

Scavenging Properties of Atmospheric Carbon by Precipitation

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Abstract

In order to investigate the scavenging property of airborne carbonaceous particles by precipitations, rainwater, snow sample, and total suspended particulate matter (TSP) were collected at a heavily industrialized urban site. Elemental carbon (EC) contents of both rainwater and snow water were determined using elemental analysis system. EC concentrations in rain samples varied from 33.6 to 166.6 $\mu\text{g L}^{-1}$ with an average 47.2 $\mu\text{g L}^{-1}$. On the other hand, those of snow samples in three times snow events were ranged from 122.4 to 293.3 $\mu\text{g L}^{-1}$. As might be expected, EC showed the significantly high scavenging rate at the initial rainfall. The average total carbon (TC) scavenging rate by washout mechanisms was 57.6% for five rainfall events. The scavenging rate of EC gradually increased in proportion to the increasing rainfall intensity and rainfall amount.

Key words : Elemental carbon, Total carbon, Rainfall, Snowfall, Wet scavenging

1. INTRODUCTION

The increasing demand for energy in the past century has led to a great increase in the rate of elemental carbon (EC or black carbon (BC): when soot particles affect the solar radiation balance) emissions. It is produced during the incomplete combustion of various fuels, with the most important sources being fossil fuel combustion and biomass burning (Ghan and Penner, 1992; Turco *et al.*, 1983). EC is characterized by chemical inertness, large surface area, and low density. Because of its high optical absorption, an increase in BC may affect the solar radiation balance, surface tempera-

ture and visibility. Also EC has a strong ability to absorb SO_2 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 hydroscopic by surface reactions and mixing processes (Weingartner *et al.*, 1997; Chughtai *et al.*, 1996). This enables the transfer of EC into cloud droplets via nucleation scavenging (Hitzenberger *et al.*, 1999; Hallett *et al.*, 1989).

When aerosol particles are activated and form cloud droplets, both the optically scattering and absorbing properties of the cloud are influenced by the aerosol. If BC is present in cloud droplets, the net radiative effect of the cloud could change from negative to positive. In a recent report by Chýlek *et al.* (1996), EC concentration was ranged from 1.2×10^{-4}

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mg L⁻¹ to 8 mg L⁻¹ in cloud water. This nucleation scavenging is one of the most important processes governing the atmospheric lifetime of aerosol particles including EC. Aerosol nucleation properties, i.e. the characteristic of particles to form water droplets at atmospheric supersaturation levels, are determined by the particle size, chemical composition, and surface characteristics (Winkler, 1988). The nucleation properties of water-soluble aerosols, such as sulfates, are well documented. However much less is known about water vapor nucleation on carbonaceous particles. Once emitted into the atmosphere, carbonaceous particles will undergo several aging processes. The hygroscopicity of aerosol particles has an important influence on their residence time in the atmosphere as well as on their optical properties. The absorption of solar radiation by soot particles is increased if the particles are incorporated into cloud droplets (Chýlek and Marlow, 1995). This process leads to a decrease of the cloud albedo. In contrast, if soot particles act as cloud condensation nuclei (CCN) (i.e. act as nuclei for CCN) they can increase the cloud albedo by changing the droplets size distribution towards smaller droplets (Tomey *et al.*, 1984). Soot particles that are effective CCN will therefore have shorter atmospheric lifetimes than those unaffected by nucleation. Furthermore added to this CCN scavenging, the large amount of soot particles suspended in the air can be removed by precipitation scavenging mechanisms.

As an obvious manifestation of the degree of atmospheric pollution by soot particles, Malissa and Pell (1989) report the "black snow" episode occurred in the Salzburg region of Austria. The black coloration of the snow was caused by the presence of soot particles. As the results of their experiment about black snow, carbon concentration was found to be 30% of solid fraction of the black snow. Also they said that the carbon content of the black snow was 1000 times higher than that of the "white snow".

