

The Distribution Characteristics and Long-term Trend of Carbonaceous Species in Airborne Particulate in Seoul between 1986 and 1996

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Abstract

To characterize airborne particulate carbon and its temporal variation in the heavily industrialized metropolitan city, Seoul in South Korea, aerosol sampling was performed from 1986 to 1996. Correlation coefficients of elemental carbon (EC) and organic carbon (OC) with mass concentration of fine particles ($\leq 2.1 \mu\text{m}$) are 0.73 and 0.51, respectively. EC concentrations of the fine particle mode are 10.1, 5.9, 4.5, and $7.4 \mu\text{g m}^{-3}$ in winter, spring, summer, and autumn, respectively. On the other hand, OC concentration shows maximum value in winter and followed by autumn, summer, and spring. A seasonal peak in the ratio of OC to EC in fine particles was observed during the summer photochemical season from June to August. Concentrations of EC and OC in Asian dust storm events are generally higher than in non-Asian dust storm events except in 1990. The difference of EC concentrations between Asian dust storm periods and non-Asian dust storm periods are much larger than those of OC concentrations. There are slight increases of EC concentration between 1987 and 1990 and a gradual decrease between 1990 and 1996.

Key words : Elemental carbon, Organic carbon, Asian dust storm, Long-term trend

1. INTRODUCTION

It is generally accepted that carbonaceous material is a principal component of airborne particulate matter, with carbon responsible for 10~20% of aerosol mass (Shah *et al.*, 1986; Malissa *et al.*, 1984). Carbonaceous aerosol is mainly a combustion aerosol of primary and

secondary origin. Its shape, size, and origins are extremely variable depending on the different type of fuels, combustion processes, and the age and history of air masses. It mainly resides in the submicron particle size range (Liousse *et al.*, 1993; Clarke *et al.*, 1987). This aerosol is usually separated into two main components: one is called the elemental carbon (EC) and the other the organic carbon (OC) component. EC (the optically absorbing component of soot and smoke) is spread throughout the world's atmosphere. It is produced during the incomplete combustion of various fuels, with

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the most important sources being fossil fuel combustion and biomass burning (Ghan and Penner, 1992; Turco *et al.*, 1983). The increasing demand for energy in the past century has led to a great increase in the rate of EC emissions. EC particles are characterized by chemical inertness, large surface area, and low density. Because of its high optical absorption, an increase in EC may affect the solar radiation balance, surface temperature and visibility. Also, EC has a strong ability to absorb SO₂ gas, which can be catalytically converted to sulfate to enhance EC cloud condensation activity. Although fresh soot is mostly hydrophobic, aged EC-containing aerosols become more hygroscopic by surface reactions and mixing processes (Weingartner *et al.*, 1997; Chughtai *et al.*, 1996; Lioussé *et al.*, 1993). This enables the transfer of EC into cloud droplets via nucleation scavenging (Hitzenberger *et al.*, 2001). Although the effect of increasing EC in the atmosphere may be considerable, knowledge of EC concentrations, distributions, characteristics, and potential effects is still seriously lacking. Organic particulate carbon is both emitted in primary particulate form, and formed in the atmosphere through photochemical reactions. Low-volatility products from the gas-phase oxidation of reaction organic gases condense or absorb onto particle surfaces, or absorb into pre-existing particulate matter resulting in the addition of material to atmospheric particles. This is termed "secondary organic aerosol formation". Both biogenic and anthropogenic sources contribute to primary and secondary organic particulate matter (Hildemann *et al.*, 1996; Grosjean, 1992).

Until now, several studies on airborne particulate carbon have been carried out in urban and remote areas of South Korea (Park *et al.*, 2001; Kim *et al.*, 2000; Kim *et al.*, 1999). However, only a few measurements of the long-term variation of carbonaceous species have so far been made at heavily industrialized areas in South Korea.

The primary goal of this study is to report the characteristics of airborne particulate carbon and its long-term variation in a heavily industrialized metropolitan

city in South Korea. The investigation of long-term variation air pollution plays a considerable part in the regulation of emission sources and environmental policies. From this point of view, the information reported in this work showed be helpful to produce further emission source control strategies in Seoul, Korea.

