanazawa were high during winter, spring, and summer,

respectively. Generally, the concentrations of carbonaceous species were high during fall and winter because of the burning of fossil fuels for heating. However, in this study, the results were different with previous studies because of the limited number of samples. The average concentrations of SOC were high during spring in all three cities. In addition, the average fraction of SOC was high during spring in Seoul and Kanazawa, while the one was high during winter in Taipei.

Table 4 Concentrations of POC, SOC, and EC of PM<sub>2.5</sub> measured in Seoul, Taipei, and Kanazawa (unit in  $\mu\text{g}/\text{m}^3$ )

		Seoul			Taipei			Kanazawa		
		POC	SOC	EC	POC	SOC	EC	POC	SOC	EC
Total	N		132			118			111	
	Mean	4.67	0.39	4.43	2.88	0.31	1.95	2.55	0.72	1.19
Spring	N		56			45			71	
	Mean	3.59	1.00	3.52	3.12	0.39	2.47	2.35	1.31	1.20
Summer	N		21			9			6	
	Mean	4.19	0.54	3.36	3.24	0.23	1.42	3.78	0.29	1.44
Fall	N		29			25			7	
	Mean	4.60	0.66	4.47	2.56	0.42	1.97	2.27	0.25	1.40
Winter	N		26			39			27	
	Mean	6.31	0.48	6.36	2.60	0.34	1.94	1.80	1.04	0.72

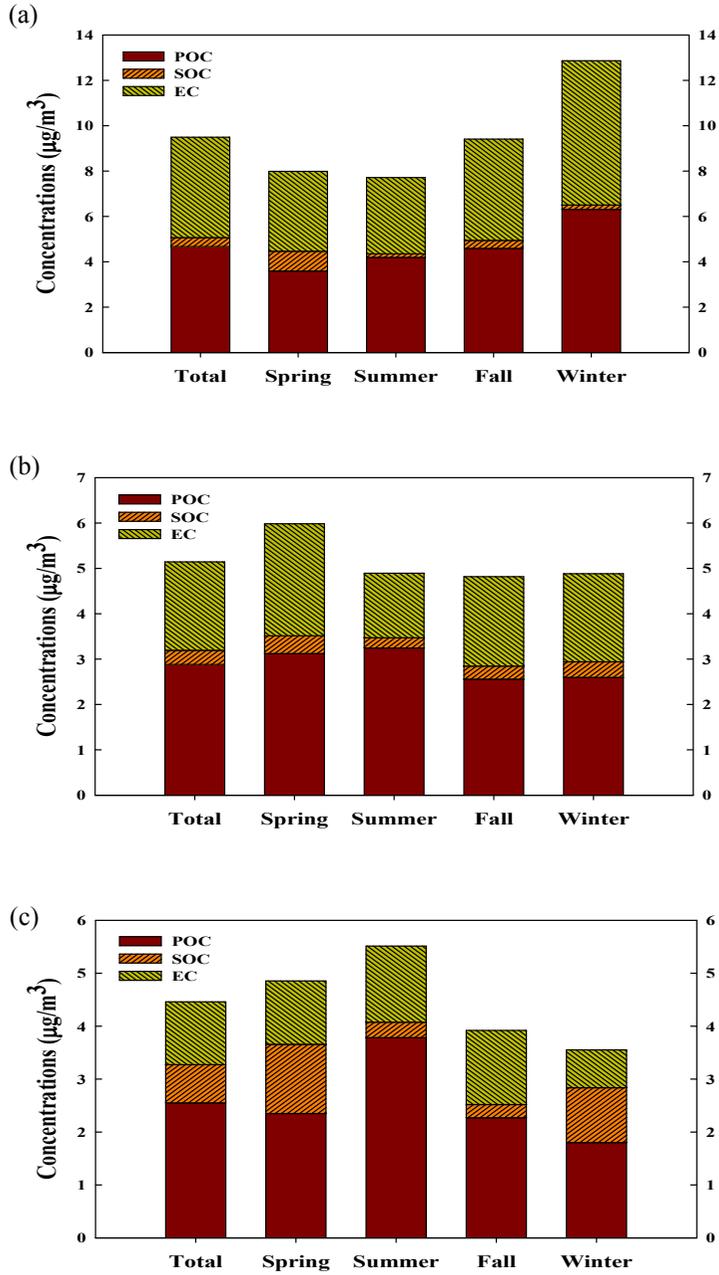


Figure 10. Seasonal variations of POC, SOC, and EC (a) Seoul, (b) Taipei, and (c) Kanazawa.