Till now, many studies on airborne EC concentration have been carried out in urban and remote areas (Hwang and Ma, 2003; Jennings *et al.*, 1997; Kuhlbusch *et al.*, 1996; Pinnick *et al.*, 1993; Clarke,

1989; Hansen *et al.*, 1988; Andreae *et al.*, 1984; Wolff *et al.*, 1981). However, there are limited data available concerning the EC concentration in rain and snow samples (Ducret and Cachier, 1992; Cachier and Ducret, 1991; Warren and Clarke, 1990; Clarke and Noone, 1985; Ogren and Charleson, 1984). One of reasons for the scarce data on carbonaceous particles in precipitation is probably due to specific problems of collection for carbonaceous material such as bacterial contamination during and after sampling and the sample treatments.

In this study, to determine the scavenging property of atmospheric carbon by precipitation, EC content of both rainwater and snow water as well as TC concentration in ambient particles were determined.

2. MATERIAL AND METHODS

2.1 Sampling

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. The surroundings of this urban sampling site are residential and commercial areas with some minor point sources such as textile mill, electronic appliance shops, and print shops. There are no nearby structures taller than sampling site. Six-lane roads, with usually heavy traffic, are located 0.1 km all around of the sampling site. Rainwater was collected using a glass pluviometer (Tokyo Co., 8065) consisted of reservoir basket, reservoir bottle, and body. Prior to sampling, this pluviometer was cleaned with 1, 1, 1-trichloroethane. Rainwater samples were collected five times from the end of March to the middle of June 1998. Also three snowfall samples were collected using a polycarbonate collection plate in January 1998. Collected fresh snow samples were melted at room temperature. To hinder the growth of biogenic materials like bacteria, algae, or fungi, every sample was refrigerated until analysis.

In addition, to determine the scavenging rate of TC by rainfall, TSP ($\leq 100 \mu\text{m}$) in the atmosphere was sampled before and during five rain events. For

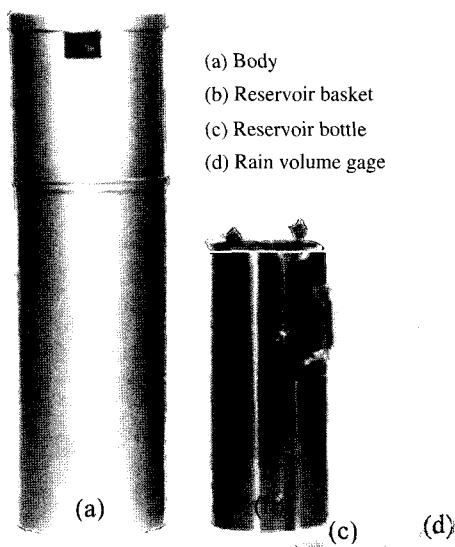


Photo 1. A reservoir type pluviometer (Tokyo Co., 8065)

the sampling of TSP, a high volume air sampler (Kimoto Co., 120F) was operated for 6 hours with $1.2 \text{ m}^3 \text{ min}^{-1}$ flow rate. As the substrate filter, Whatman 8×10 in. quartz micro fiber filters were used. Prior to sampling, to remove carbonaceous contaminants, quartz fiber filters were pretreated by heating in an electric furnace at 800°C for 3 hours. And every preheated quartz fiber filter was placed in clean polyethylene bags. After sealing and wrapping with aluminum foil, all bags were refrigerated until sampling.

2.2 Pretreatments of rain and snow water

Figure 1 shows the pretreatment procedure of liquid sample for the subsequent EC analysis. After agitating, rainwater was passed through a nylon filter (Gelman, Nylasorb) with $5 \mu\text{m}$ pore size, to remove impurities such as leaves, pollens, and insects. It is widely known that carbonaceous aerosols mainly reside in the submicron particle size range (Clarke *et al.*, 1987). Therefore, carbonaceous aerosols can be passed through a nylon filter with $5 \mu\text{m}$ pore size.