2. MATERIAL AND METHODS

2.1 Location

Seoul, the capital city of South Korea, is located in the central western part of the Korean Peninsula. Seoul has a population of 10.6 million people over an area of 605 km². The annual average temperature is 11.1 degrees Celsius (°C) in Seoul but its monthly temperature varies according to season. For instance, it may reduce to as low as minus 4.9°C in January and rise to 33.4°C in August. Annual rainfall is relatively high at 1,295 mm but it rains heavily in the summer from June to September but very little in the winter. As a major source of air pollution, about 2.4 million motor vehicles are in Seoul. The delta area forming the Seoul-Inchon-Suwon triangle is the largest industrial zone in South Korea with several satellite cities such as Anyang, Puchon and Uijonbu.

Sampling was carried out on the roof of a five-story building (15 m) of Konkuk University (37° 34'N; 126° 58'E) in Seoul, Korea. The surroundings of this urban sampling site are residential and commercial areas with some minor point sources such as a textile mill, electronic manufactories, and print shops. There are no nearby structures taller than the sampling site. Six-lane roads, usually with heavy traffic, are located 0.1 km all around the sampling site.

2.2 Instrumentation

A high volume air sampler was operated three times for 24 h per month with a flow rate of 1.2 m³ min⁻¹ from 1986 to 1996. Whatman 8 × 10 in. quartz micro fiber filters were used for the sampling of total suspended particulate (TSP) (≤ 100 μm). By using this high

volume air sampler, 372 TSP samples were collected during the whole of sampling period. A 4-stage Andersen air sampler was also used for the sampling of aerosols as a function of their size. Aerosol particles were collected onto a 80 mm diameter quartz fiber filter (Whatman) at each stage. Sampling duration time was 72 h and the airflow was maintained at approximately 23.8 l min^{-1} . The 50% cut-off sizes of each stage for the Andersen air sampler are 7, 2.1, 0.43, $0.08 \mu\text{m}$ from the 1st- to 4th stage, respectively.

In this work, quartz fiber filters were pretreated by heating in an electric furnace at 800°C in air for 1 h to remove carbonaceous contaminants. Each pretreated quartz fiber filter was placed in a clean polyethylene bag. All bags were refrigerated until field measurement after sealing and wrapping with aluminum foil.

Quartz fiber filters are often used for the measurement of carbonaceous species due to their suitability for thermal carbon analysis and extraction. However, they easily adsorb gas-phase organic species because of their large specific surface area (Turpin *et al.*, 2000). This adsorption of gas-phase organic species onto the filter during sampling and handling processes cause positive interferences. On the contrary, negative artifacts caused by the volatilization of organics from the particle sample is possible (Turpin *et al.*, 2000). In this study, several pre-heated blank filters were handled in the same manner as the sampling filter during every process of sampling, weighing, and analysis. The OC content of blank filters were used to calculate the positive artifacts, in the analysis.

2.3 Analysis

It is important to try to distinguish between organic and elemental carbon because of their different origins and different atmospheric chemical and optical properties. Elemental carbon may be first considered as thermally refractory: therefore, thermal analyses are based on the separation of the two main constituents of carbonaceous aerosols, the organic and the elemental carbon fraction; the organic matter volatilizing first during a heat treatment (Hansen and Novakov, 1990).