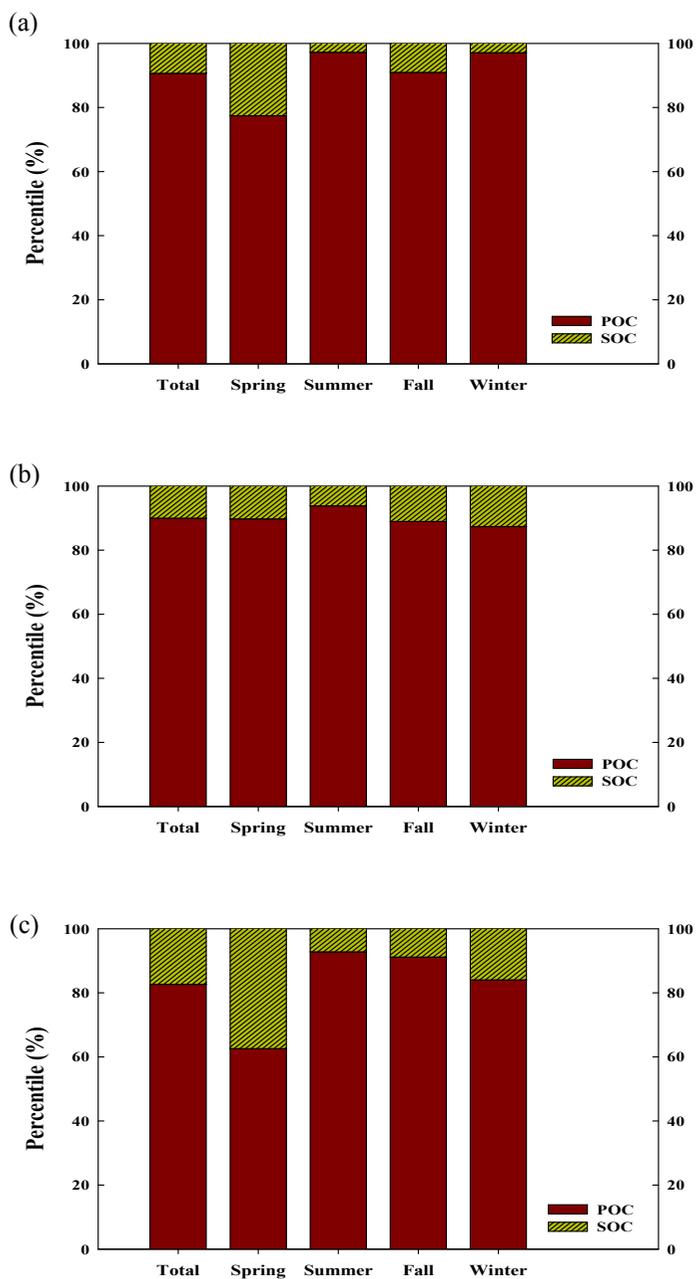


Figure 11. Seasonal mass fractions of POC and SOC (a) Seoul, (b) Taipei, and (c) Kanazawa.

## 3.2 Identification of Possible Source Areas

### 3.2.1 HYSPLIT 4 Model

Backward trajectories were calculated by HYSPLIT modeling system (Draxler and Rolph, 2007) and GDAS was used (Karaca, 2009). Five-day backward trajectories starting at every hour at a height of 1000 m above ground level were calculated for every sample day (Heo, 2010).

Prevailing wind in Seoul (37.5° N, 127.0° E) and Kanazawa (36.3° N, 136.4° E) was westerlies, while the one in Taipei (25.0° N, 121.3° E) was trade winds (Figure 12).



### 3.2.2 PSCF Model

PSCF analysis have been applied in a number of studies to identify the possible source locations of pollution (Zeng and Hopke, 1989; Gao et al, 1993, 1994, 1996; Cheng et al., 1993a, b; Hopke et al., 1993, 1995; Lucey et al., 2001). In this study, the criterion value was set to the 75<sup>th</sup> percentile and the results were shown in maps with each grid cell equal to 1° latitude by 1° longitude in size. Figure 13 showed total number of endpoints of 5-day backward wind trajectories, indicating the residence time of the backward trajectories passing through the grid cell in the PSCF.