A 47 mm diameter quartz fiber filter (Whatman) heated at 800°C was used for the filtration of the pre-filtrated rainwater. To remove OC fraction from

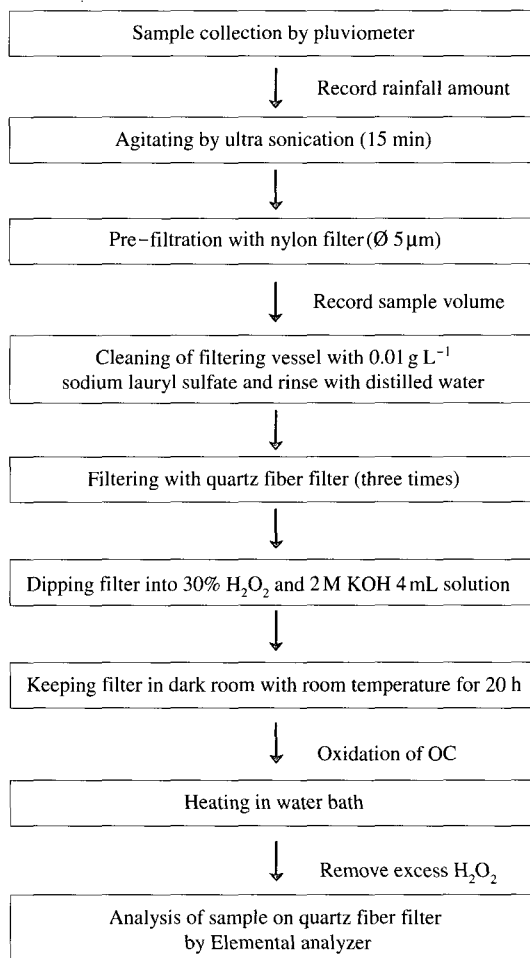


Fig. 1. Pretreatment procedure for the determination of EC in rain and snow waters.

the particles deposited on quartz fiber filter, the oxidation method in a basic peroxide solution introduced by Smith *et al.* (1975) was applied. This direct chemical oxidation method is a non-thermal, ambient pressure, and aqueous-based technology for the oxidation destruction of the organic components. Organic compounds in particles were mostly decomposed in the first 1–2 minutes, and extending reaction time had little effect on further decomposition. The oxidation rate of OC fraction in particles was to be 99.93% (Smith *et al.*, 1975). Whereas, the oxidation method does not cause the removal of EC from

particles. The melted snow water was handled in the same manner as rainwater. After finishing the pretreatment procedure of samples, 6 mm diameter discs were punched out from several portions of quartz fiber filters.

2.3 Analysis

For the analysis of EC and TC, an elemental analysis system (Fisons Co., EA1108) was employed in this work. A schematic diagram of elemental analyzer is illustrated in Figure 2. 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 samples in the combustion bath were detected by multiful 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 the analysis of standard test substance. Standard deviation was $<0.1\%$ abs. with 4–5 mg carbon test substance. Recovery rate was $>99.5\%$ for standard test substance. And then quartz fiber discs were fed into the dynamic flash combustion chamber of elemental analysis system to obtain the EC and TC

contents, respectively. The more detailed analytical procedures and experimental set-ups at elemental analysis system were described elsewhere (Ma *et al.*, 2001b).

3. RESULTS AND DISCUSSION

Quartz fiber filter was used as the filtration substrate for the separation of retained particles from rain and snow waters. However, it is a debatable point that this quartz fiber filter can completely sustain submicrometer particles. To determine the sustainability of quartz fiber filter for submicrometer particles, the artificial carbon particles with $0.2\ \mu\text{m}$ diameter were mixed with distilled water. And then this water including carbon powder was passed through a 47 mm diameter quartz fiber filter. Sustainability was calculated by weighing the carbon powder which was mixed with distilled water and that of deposited on quartz fiber filter after filtration. Consequently, sustainability determined from this method was found to be 95%. This efficiency of filtration was also tested in previous study (Chýlek *et al.*, 1999). They reported that filtration efficiency varied between 71% and 99% with an average value of 88%.