For the analysis of total carbon (TC) and EC, an Elemental Analysis System (EA1108, Fisons Co.) was used in this work. As one of the carbon analysis techniques with Thermal Manganese dioxide Oxidation technique (Fung, 1990), Thermal-Optical Reflectance Method (Chow *et al.*, 1993b), and Integrating Sphere Method (Hitzenberger *et al.*, 1996), this Elemental Analysis System has been usefully applied since it was introduced by Cachier *et al.* (1989) (Lin and Tai, 2001; Ma *et al.*, 2001b). However it is difficult to compare data on carbonaceous species measured by different techniques, because no standard method yet exists. This Elemental Analysis System combined with a high degree of analytical accuracy and a recovery rate of nearly one hundred percent enables a speedy quantitative analysis of C, H, N and S simultaneously from the sample filter. Digested sample filters in the combustion bath were detected by a multiple detector which digitizes the analogue amplification of the signal via a voltage-to-frequency converter. At the start of the analysis run, a standard test substance was analyzed to provide data on the functioning of the Elemental Analysis System, and to ensure the detector was working properly. Calibration was performed by this analysis of the standard test substance. Standard deviation is $<0.1\%$ abs. with a $4\sim 5 \text{ mg}$ carbon test substance. Recovery rate is $>99.5\%$ for the standard test substance. After sampling, several parts punched out from the sample filter were heated in a 350°C electric furnace for 1 h to exclude the OC content. Then these pretreated parts and other parts of the original sample filters were fed into the dynamic flash combustion chamber of the Elemental Analysis System to obtain the EC and TC contents, respectively. When the sample filter enters into combustion chamber, a fixed amount (45 ml min^{-1}) of oxygen is injected into the helium carrier gas, which is flowing through the combustion reactor. The OC concentration was calculated from the difference between TC and EC concentrations. The more detailed analytical procedures and experimental set-ups of the Elemental Analysis System are described elsewhere (Ma *et al.*, 2001b).

3. RESULTS AND DISCUSSIONS

It is a debatable point if the quartz micro fiber filter used for the sampling of TSP has uniformity of aerosol mass collected on the filter surface. It is necessary to examine this because we analyzed only several parts of each filter. To solve this problem, we punched out 100 parts from different portions of a sample filter, then these 100 discs were analyzed under the same analytical conditions. As a result, the average TC concentration of 100 discs was $25.4 \mu\text{g m}^{-3}$ with pretty low standard deviation ($0.31 \mu\text{g m}^{-3}$). The results show that we could not find any significant difference in concentration among discs punched out from a sample filter except of the edge portion of the sample filter.

Relationships between EC concentration and mass concentration of fine particles ($\leq 2.1 \mu\text{m}$, sum of the 3rd and 4th stage of the Andersen air sampler), OC concentration and mass concentration of fine particles, and OC concentration and EC concentration of fine particles are shown in Figure 1. Correlation coefficients of EC and OC to mass concentrations of fine particles are 0.73 and 0.51, respectively.

In the polluted urban area, carbonaceous species contributes approximately 10~20 wt% of TSP and 40 wt% of fine particles (Gray *et al.*, 1986). In a heavily polluted atmosphere, carbonaceous species contribute up to 50% of the fine particle mass (Chow *et al.*, 1993a; Gray *et al.*, 1986). In Seoul, OC and EC contribute 23 wt% and 19 wt% of $\text{PM}_{2.5}$ (Park *et al.*, 2001). And Heintzenberg (1989) reported that in remote areas, OC and EC contribute about 11 wt% and 0.3 wt% of mass of fine particles, respectively. Generally, high correlations between EC and fine particle, and OC and fine particles are usually found because EC and OC are major components of fine particles. According to Lee and Kang's study (2001), correlations between EC and $\text{PM}_{2.5}$, and OC and $\text{PM}_{2.5}$ were 0.64 and 0.63, respectively at other urban areas of South Korea. Comparing to Lee and Kang's (2001) result, poor correlation between OC and fine particles was found in this study.