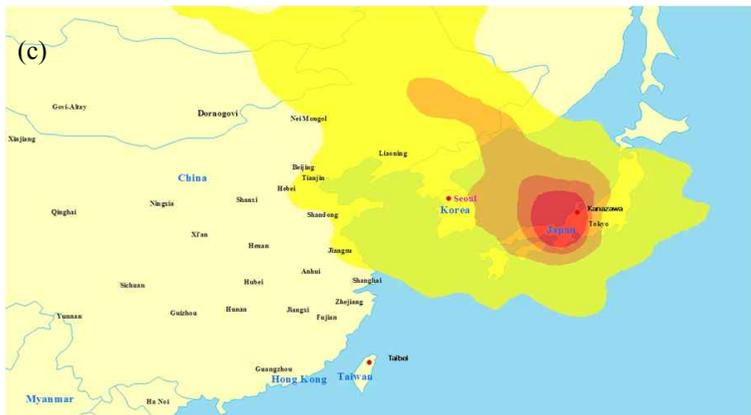
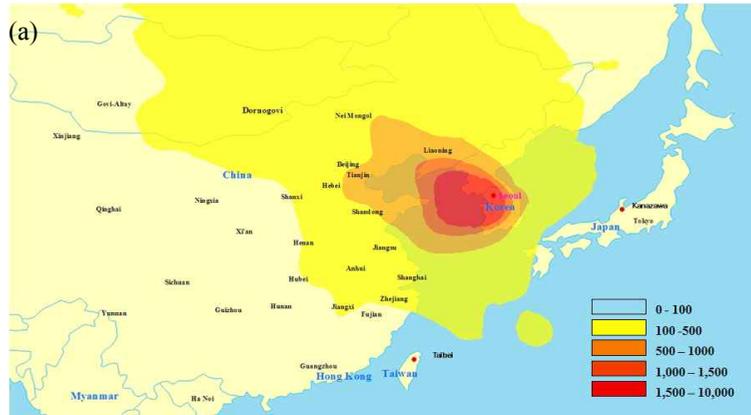


Figure 13. Total number of end points of 120-hour backward wind trajectories during the sampling period (a) Seoul, (b) Taipei, and (c) Kanazawa.

The PSCF result showed that the possible source locations of most chemical constituents measured in Seoul were the industrial areas in the eastern coast and middle regions in China and the Gobi desert (Figure 17). OC and EC originated from eastern and northern China (including Jiangxi, Zhejiang, Anhui, Shanghai, Shanxi, and Hebei) and Dornogovi were transported to Seoul as shown in Figures 14 (a) and (b). High PSCF values for nitrate in Seoul were found in heavily industrialized areas in eastern and central China like Shandong, Hebei, Beijing, and Tianjin. Dornogovi and northwestern China were also identified as possible source areas. The PSCF result of sulfate showed the Yellow Sea and industrial areas of the eastern coast in China as the possible source areas (Figure 14 (d)). The high PSCF values generated in this area were related to the thermal power plant, industrial, and residential emissions located in the coastal areas of China. In addition, there was a possible source areas of sulfate near the Yellow Sea probably due to the emissions of international and domestic shipping and fishing vessels on the sea. High PSCF values for ammonium were found in Zhejiang, Shanghai, Jiangxi, and central China areas. Zhejiang is the center of agriculture in China, and Shanghai is the main industrial area of China (Heo, 2009).

