#### 연구논문 (Article)

## 공기 궤 유입경로에 따른 한반도 서울 상공의 전체 및 유기 에어로졸 농도 변화 분석

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### Dependence of Total and Carbonaceous Aerosol Concentrations on Transport Pathways in Seoul, Korea

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**Abstract** Recently increased anthropogenic aerosols change the radiative energy balance and affect human life. The management of air quality requires monitoring both the local emissions and transported pollutants. In order to estimate the quantitative contribution of long-range transport from remote sources on aerosol concentrations in Seoul, the airmasses were classified into five types with respect to their pathways. When airmass came from west over strong emission regions in China, high concentrations of  $PM_{10}$ ,  $PM_{2.5}$ , black carbon (BC), organic carbon (OC), and elemental carbon (EC) were found, even higher than those for the stagnated airmass. High OC concentrations were found when airmass came from north while BC, EC, and  $PM_{2.5}$  concentrations were lower than those of the stagnated airmasses. During dust events, the  $PM_{2.5}$  and  $PM_{10}$  concentrations increased significantly while carbonaceous aerosol concentrations did not increased. The temporal variations of aerosol concentrations in Seoul were affected by the seasonal variations of airmass pathways. The high  $PM_{2.5}$  concentrations over 100 µg m<sup>-3</sup> appeared most frequently when the airmasses came from west.

Key words: Particulate matter, black carbon, organic carbon, elemental carbon, long range transport, East Asia

### 1. Introduction

Our earth-atmosphere system has maintained balance to natural aerosol including dust particles, pollens, and sea salt for a long time. However, recently emitted anthropogenic aerosols are changing the radiative energy balance and affecting human life such as public health and visibility (Ramanathan et al., 2001; Pope et al., 2002; Watson, 2002). Recent studies focused on the carbonaceous particles that can be classified into organic carbon (OC) and elemental carbon (EC). EC is nonvolatile and strong light absorbing particle which is generally emitted from biomass burning and incomplete

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combustion in automobile engines. Meanwhile, OC is generally volatile and light scattering particle which comprises a large number of species having large variations in volatility and sources (Seinfeld and Pandis, 2006). Black carbon (BC) has been often referred as EC and the two are well correlated. However, previous study reported that there can be distinctions between EC and BC due to the different measurement methods (Jeong et al., 2004).

Aerosol concentrations at a receptor site are typically affected by both the local emission and inflow pollutants from remote source areas. Previous studies investigated the effects of long-range transport on local air quality by using the chemical transport models, cluster methods or receptor models (Hwang and Hopke, 2007; Heo et al., 2009; Jeong et al., 2011a, b). However, estimation of the contribution of long-range transport on aerosol concentrations at a receptor site is still challenging since each method has its own advantages and limitations (Jeong et al., 2013). Seoul is one of representative megacities located at the center of Korean peninsula with populations over 20 millions including surrounding cities, thus strong aerosol emission sources are distributed throughout the metropolitan area. The aerosol concentrations in Seoul are also significantly affected by the long range transport from remote source areas at northwest direction since northwesterly wind dominates in Seoul except summer (Jeong et al., 2011b). Meanwhile, there are less source areas in the southeastern direction of Seoul. This study aims to investigate the dependence of the aerosol concentrations in Seoul with respect to the airmass pathways in order to quantify the effects of long range transport (Moy et al., 1994; He et al., 2003). The PM<sub>10</sub> (particulate matter with diameter less than 10 µm), PM<sub>2.5</sub> (particulate matter with diameter less than 2.5 µm), BC, EC, and OC samples collected in Seoul from April 2007 to March 2008 were employed for the analysis.