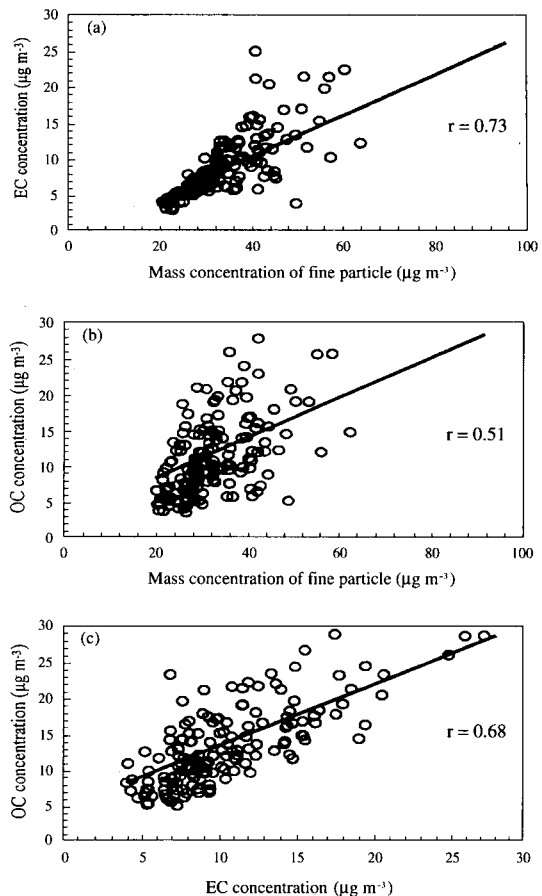


Fig. 1. Relationships between EC mass concentration ($\mu\text{g m}^{-3}$) and mass concentration of fine particles (a), OC concentration and mass concentration of fine particles (b), and OC concentration and EC concentration (c).

Particulate OC lost due to volatilization (i.e. negative artifacts) and adsorption of gas-phase organics onto the filter (i.e. positive artifact) during and/or after sampling are possible reasons for poor correlation between OC and fine particles. It is difficult to accurately measure particulate OC concentrations due largely to interference from gaseous and condensable organics. Sampling errors related to the interaction of semi-volatile and volatile organics with the filter substrate are caused (Turpin *et al.*, 2000). Eatough *et al.* (1990) reported that estimates of artifact contributions to OC

Table 1. Seasonal variation of EC and OC mass concentrations in fine and coarse mode particles, OC/EC ratio in fine mode particles, and meteorological data in Seoul.

Season		EC ($\mu\text{g}/\text{m}^{-3}$)			OC ($\mu\text{g}/\text{m}^{-3}$)			OC/EC in fine	Average temperature ($^{\circ}\text{C}$)	Average R.H. (%)
		Fine ^a	Coarse ^b	Fine/Coarse	Fine	Coarse	Fine/Coarse			
Winter (n = 20) ^c	Average	10.1	5.2	1.9	12.6	4.7	2.7	1.2	-2.6	65.2
	S.D. ^d	1.2	0.6		2.7	0.9				
Spring (n = 19)	Average	5.9	3.0	1.9	8.5	3.9	2.2	1.4	13.1	56.1
	S.D.	0.2	0.4		1.3	0.5				
Summer (n = 15)	Average	4.5	2.5	1.8	9.6	3.8	2.5	2.1	27.4	72.4
	S.D.	1.6	0.2		2.4	0.8				
Autumn (n = 18)	Average	7.4	3.7	2.0	9.7	3.4	2.9	1.3	10.2	63.6
	S.D.	1.4	1.2		1.7	0.6				
Annual (n = 72)	Average	6.9	3.6	1.9	10.1	3.9	2.6	1.5	12.0	64.3
	S.D.	1.1	0.6		2.0	0.7				

Winter (December–February), Spring (March–May), Summer (June–August), Autumn (September–November)

^aFine particle ($\leq 2.1 \mu\text{m}$, sum of the 3rd and 4th stage of Andersen air sampler)

^bCoarse particle ($\geq 2.1 \mu\text{m}$, sum of 1st and 2nd stage of Andersen air sampler)

^cNumber of samples

mass measurement range from -80% for volatilization-induced bias to $+50\%$ for adsorption-induced bias.

The origin of carbonaceous particles can be estimated (Turpin and Huntzicker, 1995) based on the relationship between OC and EC. As shown in Figure 1, the correlation coefficient between OC and EC in fine particles is 0.68. Kim *et al.* (1999) reported that relationships between the EC and OC concentration for PM_{2.5} were 0.72 at Seoul and 0.96 at Cheju remote area, respectively. Though a relatively lower relationship than that of Kim's (1999) result, this relatively good correlation suggests that the EC and OC may be emitted from the same sources.