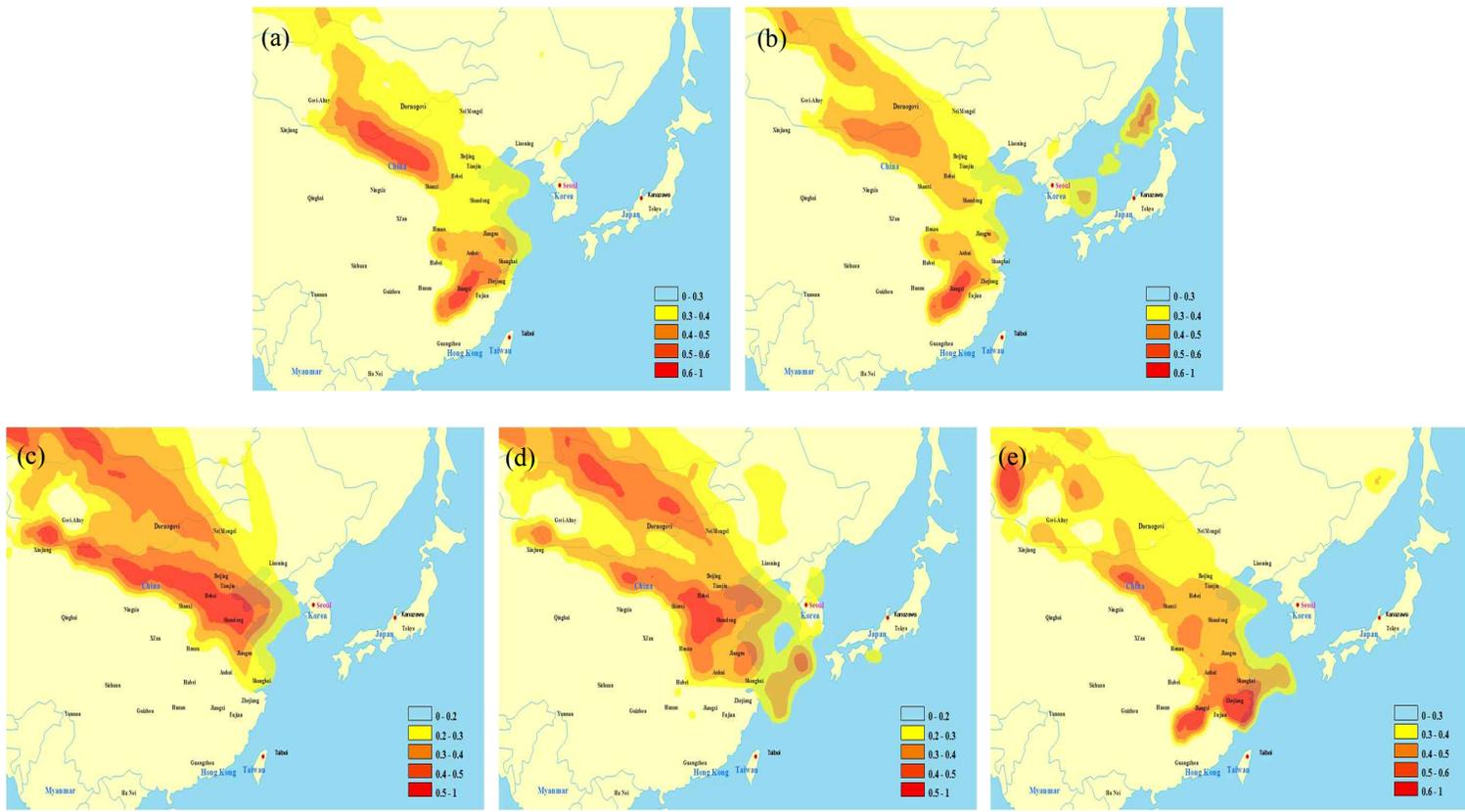


Figure 14. PSCF results for Seoul. (a) OC, (b) EC, (c) nitrate, (d) sulfate, and (e) ammonium

PSCF results for Taipei showed the southern part of China and South-East Asia as the possible source locations. OC and EC were originated from southern part of China including Guangzhou, Guangdong, Fujian, Hong Kong, and Macao. Some of the central China like Henan and Jiangxi were also identified as source areas. Jiangxi is the major regions of production for Kumquats and many crops in China, and Henan has the second largest molybdenum reserves in the world. Coal, aluminum, and alkaline metals are also present with large amount in Henan. Hong Kong and Macao are famous tourist regions, so traffic is always heavy. In addition, the second busiest container port and the busiest airport for international cargo in the world are present in Hong Kong. OC and EC were originated from the South China Sea, Hanoi, Philippines, Laos and little from Myanmar. EC was originated from the southern part of Korea, as well. High PSCF values for nitrate were found in Philippines, Hanoi, Laos, eastern part of Viet Nam, and southern part of China especially Guangzhou. Guangzhou is the third largest city in China and the largest city in southern China. The PSCF result for sulfate showed the South China Sea, Philippines, Hanoi, and southern part of China as well as the possible source areas, Dornogovi and Beijing. The PSCF result for ammonium showed the southern and the industrial areas in the eastern coast in China as the source areas (Figure 15 (e)).