### 2. Measurements

The site is located at the rooftop of Science Hall, inside the campus of Yonsei University in Seoul, Korea, located at 37° 33 N in latitude and 126° 56 E in longitude. This site is surrounded by commercial and residential areas where heavy traffic affects air quality in part but strong industrial emission source is further away. Thus, the site can be considered as a typical urban background site for air quality monitoring. The samples of BC, OC, EC, PM2.5 and PM10 have been collected from April 2007 to March 2008 in this study. The BC concentration is obtained from the 7- $\lambda$ aethalometer (AE31, Magee Scientific) which measures the optical attenuation (absorbance) of light from LED lamps emitting at seven wavelengths (370, 470, 520, 590, 660, 880, and 950 nm) with a typical half-width of 0.02 nm (Hansen et al., 1984). Details of the instrument can be found in previous works (Weingartner et al., 2003; Schmid et al., 2006). The PM<sub>10</sub> and PM<sub>2.5</sub> were measured by an aerosol spectrometer (Model 265, Grimm Labortechnik Ltd., Ainring, Germany). The aerosol spectrometer estimates the mass concentration of aerosols by measuring the scattered light from a laser diode by dry particles (RH <40%). The detailed principles and applications are reported in Cheng (2008) and Cheng and Lin (2010). The uncertainties of the aerosol spectrometer are typically caused by the assumptions of refractive index, particle morphology, density, and size distribution (Cheng, 2008; Cheng and Lin, 2010). Thus, the measurement accuracy depends on the optical properties of the aerosols. The coarse mass is defined as particles with diameter larger than 2.5  $\mu$ m but smaller than 10  $\mu$ m (PM<sub>10</sub>-PM<sub>2.5</sub>) in this study. The OC and EC were sampled using a Sunset Laboratory semi-continuous organic-carbon/elementalcarbon (OC/EC) analyzer with a thermal-optical transmittance (TOT) protocol for pyrolysis correction (Birch and Cary, 1996; Jeong et al., 2004; Kim et al., 2006). The uncertainties of the instrument were reported to be about 5% (Polidori et al., 2006). In order to complement the missing period, both measurements were used despite BC and EC are well correlated.

# 3. Calculation of backward trajectory and airmass pathway classification

The airmass pathways to Seoul, Korea were classified into the five types (Type N - from north; Type W from west; Type S - from south; Type E - from east; Type C - stagnated in the local area, around Seoul) with respect to the 48-hour backward trajectory simulations as shown in Fig. 1. The backward trajectories arriving at Seoul at the height of 70 m above the ground level were calculated using HYSPLIT (Hybrid Single-Particle Lagrangian Integrated Trajectory) model developed by NOAA ARL (Air Resources Laboratory) every day



**Fig. 1.** Spatial distribution of AOD and classified pathways of aerosol transport to Seoul. (a) level 3 AOD at 550 nm from MODIS Aqua during measurement period; (b) type N: from north; (c) type S: from south; (d) type E: from east; (e) type W: from west; (f) type C: stagnated in the local area.

at 0900 LST. The NCEP (National Centers for Environmental Prediction) reanalysis data were used for the information of meteorological condition. Despite the HYSPLIT simulations contain non-negligible uncertainties, it represents the general tendencies of airflow with its position errors reported to be about 20% of the travel distance (Stohl, 1998).

In order to see the spatial distribution of the aerosols

 Table 1. Monthly variations of number of days for each type pathway.

		Type N	Type S	Type E	Type W	Type C
2007	Apr	12	1	0	9	8
	May	10	5	0	12	4
	Jun	0	2	11	4	13
	Jul	0	4	6	6	15
	Aug	1	15	2	6	7
	Sep	8	3	12	3	4
	Oct	11	1	4	4	11
	Nov	14	0	1	12	3
	Dec	14	0	1	11	5
2008	Jan	20	0	1	9	1
	Feb	18	1	0	9	1
	Mar	15	0	3	7	6
	Total	123	32	41	92	78