Seasonal variation of EC and OC concentrations in fine and coarse particles, OC/EC ratios of fine particles, and meteorological data are summarized in Table 1. EC concentrations in the fine particle mode were 10.1, 5.9, 4.5, and 7.4 $\mu\text{g m}^{-3}$ in winter, spring, summer, and autumn, respectively. On the other hand, OC concentration shows the maximum value in winter and followed by autumn, summer, and spring. The seasonal variation of OC/EC ratio in fine particles was ranged from 1.2 to 2.1. On the other hand, OC/EC ratios in coarse particles were 0.9, 1.3, 1.5, and 0.9 in winter, spring, summer and fall, respectively. The considerable

increasing of OC/EC ratios in coarse particles in spring is like due to the high input of local biogenic organic aerosols (Gelencser *et al.*, 2000). Though OC/EC ratios in coarse particles marked lower levels than those of fine particles, they exhibit similar seasonal fluctuation with the relatively high ratio in spring and summer and low ratio in fall and winter. A seasonal peak in the ratio of OC to EC in fine particles was observed during the summer photochemical season from June to August, reflecting an increased formation of secondary organic carbon because of more favorable meteorological conditions for the gas/particle conversion of gaseous hydrocarbon precursors as a result of photochemical activity. This could result from the large amount of volatile organic compounds emitted from various sources in heavily industrialized Seoul.

In order to identify secondary OC, an equation was introduced from total OC and OC/EC primary ratio by Turpin and Huntzicker (1995), and also Castro *et al.* (1999) suggested an equation with an assumption that particle samples with minimum OC/EC ratios contain almost exclusively primary carbonaceous compounds. However, generally a value of 2.0 for the ratio of OC-EC has been used to identify the presence of secondary organic aerosols formed in the atmosphere through

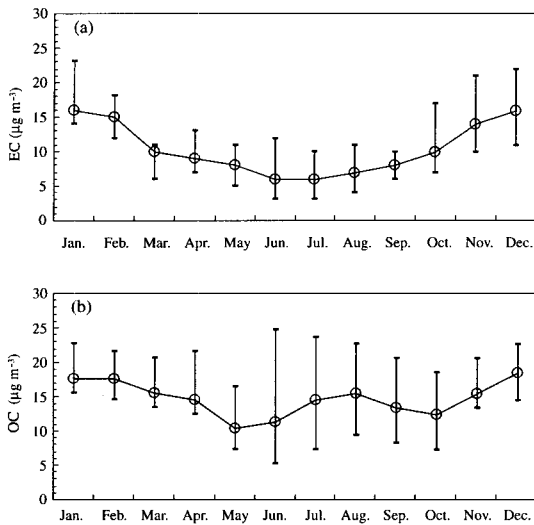


Fig. 2. Monthly average mass concentrations ($\mu\text{g m}^{-3}$) of EC (a) and OC (b) in TSP.

volatile organic compound gas-to-particle conversion processes (Chow *et al.*, 1994). In this study, high OC-EC correlation and less than 2.0 OC/EC ratios except for summer time may be evidence that OC is considered to be emitted from primary anthropogenic sources along with EC.

The summertime OC/EC ratio of 2.1 in fine particles measured in this study is comparable to those observed at urban sites in other countries. Wolff *et al.* (1991) performed measurements in Claremont, USA and found OC/EC ratio of 3.9. They also reported that OC/EC ratios in Long Beach, USA was 2.4. Furthermore, Miura *et al.* (2000) and Kadowaki (1990) carried out the measurements of OC/EC ratios in Japan showing 1.4 in Tokyo and 1.2 in Nagoya, respectively. We found that OC/EC ratio obtained in this study was relatively low compared to those reported at other urban sites in USA, whereas the summertime OC/EC ratio of 2.1 in this study showed higher level than those of two urban sites in Japan.

Figure 2 shows the monthly average concentrations of EC (a) and OC (b) in TSP. EC concentration is relatively low and constant from April to September. The

average concentration during these periods was approximately $8 \mu\text{g m}^{-3}$. Starting in October, however, higher EC concentrations are shown. The maximum EC concentration reached nearly $23 \mu\text{g m}^{-3}$ in January. During the entire winter, EC concentrations greater than $20 \mu\text{g m}^{-3}$ were frequently measured. It is evident that the period of high EC concentration corresponds to lower temperatures, suggesting that space heating is a significant source of ambient EC during cold periods. Stagnant air conditions occurring during cold days would, of course, result in maximum EC concentration (Dod *et al.*, 1986).