It is also questionable point that the quartz micro fiber filter, which was used as the substrate material

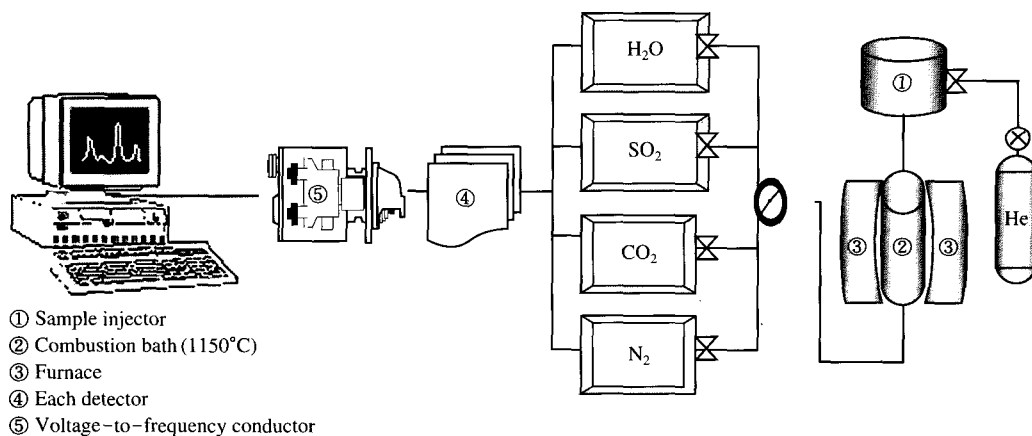


Fig. 2. A schematic diagram of elemental analyzer.

of insoluble fraction in the procedure of liquid sample filtration, has the uniform mass thickness for deposited particles. It is necessary to examine this point because we analyzed only some parts of each filter. To solve this problem we punched out 20 parts from different portions of a quartz fiber filter, then these discs were analyzed under the same analytical conditions. As a result, we could not find any difference of concentration among discs punched out from sample filter with the exception of the edge portion of filter.

3.1 EC in rainwater

Table 1 shows the EC mass concentrations in rainwater collected in five rainfall events. They varied between $33.6 \mu\text{g L}^{-1}$ and $166.6 \mu\text{g L}^{-1}$ with an average $47.2 \mu\text{g L}^{-1}$. These values are comparable to those found in other urban areas. EC concentrations in rainwater collected at an urban location of USA (on the University of Washington campus in Seattle) during the winter of 1980–1981 were ranged from $30 \mu\text{g L}^{-1}$ to $400 \mu\text{g L}^{-1}$ with a median of $60 \mu\text{g L}^{-1}$ (seven samples) (Ogren and Charlson, 1984). And the monthly samples collected at 12 rural sites from April to August, 1981 had a range of $20 \mu\text{g L}^{-1}$ to $600 \mu\text{g L}^{-1}$ with a median of $100 \mu\text{g L}^{-1}$ (58 samples) (Ogren and Charlson, 1984). Other references to EC in rain were summarized in Table 2. To investigate the variation of scavenging rate of EC according to rainfall amount, rain samples were collected during three continuous rainfall events (Apr. 4–5, Apr. 12–13, and Jun. 13–14, 1998). The

scavenging rate of EC at the initial rainfall was calculated by $[\text{EC concentration at initial rainfall} / (\text{Sum of EC concentration at both initial and subsequent rainfalls}) \times 100]$. As shown in Table 1, EC show the significantly high concentrations at the initial rainfall in the all sample. The scavenging rate of EC at initial rainfall was found to be 68.3%, 56.5%, and 78.5% at three rainfall events, respectively. These scavenging rates can be comparable to those in the study of Ma *et al.* (2002). They determined the elemental concentration in 0.35 mm radius raindrops collected at the beginning and subsequent rainfalls, and calculated an average 74% scavenging rate (Elemental concentration in beginning rainfall/The sum of elemental concentration in beginning and subsequent rainfalls \times 100) of eight elements at the beginning rainfall.

3.2 EC in snow

The EC mass concentrations in snow samples collected in three snow events were shown in Table 3. The EC mass concentrations in three times snow events were $122.4 \mu\text{g L}^{-1}$, $151.9 \mu\text{g L}^{-1}$, and $293.3 \mu\text{g L}^{-1}$, respectively. They show significant variation depending on snow events. We note that EC concentration in snow sample is considerably higher than that in rainwater. Recently, a similar result at urban location was reported (Chýlek *et al.*, 1999). Previously reported data (Mitra *et al.*, 1990; Murakami *et al.*, 1981; Knutson *et al.*, 1975;) on the aerosol scavenging by individual snow crystals or flakes indicate that aerosol scavenging is primarily

Table 1. EC mass concentrations in rainwater collected in five rain events.