around Seoul, the mean aerosol optical depth (AOD) at 550 nm from April 2007 to March 2008 in East Asia measured by the Moderate Resolution Imaging Spectroradiometer (MODIS) onboard Aqua (EOS, earth observing system) was shown in Fig. 1 a (http://modisatmos.gsfc.nasa.gov/MOD08 M3/index.html). While the absolute values of AOD and surface level dry aerosol concentrations are not well correlated due to the variation of atmospheric boundary layer and hygroscopic properties of the aerosols, the spatial distribution of aerosols can be described quite well by AOD (Wang and Christopher, 2003). There are significant differences in AOD at northern and western region of Seoul, where the boundary between the two areas is selected to separate the type N and W. The type W represents the cases with airmass passed over the strong sources of aerosols in China, while type N represents cases with airmass passed over the less polluted continental area including North Korea. The type C represents the stagnated airmass for more than 2 days, thus affected by the local emission in Seoul. The type S and type E represents the airmass passed over the southern and eastern Seoul, respectively. The type S and type E also includes the marine aerosols. Corresponding numbers of dates for each airmass pathway type during the observational period are listed in Table 1. Due to the latitudinal location of Seoul, westerly and northerly winds dominated the airflow direction except summer (occurrence frequencies of Type N and W occupied 59% of the whole airmass types during the observational period). By the effects of monsoon, there are significant monthly variations in number of days for different pathway types.

### 4. Results

The average concentrations of PMs and carbonaceous aerosols for each airmass were listed in Table 3. In order to provide the quantitative intuition of health effects of the each airmass type, Air Quality Category (AQC) suggested by US Environmental Protection Agency (USEPA) of the each airmass type were also shown. The mean PM10, PM25, BC, OC, and EC concentrations of type W were 66.32, 54.76, 3.82, 5.72, and 3.44  $\mu$ g m<sup>-3</sup>, respectively, which are noticeably higher than those of type C (52.37, 43.77, 3.53, 4.79, and 2.92  $\mu$ g m<sup>-3</sup> for PM<sub>10</sub>, PM<sub>2.5</sub>, BC, OC, and EC, respectively). Pope et al. (2002) reported that 10 µg m<sup>-3</sup> increase of long-term average PM<sub>25</sub> was associated with about 8% increased risk of lung cancer mortality. The PM<sub>2.5</sub> concentration of type W was higher than that of type C by 10.99  $\mu$ g m<sup>-3</sup>. This value possibly increases the relative risk (RR) of lung cancer mortality ratio by about 0.08 according to the Pope et al. (2002). The mean concentrations of EC, BC, PM2,5 of type N (2.85, 2.95, and 37.78  $\mu$ g m<sup>-3</sup>, respectively) were lower than those of type C (2.92, 3.53, and 43.77  $\mu$ g m<sup>-3</sup>, respectively) while OC concentration of type N (5.75  $\mu$ g m<sup>-3</sup>) was higher than that of type C  $(4.79 \ \mu g \ m^{-3})$ . The coarse mass concentrations of type N and type W (12.68 and 11.56  $\mu$ g m<sup>-3</sup>, respectively)

Table 2. AQI and its corresponding 24 hourly mean PM<sub>2.5</sub> (µg m<sup>-3</sup>) and Air Quality Category (AQC).

	AQI								
	0-50	51-100	101-150	151-200	201-300	301-400	401-500		
PM <sub>2.5</sub>	0-15.4	15.5-40.4	40.5-65.4	65.5-150.4	150.5-250.4	250.5-350.4	350.5-500.4		
AQC	Good	Moderate	USP	Unhealthy	Very Unhealthy	Hazardous	Hazardous		

USP denotes unhealthy conditions for special groups such as elderly and children.