The washout of EC by more frequent rain event in summer will be one of the possible causes of lower values in the summer. In Seoul, the average number of rainy days per season is 28 days in summer and 7 days in winter, respectively. Ogren *et al.* (1984) and Harrison *et al.* (1997) reported that rainfall scavenging could be an efficient sink for EC aerosols. The removal efficiency of EC by precipitation is expected to vary widely. In general, hydrophilic particles are more easily scavenged because they are able to act as cloud concentration nuclei (CCN) (Schumann *et al.*, 1988). EC, if present as pure graphite, is hydrophobic. However, after aging most EC should become internally mixed with hydrophilic aerosol components. These EC-containing particles would act as CCN and undergo removal if precipitation develops. Even though the fluctuating OC concentration has not shown the same monthly variation as EC concentration, lower OC concentration occurred from May to June. The average concentration of OC during these periods was approximately $10.5 \mu\text{g m}^{-3}$. Likens *et al.* (1983) reported that wet deposition of organic carbon was higher in summer and lower in winter at Ithaca, NY and Hubbard Brook, NH, USA. The concentration of OC in wet-only deposition in Sweden, W Ontario, and NE United States is reported to range from 1.2 to 2.4 mg C l^{-1} . The monthly maximum in OC concentration was observed during the photochemical season from June to July. This summertime peak of OC concentration is probably reflecting the enhanced secondary organic aerosol generation

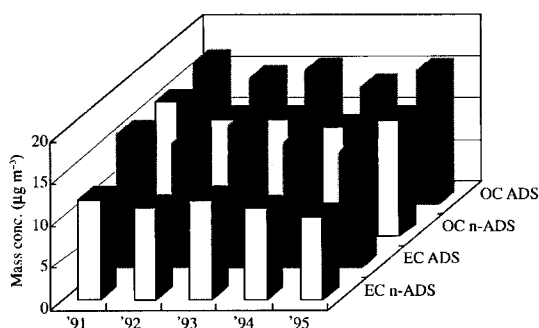


Fig. 3. Yearly variation of average carbon mass concentration ($\mu\text{g m}^{-3}$) during Asian dust storm events and non-Asian dust storm periods. ADS and n-ADS mean Asian dust storm and non-Asian dust storm, respectively.

because of more favorable conditions for the gas to particle conversion of VOCs as a result of photochemical episodes.

EC particles are characterized by chemical inertness, large surface area, and low density. They may have long residence times in the free troposphere and can be transported over great distances. Even at the most remote locations such as the South Pole, detectable amounts of EC have been measured (Hansen and Novakov, 1988). Wolff *et al.* (1982) attributed higher than average elemental carbon concentration ($2.5 \mu\text{g m}^{-3}$) for air samples collected on the Louisiana Gulf coast to the long-range transport of emissions from the midwestern and northeastern United States as compared to a concentration of $1.2 \mu\text{g m}^{-3}$ obtained for samples collected from maritime tropical air masses originating in the Gulf.

Asian dust storm (ADS) is a serious and growing environmental problem in East Asia as well as the Pacific Basin. This dust is blown up by strong winds behind cyclones and delivered up to the free troposphere by westerly jet. Even though ADS are a peculiar phenomenon occurring in the China continent, these ADS have led to significant environmental changes in East Asia and North Pacific Ocean for a long time (Ma *et al.*, 2001a). To investigate the effect of Asian dust storms on particulate carbon concentration, aerosol

samplings were performed using a high volume air sampler during Asian dust storm events from 1991 to 1995. Figure 3 shows the yearly variation of mean carbon concentration during Asian dust storm and non-Asian dust storm periods. Here, non-Asian dust storm period means the springtime when Asian dust storm event was not occurred. Concentrations of EC and OC in Asian dust storm events are higher than in non-Asian dust storm periods. The differences in EC concentration between Asian dust storm periods and non-Asian dust storm periods are much larger than those of OC concentrations. Although more detailed information on the source profiles and meteorological conditions are desired, the probable explanation of this reason is Asian dust storm streamed through the industrialized area.