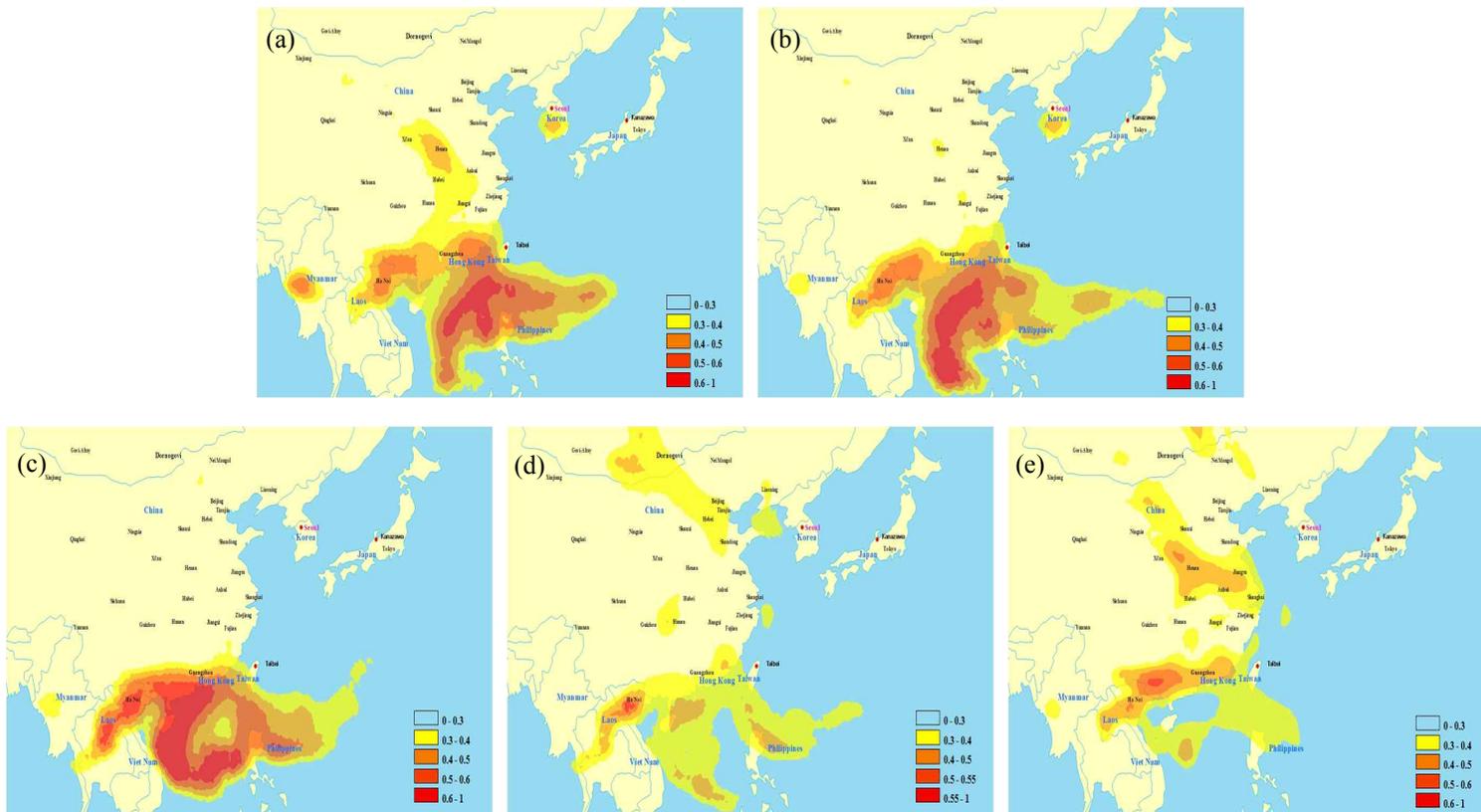


Figure 15. PSCF results for Taipei. (a) OC, (b) EC, (c) nitrate, (d) sulfate, and (e) ammonium

Kanazawa was mostly affected by eastern coastal regions of China, southern part of Korea, and Pacific Ocean. The PSCF results of OC and EC showed the eastern coastal regions of China including Jiangsu, Shandong, Hebei, and Tianjin as possible source areas. Also, PSCF results showed the southern industrial regions of Korea as possible source location. Nitrate was originated from the eastern part of China and southern Korea, but mostly from local source areas. High PSCF values for sulfate were shown in industrial regions of China like Shandong, Jiangsu, Shanghai, and Beijing. South industrial regions in China were also shown as the possible source areas. High PSCF values for ammonium were found in Beijing, Tianjin, Hebei, Shandong, and Jiangsu in China.