Sampling period	Rainfall amount (mm)	Rain intensity (mm h^{-1})	Total rainwater (mL)	Filtered rainwater (mL)	EC concentration ($\mu\text{g L}^{-1}$)
Mar. 26, 1998	3.1	0.39	135	135	166.6
Apr. 1–2, 1998	31.8	1.35	801	801	33.6
Apr. 4–5, 1998	19.1	1.17	200 ^a	200 ^a	98.6 ^a
			350 ^b	350 ^b	45.7 ^b
Apr. 12–13, 1998	27.4	1.31	200 ^a	200 ^a	57.6 ^a
			530 ^b	530 ^b	44.4 ^b
Jun. 13–14, 1998	23.7	0.99	200 ^a	200 ^a	98.2 ^a
			580 ^b	580 ^b	26.9 ^b

^a Initial rainfall (2 mm rainfall amount from raining start); ^b Subsequent rainfall

Table 2. Reference data on EC concentrations in rainwater collected at different sites.

		Number of samples	Concentration ($\mu\text{g L}^{-1}$)		Reference
Urban	Fif sur Yvette, France	54	336		Ducret and cachier (1992)
	Detroit, USA	40	160		Orgen and Charlson (1984)
Rural	Michigan, USA	25	72		Orgen and Charlson (1984)
	Sweden	5	100		Orgen and Charlson (1984)
Remote	Mace Head, Ireland	23	27		Ducret and cachier (1992)
Tropical	Lamto, Ivory Coast	62	143		Ducret and cachier (1992)
	Enyele, Congo forest	46	73		Ducret and cachier (1992)

Table 3. EC concentration in snow water collected in three snow events.

Sampling date	Snowfall amount (cm)	Total melted snow amount (mL)	Filtered amount of melted snow (mL)	EC concentration ($\mu\text{g L}^{-1}$)
Jan. 4, 1998	1.9	705	400	122.4
Jan. 8, 1998	3.8	750	370	151.9
Jan. 20, 1998	3.8	710	350	293.3

of the washout type and that snow scavenges aerosol much more efficiently than rain, when based on equal weights of precipitation. Also Gray and Male (1981) reported that because a snowflake falls at a slower speed than a raindrop and sweeps out a larger area, the snowflake will have had a greater exposure to pollutants and therefore would be a better indicator of their presence. In addition, one of the possible reasons for the overwhelmingly high EC concentration in snow water is that snow samples were collected in wintertime. According to the study of Hwang and Ma (2003), during the cold season the airborne EC concentration in Seoul was increased due to domestic and commercial heating. They reported that the range of airborne EC concentrations in the wintertime was between $8.9 \mu\text{g m}^{-3}$ and $11.3 \mu\text{g m}^{-3}$ in fine particles ($\leq 2.1 \mu\text{m}$). This wintertime EC concentration was about two times higher than other seasons.

3. 3 TC scavenging by washout mechanisms

Rainout usually refers to in-cloud scavenging and washout to below-cloud scavenging by falling rain, snow, and so on. Three steps are necessary for wet removal of atmospheric pollutants. Specifically,

Table 4. Scavenging rate of TC by washout mechanism of rainfall.

Sampling period	TC concentration in air ($\mu\text{g m}^{-3}$)		Scavenging rate ^c (%)
	Before raining ^a	During raining ^b	
Mar. 23-26, 1998	40.6	25.4	37.4
Mar. 28-Apr. 2, 1998	24.7	5.40	78.1
Apr. 3-5, 1998	34.7	13.4	61.3
Apr. 7-13, 1998	33.3	12.5	62.4
Jun. 8-14, 1998	30.8	15.7	48.9

$$c = (a-b)/a \times 100$$

the species like gas and aerosol must first be brought into the presence of condensed water. Then, the species must be scavenged by the hydrometeors, and finally it needs to be deposited on the Earth's surface. To evaluate the scavenging rate of TC by only washout mechanism of rainfall, TSP in the air was sampled before and after five rainfall events using a high volume air sampler. Table 4 shows the TC concentrations both before and after five rainfall events, and scavenging rate of TC by washout mechanism was also listed. In here, we adopted two assumptions that TC was continuously emitted with the same concentration and the constant dry deposition rate was maintained for sampling duration. TC scavenging rate by washout was ranged from 37.4% to 78.1% with an average 57.5%. As might be expected, the high TC scavenging rate by washout mechanism was found. Generally carbonic aerosols have no significant chemical sinks and accumulate in the $0.1-1 \mu\text{m}$ size range. And they are assumed to have a dry deposition of 0.1 cm s^{-1} but their predominant sink mechanism is wet deposition (Harrison