		BC	OC	EC	$PM_{10}$	PM <sub>2.5</sub>	Coarse	OC/EC	N <sup>a</sup>	AQC <sup>b</sup>
Туре С	AVG <sup>e</sup> STD <sup>d</sup> CNT <sup>e</sup>	3.53 2.11 1829	4.79 2.84 1361	2.92 1.72 1359	52.37 32.93 1517	43.77 29.62 1517	8.60 7.34 1416	1.64	78	USP
Type E	AVG STD CNT	2.47 1.85 520	3.86 2.87 384	2.23 1.44 380	28.27 25.97 515	22.46 22.09 515	5.81 6.42 515	1.73	41	Moderate
Type S	AVG STD CNT	2.88 1.62 733	3.24 2.24 637	2.38 1.34 636	37.65 29.90 719	32.55 26.71 719	5.12 5.06 409	1.36	32	Moderate
Type N	AVG STD CNT	2.95 2.01 2817	5.75 2.78 2133	2.85 1.82 2133	50.47 27.81 2695	37.78 24.02 2695	12.68 9.91 2646	2.02	120	Moderate
Type W	AVG STD CNT	3.82 2.32 2058	5.72 4.89 1798	3.44 2.26 1798	66.32 35.44 2094	54.76 31.83 2094	11.56 7.75 2020	1.66	89	USP
Type N Asian dust	AVG STD CNT	3.29 2.15 96	6.67 3.48 83	3.27 2.13 83	130.06 35.26 92	56.32 23.94 92	73.74 26.86 92	2.04	3	USP
Type W Asian dust	AVG STD CNT	3.61 1.85 83	4.12 1.70 65	3.46 1.49 65	203.52 86.73 82	106.24 46.82 82	97.28 62.04 82	1.19	3	Unhealthy

**Table 3.** Average aerosol concentrations of each airmass pathway type ( $\mu g m^{-3}$ ).

<sup>a</sup>Number of days for each type.

<sup>b</sup>Air quality category.

<sup>c</sup>Average.

<sup>d</sup>Standard deviation.

<sup>e</sup>Total number of data (sampled with 1 hr resolution).

were relatively higher than those of type C, type E, and type S (8.60, 5.81, and 5.12  $\mu$ g m<sup>-3</sup>, respectively). Both the aerosol concentrations and corresponding number of days of type E and type S were lower than those of other type of airmasses. The AQCs of type W and type C were USP (unhealthy conditions for special groups such as elderly and children) while those of type E, type S, type N were at moderate level.

There were 6 days of dust events during the observational period. The airmass types of dust events on 8th May 2007, 12th Feb, and 16th Mar 2008 were type N and that on 25th May, and 29th Dec 2007 were type W. During the dust events, the coarse mass concentrations of the both airmass types increased significantly (73.74 and 97.28  $\mu$ g m<sup>-3</sup> for type N and type W, respectively). Especially when dust came from west, PM<sub>2.5</sub> (106.24  $\mu$ g m<sup>-3</sup>) increased significantly since the airmass passed over the large emission source areas

of the aerosols in East China with the strong wind advection. AQC during the dust event of type N and type W increased from Moderate to USP and from USP to Unhealthy, respectively. However, carbonaceous aerosol concentrations of type N and type W during the dust event showed comparable amounts with the corresponding type of airmasses without dust.

Temporal variations of PM and carbonaceous aerosol concentrations are shown with the temporal variations of airmass type in Fig. 2. The Airmasses of the type N, type W, and type C dominated throughout the measurement period except summer. In summer (from June to September), the type E and type S were dominant during the period. Coarse mass increased in type W and type N, while  $PM_{2.5}$  increased in type W and C. The high  $PM_{2.5}$  concentrations over 100 µg m<sup>-3</sup> appeared frequently in type W. Low aerosol concentrations appeared in type E and type S. When the dust events appeared, coarse mass concentration



**Fig. 2.** Temporal variations of (a)  $PM_s$  (gray :  $PM_{2.5}$ ; dark gray: Coarse mass) and (b) carbonaceous aerosols (gray: OC; dark gray: EC) during the measurement period. Background colors represent the airmass type (yellow: type W; blue: type C; green: type N; white: type E and type S; red: Dust event).

increased over 100  $\mu$ g m<sup>-3</sup> as shown in Fig. 2. In Fig. 2, high EC concentration appeared in type W and type C, while high OC concentration appeared in type W and type N. It was found that the temporal variations of aerosol concentrations in Seoul were strongly affected by the seasonal variations of airmass pathways.