Some results showed oceans like East Sea, Yellow Sea, and Pacific Ocean as possible source areas. Because PSCF modeling evenly apportions weight along the path of trajectories, the PSCF results represent possible source directions rather than locations (Hsu et al., 2003). Therefore, the trailing effect usually appears, but it can be useful in finding the pathway between the possible source areas and the receptor site (Han et al., 2004).

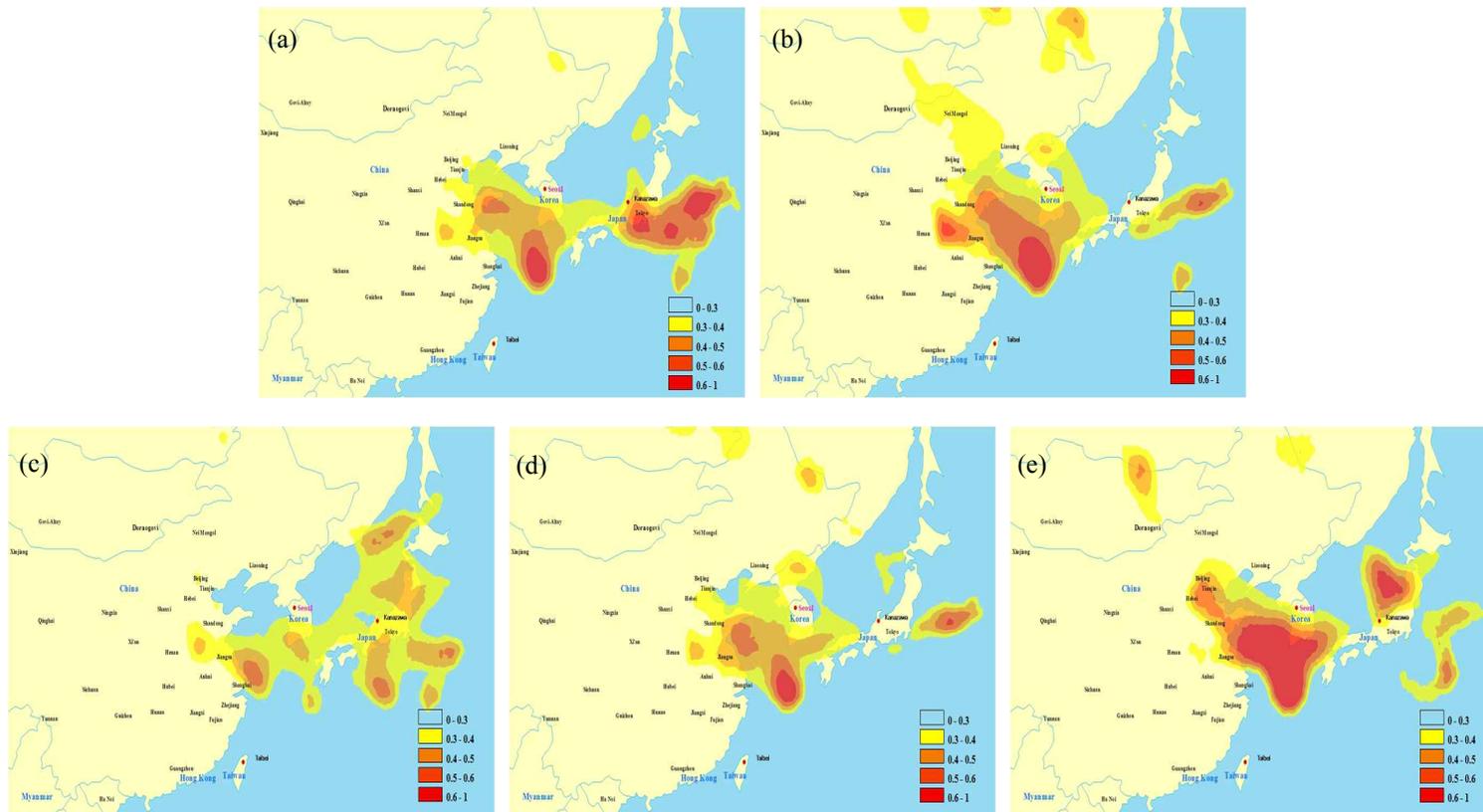


Figure 16. PSCF results for Kanazawa. (a) OC, (b) EC, (c) nitrate, (d) sulfate, and (e) ammonium

## 4. Conclusions

The overall objectives of this study were to investigate the major chemical constituents of PM<sub>2.5</sub> and to identify the possible source locations.