Parungo *et al.* (1994) reported that the major source of EC over the Yellow Sea (between Korea and China) and the Western Pacific Ocean is probably anthropogenic air pollution from China. One can estimate that the average horizontal EC flux from China continent to the atmosphere over the nearby sea is $5 \mu\text{g m}^{-2} \text{ s}^{-1}$, $0.45 \text{ g m}^{-2} \text{ d}^{-1}$ and $165 \text{ g m}^{-2} \text{ yr}^{-1}$. Taking the China coastline as approximately 2500 km in length and coastal boundary layer height as about 1000 m, Parungo *et al.* (1994) estimate an annual export of EC at $4 \times 10^{11} \text{ g}$. Also, recently Ro *et al.* (2001) characterized the Asian dust deposited with rainwater in Seoul. According to their study, even though the carbonaceous particles could not be classified into elemental, organic, and biogenic particles using their low-Z EPMA technique, carbonaceous particles were observed in various types, e.g. as a single species (4.0%), and mixtures with aluminosilicates (36%), SiO_2 (3.4%), and CaCO_3 (2.4%). Their results also suggest that the carbonaceous species should be considered as one of the important chemical species in Asian dust particles.

As mentioned earlier, the investigation of the long-term variation of particulate carbon concentration is one of the important steps required to not only model the atmospheric consequences of combustion particles but also regulate the emission sources. Figure 4 shows

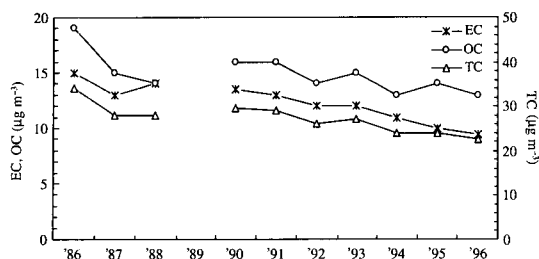


Fig. 4. Average mass concentration ($\mu\text{g m}^{-3}$) of particulate carbon from 1986 to 1996 in Seoul, Korea.

the average mass concentration of particulate carbon from 1986 to 1996 in Seoul, Korea. Owing to a small sample number, data in 1989 were excluded. There are slight increases of EC concentration between 1987 and 1990 and a gradual decrease between 1990 and 1996. Before the 24th Seoul Olympic games in 1988, traffic was regulated and the number of vehicles on Seoul city streets was very low. The slight decrease of EC concentration in 1987 shows the effect of the reduction of traffic on the concentration of particulate EC in the city center. When the Seoul Olympic games were finished, both traffic and air pollution began to return to normal in the city. With regard to the above results, we would like to point out that the regulation of traffic transportation may give a temporary positive effect on the environment. The slight decrease of EC concentration from 1990 to 1996 is caused by the rigorous emission regulation of diesel vehicles including large-sized buses and a campaign of clean energy supply in this period.

4. CONCLUSIONS

Characteristics of airborne particulate carbon, its long-term variation, and the effect of Asian dust storms on particulate carbon levels were studied in Seoul, Korea. Correlation coefficients between EC and mass of fine particles ($\leq 2.1 \mu\text{m}$), OC and mass of fine particles, and OC and EC are 0.73, 0.51 and 0.68 respectively. EC concentrations in fine particles are 10.1, 5.9, 4.5, and $7.4 \mu\text{g m}^{-3}$ in winter, spring, summer, and

autumn, respectively. On the other hand, OC concentration shows a maximum value in winter and followed by autumn, summer, and spring. A seasonal peak in the ratio of OC to EC in fine particles was observed during the summer photochemical season from June to August, reflecting an increased formation of secondary organic carbon because of more favorable meteorological conditions for the gas/particle conversion of gaseous hydrocarbon precursors as a result of photochemical activity. Both EC and OC concentrations in Asian dust storm events are higher than in non-Asian dust storm periods. Rigorous emission regulation of diesel vehicles, and a campaign of clean energy supply has caused a gradual decrease in EC mass concentration between 1990 and 1996 in Seoul, Korea.

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