and Grieken, 1998). Mean residence time has been estimated by Orgen and Charlson (1984) to be of the order of 7 days, allowing for synoptic influence of the particles. Residence time, however, is likely to span over a vast range of values from a few days to a month. Such variability is caused by different possible precipitation patterns in source regions but also in height of injection. Another parameter to take into account is the formation of a hydrophilic coating which is likely to facilitate incorporation of particles in cloud droplets.

3.4 EC scavenging rate as the functions of rainfall intensity and rainfall amount

Figure 3 shows the relationships between the scavenging rate of EC and rainfall intensity (I_r) [a], and between the scavenging rate of EC and rainfall amount (A_r) [b], respectively. As shown in the figure, the scavenging rate of EC is gradually increasing in proportion to the increasing of rainfall intensity and rainfall amount. This variation of EC scavenging rate can be explained by the theory of Brownian capture by raindrops. Generally, there are three main processes contributing towards the scavenging of uncharged aerosols by raindrops: [1] Brownian motion, which dominates for aerosol radii less than about $0.05 \mu\text{m}$, [2] inertial collection, which becomes significant only for aerosols larger than a few microns in size, and [3] phoretic forces which are important for intermediate sizes. Pruppacher and Klett (1997) introduced the half-life, $T_{H.L.}$, of an interstitial aerosol with respect to the convective Brownian capture by raindrops as follow;

$$T_{H.L.} = \frac{\ln(2) \rho \alpha^2}{1.35 D \omega f} \quad (1)$$

where, ρ is the density of water, α is the mean radius of the raindrop, D is the diffusivity of EC in air, ω is the liquid water content (LWC). The ventilation coefficient, f , is an empirical enhancement of the particle flux due to the terminal velocity of raindrops.

According to this equation, $T_{H.L.}$ decrease with increasing LWC. Also it can be said that smaller raindrop should have higher EC scavenging rate

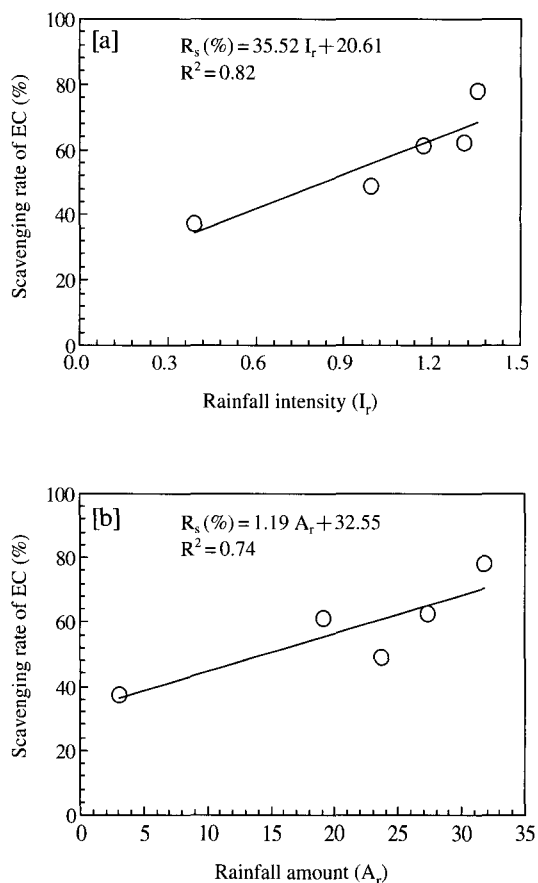


Fig. 3. Scavenging rate of EC according to rainfall intensity (I_r) [a] and rainfall amount (A_r) [b], respectively.

than larger one. A similar scavenging rate for other elements was found in a single raindrop study by Ma *et al.* (2001a).