PM<sub>2.5</sub> samples were collected at two urban monitoring sites in Seoul, Taipei, and at a one rural monitoring site in Kanazawa. PM<sub>2.5</sub> samples were collected from midnight to midnight (00:00 ~ 24:00) mostly every third day (Seoul: April 2010 - May 2013), (Taipei: December 2010 - May 2013), (Kanazawa: November 2011 - May 2013). Major constituents of PM<sub>2.5</sub>, OC, EC, nitrate, sulfate, ammonium, and other trace elements, were analyzed in this study.

Backward trajectories were calculated by HYSPLIT modeling system and PSCF analysis was applied to identify the possible sources locations of pollution. In this study, the criterion value was set to the 75<sup>th</sup> percentile and the results were showed in maps with each grid cell equal to 1° latitude by 1° longitude in size.

The main findings of this study were summarized as follows:

(1) The average mass concentration of PM<sub>2.5</sub> was 37  $\mu\text{g}/\text{m}^3$  in Seoul, and its standard deviation was 82  $\mu\text{g}/\text{m}^3$ . This result indicate that the large variation of PM<sub>2.5</sub> concentration in Seoul was possibly due to the long-range transport. Nitrate was the highest in Seoul (9.31  $\mu\text{g}/\text{m}^3$ )  $\text{SO}_4^{2-}$  (7.30  $\mu\text{g}/\text{m}^3$ ) >  $\text{NH}_4^+$  (6.59  $\mu\text{g}/\text{m}^3$ ) > OC (4.96  $\mu\text{g}/\text{m}^3$ ) > EC (4.26  $\mu\text{g}/\text{m}^3$ ) > crustal (3.50  $\mu\text{g}/\text{m}^3$ ) >

anthropogenic ( $2.96 \mu\text{g}/\text{m}^3$ ) > mixed ( $1.24 \mu\text{g}/\text{m}^3$ ).

(2) The average mass concentration of  $\text{PM}_{2.5}$  measured in Taipei was  $36 \pm 35 \mu\text{g}/\text{m}^3$ . Sulfate was the highest constituent in Taipei ( $9.90 \mu\text{g}/\text{m}^3$ ) followed by  $\text{NO}_3^-$  ( $9.31 \mu\text{g}/\text{m}^3$ ) >  $\text{NH}_4^+$  ( $6.59 \mu\text{g}/\text{m}^3$ ) > OC ( $3.18 \mu\text{g}/\text{m}^3$ ) > crustal ( $3.10 \mu\text{g}/\text{m}^3$ ) > anthropogenic ( $2.95 \mu\text{g}/\text{m}^3$ ) > EC ( $2.11 \mu\text{g}/\text{m}^3$ ) > mixed ( $1.17 \mu\text{g}/\text{m}^3$ ).

(3) The average mass concentration of  $\text{PM}_{2.5}$  in Kanazawa was  $25 \mu\text{g}/\text{m}^3$  and its standard deviation was  $23 \mu\text{g}/\text{m}^3$ . Sulfate was the highest in Kanazawa ( $7.35 \mu\text{g}/\text{m}^3$ ) followed by OC ( $3.41 \mu\text{g}/\text{m}^3$ ) > crustal ( $2.74 \mu\text{g}/\text{m}^3$ ) >  $\text{NO}_3^-$  ( $2.65 \mu\text{g}/\text{m}^3$ ) > anthropogenic ( $2.49 \mu\text{g}/\text{m}^3$ ) >  $\text{NH}_4^+$  ( $1.71 \mu\text{g}/\text{m}^3$ ) > EC ( $1.11 \mu\text{g}/\text{m}^3$ ) > mixed ( $0.67 \mu\text{g}/\text{m}^3$ ).