To evaluate the significance of the differences between average values of each airmass pathway type, T-test of independent sample has been performed using SPSS (Statistical Package for Social Sciences) version 12.0 K. The calculated P-values (intercorrelation coefficients) are listed in Table 4. The difference of OC concentrations between type N and type W, that of BC concentrations between type S and type N, that of EC concentrations between type N and type C, and that of  $PM_{10}$  concentrations between type N and type C were identified to be meaningless with respect to the significance level of 0.05. The differences of the other species between different types of airmasses were identified to be meaningful with respect to the significance level of 0.05.

### 5. Summary and conclusions

In order to quantify the effects of the long range transport from remote sources on aerosol concentrations in Seoul, the airmasses were classified into the five types with respect to their pathways. The  $PM_{10}$ ,  $PM_{2.5}$ , BC, OC, and EC samples were collected at Yonsei University site during the period from April 2007 to March 2008. The aerosol concentrations of airmasses

BC	OC										
EC	$\mathbf{PM}_{10}$	Type C		Type W		Type S		Type N		Type E	
PM <sub>2.5</sub>	CM <sup>a</sup>										
Туре С				0	0	0	0	0	0	0	0
				0	0	0	0	0.232 <sup>b</sup>	$0.057^{b}$	0	0
				0	0	0	0	0	0	0	0
Type W		0	0			0	0	0	0.822 <sup>b</sup>	0	0
		0	0			0	0	0	0	0	0
		0	0			0	0	0	0	0	0
		0	0	0	0			0.324 <sup>b</sup>	0	0	0
Тур	be S	0	0	0	0			0	0	0.046	0
		0	0	0	0			0	0	0	0.016
		0	0	0	0.822 <sup>b</sup>	0.324 <sup>b</sup>	0			0	0
Type N	0.232 <sup>b</sup>	$0.057^{b}$	0	0	0	0			0	0	
		0	0	0	0	0	0			0	0
		0	0	0	0	0	0	0	0		
Type E	e E	0	0	0	0	0.046	0	0	0		
		0	0	0	0	0	0.016	0	0		

Table 4. P-values between the each aerosol species of different airmass pathway types.

<sup>a</sup>Coarse mas.

<sup>b</sup>Difference between the average values is not significant where significance level is 0.05.

of type W (PM<sub>10</sub>: 66.32  $\mu$ g m<sup>-3</sup>, PM<sub>2.5</sub>: 54.76  $\mu$ g m<sup>-3</sup>, BC: 3.82 μg m<sup>-3</sup>, OC: 5.72 μg m<sup>-3</sup>, EC: 3.44 μg m<sup>-3</sup>) were even higher than those of type C ( $PM_{10}$ : 52.37  $\mu g m^{-3}$ , PM<sub>2.5</sub>: 43.77  $\mu g m^{-3}$ , BC: 3.53  $\mu g m^{-3}$ , OC: 4.79  $\mu$ g m<sup>-3</sup>, EC: 2.92  $\mu$ g m<sup>-3</sup>). The EC, BC, and PM<sub>2.5</sub> concentrations of type N were 2.85, 2.95, and  $37.78 \ \mu g m^{-3}$ , respectively, which were lower than those of type C (2.92, 3.53, and 43.77  $\mu$ g m<sup>-3</sup> for EC, BC, and PM<sub>2.5</sub>, respectively). Meanwhile, the OC concentration of type N (5.75  $\mu$ g m<sup>-3</sup>) was higher than that of type C (4.79  $\mu$ g m<sup>-3</sup>). Both the coarse mass and PM2.5 concentrations increased significantly during the dust events due to the large amount of transported aerosols with strong wind advection. The temporal variations of aerosol concentrations in Seoul were found to be affected by the seasonal variations of airmass pathways. Especially high PM<sub>2.5</sub> concentrations over 100 µg m<sup>-3</sup> in Seoul were appeared most frequently when the airmasses came from west.

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