4. SUMMARY

In this study, the EC content in both rainwater and snow water as well as the EC scavenging rate by rainfall were determined. EC concentration in rainwater varied between $33.6 \mu\text{g L}^{-1}$ and $166.6 \mu\text{g L}^{-1}$. While on the other, that in three snow samples were ranged from $122.4 \mu\text{g L}^{-1}$ to $293.3 \mu\text{g L}^{-1}$. These values were compared to those found in other urban areas. As might be expected, EC show the

significantly high scavenging rate at the initial rainfall. The average TC scavenging rate by washout mechanism was 57.6% for five rainfall events. Also the increase of scavenging rate with increasing rainfall amount and rainfall intensity was found.

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REFERENCES

- Andreae, M.O., T.W. Andreae, R.J. Ferek, and H. Raemdonck (1984) Long range transport of soot carbon in the marine atmosphere. *Science of Total Environment*, 36, 73–80.
- Cachier, H. and J. Ducret (1991) Influence of biomass burning on equatorial African rain. *Nature*, 352, 228–230.
- Chughtai, A.R., M.E. Brooks, and D.M. Smith (1996) Hydration of black carbon. *Journal of Geophysical Research*, 101, 19505–19514.
- Chýlek, P. and W.H. Marlow (1995) Enhanced absorption of solar radiation by cloud droplets containing soot particles in their surface. *Q. Jl. R. Met. Soc.*, 118, 167–172.
- Chýlek, P., G.B. Lesins, G. Videen, J.G.D. Wong, R.G. Pinnick, D. Ngo, and J.D. Klett (1996) Black carbon and absorption of solar radiation by clouds. *Journal of Geophysical Research*, 101, 23367–23371.
- Chýlek, P., L. Kou, B. Johnson, F. Boulada, and G. Lesins (1999) Black carbon concentrations in precipitation and near surface air in and near Halifax, Nova Scotia. *Atmospheric Environment*, 33, 2269–2277.
- Clarke, A.D. (1989) Aerosol light absorption by soot in remote environments. *Aerosol Science and Technology*, 10, 161–171.
- Clarke, A.D. and K.J. Noone (1985) Soot in the arctic snowpack: a cause for perturbations in radiative transfer. *Atmospheric Environment*, 19, 2045–2053.
- Clarke, A.D., K.J. Noone, J. Heintzenberg, S.G. Warren, and D.S. Covert (1987) Aerosol light absorption measurement technical: analysis and intercomparison. *Atmospheric Environment*, 21, 1455–1465.
- Ducret, J. and H. Cachier (1992) Particulate carbon in rain at various temperate and tropical locations. *Journal of Atmospheric Chemistry*, 15, 55–67.
- Ghan, S.J. and J.E. Penner (1992) Smoke, effects on climate. In *Encyclopedia of Earth System Science* 4, Academic Press, pp. 191–198.
- Gray, D.M. and D.H. Male (1981) *Handbook of snow; Principles, processes, management and use*. Pergamon Press, Ontario, pp. 776–786.
- Hallett, J., J.G. Hudson, and C.F. Rogers (1989) Characterization of combustion aerosols for haze and cloud formation. *Aerosol Science Technology*, 10, 70–83.
- Hansen, A.D.A., B.A. Bodhaine, E.G. Dutton, and R.C. Schnell (1988) Aerosol black carbon measurement at the South Pole: initial results, 1986–1987. *Geophysical Research Letters*, 15, 1193–1196.
- Harrison, R.M. and R.E. Van Grieken (1998) *Atmospheric particles (Vol. 5)*. John Wiley and Sons press, Chichester, pp. 333–336.
- Hitzenberger, R., A. Berner, H. Giebl, R. Kromp, S.M. Larson, A. Rouc, A. Kosh, S. Marischka, and H. Puxbaum (1999) Contribution of carbonaceous material to cloud condensation nuclei concentrations in European back-ground (Mt. Sonnblick) and urban (Vienna) aerosols. *Atmospheric Environment*, 33, 2647–2659.
- Hwang K.C. and C.J. Ma (2003) The distribution characteristics and Long-term trend of Carbonaceous Species in Airborne Particulate in Seoul between 1986 and 1996, *Journal of Korea Soc. for Atmospheric Environment*, 19, 11–20.
- Jennings, G.S., M. Geever, F.M. McGovern, J. Francis, G. Spain, and T. Donaghy (1997) Micro-physical and physico-chemical characterization of atmospheric marine and continental aerosol at Mace Head. *Atmospheric Environment*, 31, 2795–2808.
- Knutson, E.O., S.K. Sood, and J.D. Stockham (1975) Aerosol collection by snow and ice crystals. *Atmospheric Environment*, 10, 395–402.