(4) The mean values of J during the sampling period were 127% for Seoul, 112% for Taipei, and 74% for Kanazawa. There were enough ammonium present to neutralize sulfate and nitrate in Seoul and Taipei, and also  $(\text{NH}_4)_2\text{SO}_4$  and  $\text{NH}_4\text{NO}_3$  were generally present in the atmosphere in Seoul and Taipei, while nitrate and sulfate are acidic in Kanazawa.

(5) Total concentrations of carbonaceous species in Seoul, Taipei, and Kanazawa were high during winter, spring, and summer, respectively. The average concentrations of SOC were high during spring in all three cities. In addition, the average fraction of SOC was high during spring in Seoul and Kanazawa, while the one was high during winter in Taipei.

(6) Seoul (37.5° N, 127.0° E) and Kanazawa (36.3° N, 136.4° E) which are located on middle latitude (30° - 60° N) underwent the influence of westerlies, while the wind blew over to Taipei (25.0° N, 121.3° E) on the trade winds.

(7) The PSCF result shows the possible source locations of most constituents measured in Seoul were the industrial areas in the eastern coast and middle region in China and Gobi Desert. PSCF results for Taipei identify the southern part of China and South-East Asia as the possible source location. Kanazawa is affected by eastern coastal region of China and southern part of Korea.

There have been several studies analyzed in downwind countries from China such as Korea, Taiwan, and Japan. In this study, PM<sub>2.5</sub> and its chemical speciated samples were collected simultaneously by using same standard operating procedures (SOPs) in three cities. This study suggests that regional-scale study needs to be continued and developed, and the control of the long-range transport as well as local sources should be considered to control the level of PM<sub>2.5</sub>.

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## 초 록

# 대한민국 서울, 대만 타이베이, 일본 카나자와의 대기 중 PM<sub>2.5</sub> 성분 특성 파악

박은하

서울대학교보건대학원

환경보건학과

본 연구에서는 서울, 타이베이, 그리고 카나자와에서 채취한 PM<sub>2.5</sub> 성분의 특성을 살펴보았다. 서울은 2010년 4월부터 2013년 5월까지 서울시 중로구 연건동에 위치한 (구) 서울대학교 보건대학원 옥상(37.5° N, 127.0° E)에서 측정된 결과를 사용하였고, 타이베이는 2010년 12월부터 2013년 5월까지 타이베이시의 일반 모니터링 사이트 중 하나인 Gu-ting에서 측정된 결과를 사용하였다. 일본은 Kanazawa에 있는 Kanazawa 대학에서 2011년 11월부터 2013년 5월까지 측정하였고, 이 장소는 일본 내의 국지적인 오염원의 영향 보다는 외부에서 장거리 이동되어 오는 오염물질을 측정할 수 있는 장소이다. 샘플은 3일에 한번씩 24시간 동안 채취하였다.

연구 기간 중 PM<sub>2.5</sub>의 평균 농도는 서울은 37  $\mu\text{g}/\text{m}^3$ , 타이베이는 36  $\mu\text{g}/\text{m}^3$ , 카나자와는 25  $\mu\text{g}/\text{m}^3$  이었다. PM<sub>2.5</sub>의 화학적 조성 중 탄소성분은 유기탄소(OC)와 원소탄소(EC)로 분석하고, 이온성분( $\text{NO}_3^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NH}_4^+$ ), 그리고 미량원소 등 총 25가지로 분석하였다. 타이베이와 카나자와에서는  $\text{SO}_4^{2-}$  성분이 각각 9.90  $\mu\text{g}/\text{m}^3$ , 7.35  $\mu\text{g}/\text{m}^3$ 로 높았고, 서울은  $\text{NO}_3^-$  성분이

9.31  $\mu\text{g}/\text{m}^3$ 로 가장 높았다.

HYSPLIT4 모델을 이용하여 5일 동안의 역궤도 분석한 결과와 PSCF를 이용하여 성분들의 오염원 지역을 파악하였다. 그 결과 서울은 중국의 주요 공업 지역들과 몽골의 사막 지역이 오염원 지역으로 나타났고, 타이베이의 경우는 중국의 남부지역과 남동아시아 지역이 오염원 지역으로 나타났다. 카나자와는 중국 동부 해안지역과 한국 남쪽의 공업 지역, 그리고 대서양의 영향도 일부 받은 것으로 나타났다.

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주요어 : PM<sub>2.5</sub>, 화학적 조성, 역궤도 분석, HYSPLIT 4,  
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