- Kuhlbusch, T.A.J., M.O. Andreae, H. Cachier, J.G. Goldammer, J-P. Lacaus, R. Shea, and P.J. Crutzen (1996) Black carbon formation by savanna fires: measurements and implications for the global carbon cycle. *Journal of Geophysical Research*, 101, 23651–23666.
- Ma, C.J., K.C. Choi, and J.H. Park (2001a) New attempts to the study of individual raindrops. *Korean Environmental Engineering Research*, 6, 81–89.
- Ma C.J., M. Kasahara, S. Tohno, and K.C. Hwang (2001b) Characterization of the winter atmospheric aerosols in Kyoto and Seoul using PIXE, EAS and IC. *Atmospheric Environment*, 35, 747–752.
- Ma, C.J., M. Kasahara, and S. Tohno (2002) Application of PIXE to characterization of size-segregated single raindrops. *International Journal of PIXE*, 12, 7–18
- Malissa, H. and E. Pell (1989) Black snow episodes in Austria. *Aerosol Science and Technology*, 10, 140–150.
- Mitra, S.K., U. Barth, and H.R. Pruppacher (1990) A laboratory study of the efficiency with which aerosol particles are scavenged by snowflakes. *Atmospheric Environment*, 24A, 1247–1254.
- Murakami, M., C. Hiramatsu, and C. Magono (1981) Observation of aerosol scavenging by falling snow crystals at two sites of different heights. *Journal of the Meteorological Society of Japan*, 59, 763–771.
- Ogren, J.A. and R.J. Charlson (1984) Measurement of the removal rate of elemental carbon from the atmosphere. *Science of Total Environment*, 36, 329–338.
- Pinnick, R.G., G. Fernandez, E. Martinez-Andazola, B.D. Hinds, A.D.A. Hansen, and K. Fuller (1993) Aerosol in the arid southwestern United States: measurements of mass loading, volatility, size distribution, absorption characteristics, black carbon content and vertical structure to 7 km above sea level. *Journal of Geophysical Research*, 98, 2651–2666.
- Pruppacher, H.R. and J. Klett (1997) *Microphysics of Clouds and Precipitation*, Kluwer Academic Publisher, Dordrecht. pp. 720–724.
- Smith, D.M., J.J. Griffin, and E.D. Goldberg (1975) Spectrometric method for the quantitative determination of elemental carbon. *Analytical Chemistry*, 47, 233–238.
- Turco, R.P., O.B. Toon, R.C. Whitten, J.B. Pollack, and P. Hamill (1983) The global cycle of particulate elemental carbon: a theoretical assessment. In *Precipitation Scavenging, Dry Deposition, and Resuspension*. Elsevier Science, New York, pp. 1337–1351.
- Twomey, S., M. Piepgrass, and T.L. Wolfe (1984) An assessment of the impact of pollution on the global cloud albedo. *Tellus*, 36B, 356–366.
- Warren, S.G. and A.D. Clarke (1990) Soot in the atmosphere and snow surface of Antarctica. *Journal of Geophysical Research*, 95, 1811–1816.
- Weingartner, E., H. Burtscher, and U. Baltensperger (1997) Hygroscopic properties of carbon and diesel soot particles. *Atmospheric Environment*, 31, 2311–2327.
- Winkler, P. (1988) The growth of atmospheric aerosol particles with relative humidity. *Physica Scripta*, 37, 223–230.
- Wolff, G.T., R.J. Countess, P.J. Groblickie, M.A. Ferman, S. Cadle, and Muhlbaier (1981) Visibility-reducing species in the Denver 'Brown Cloud': II sources and temporal patterns. *Atmospheric Environment*, 15, 2485–2502.