

Chemical composition and source signature of spring aerosol in Seoul, Korea

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Abstract. The measurement of atmospheric aerosol was made in Seoul during the spring of 1998. The objective of this study was to investigate the chemical characteristics of atmospheric aerosol with an emphasis on the effect of Asian dust. Total suspended particles (TSP) and particles smaller than 10 μm (PM_{10}) were collected during March–May 1998. For PM_{10} , water-soluble ions and trace elements were analyzed: NO_3^- , SO_4^{2-} , Na^+ , NH_4^+ , K^+ , Mg^{2+} , Ca^{2+} , Na, Mg, Al, Ca, Fe, Mn, Cr, Co, Cu, Zn, Cd, and Pb. All data were sorted into three groups on the basis of the intensity of Asian dust observed in Seoul: nondust, regular dust, and heavy dust. Chemical characteristics of aerosol were distinct for the heavy dust incidence compared to the other two cases. The average mass concentration of TSP and PM_{10} increased substantially during the heavy dust period. For water-soluble ions, concentrations of SO_4^{2-} and NO_3^- were the lowest, whereas Ca^{2+} and Mg^{2+} concentrations were remarkably enhanced. Concentrations and mass fraction of crustal elements such as Na, Mg, Ca, Fe, and Mn were highly elevated, but those of pollution-derived heavy metals were appreciably decreased. Enrichment ratios of metallic components indicated that soil dust was the primary source of spring aerosols over Seoul. This was also confirmed by factor analysis. Trajectory analysis provided a convincing evidence for the air mass coming from the Asian desert areas, which are the source region of mineral dust. In contrast, the anthropogenic influence was more pronounced for the regular dust period: concentrations of heavy metals, NO_3^- , and SO_4^{2-} were the highest. It could be associated with the transport of air mass over the industrialized regions of China, which is demonstrated by backward trajectory analysis. The result of factor analysis with high factor loadings in SO_4^{2-} and NO_3^- implied the considerable influence of industrial emission on the composition of all spring aerosols over Korea.

1. Introduction

Atmospheric particles are emitted from natural sources such as sea spray and volcanic emissions and from anthropogenic processes such as fossil fuel combustion and industrial emissions. Some of the sources are neither purely natural nor purely anthropogenic. Soil dust is classified as a natural source but with anthropogenic influence, and biomass burning is vice versa. Since the source of atmospheric particles is highly inhomogeneous and their atmospheric lifetime is short, concentrations and chemical composition of aerosols are widely variable in time and space. Furthermore, anthropogenic aerosols have increased dramatically over the past century and are implicated in global climate change through their direct and indirect role in the Earth's radiation balance [Charlson *et al.*, 1992]. As a result, there is a large uncertainty associated with estimating the climatic effect of aerosol.

Soil dust is a major source for coarse aerosol particles. Its

annual global emission amounts to ~ 1500 Tg, which is about 40% of the total aerosol emission [IPCC, 1994]. Because of the large amounts involved, mineral dust is thought to have a considerable effect on the planetary radiation balance [Seinfeld and Pandis, 1998]. The dust also affects human health and the life of terrestrial and aquatic ecosystems, after being deposited onto the soil and surface waters [Molnar *et al.*, 1993]. Mineral dust originates mainly from desert and semidesert regions in northern Africa and central Asia and is transported long distance to the North Atlantic and North Pacific [Prospero, 1989]. Arid and semi-arid regions in northern and northwestern China are a major source for mineral aerosol particles and for various trace elements, including iron and aluminum in the air and water of the North Pacific region [Martin and Gordon, 1988; Arimoto *et al.*, 1989; Prospero *et al.*, 1989]. A geochemically significant quantity of Asian dust, currently estimated to be 400–500 Tg, is deposited in the North Pacific each year [WMO, 1989]. In spring, dust storms take place in deserts and dry loess plateau (e.g., Gobi and Taklamakan in Mongolia and northwest China). These vast areas are located in the lee of the massive East Asian Mountains where a convergent airflow and a lee cyclone take place on a synoptic scale. The downslope forms a very dry surface, and annual rainfall in the majority of the desert and/or semidesert area is less than 300 mm. In particular, a convergent flow in a lee cyclonic system enhances an upward motion, and dust produced by mechanical and dynamic turbulence can be transported to the midtroposphere [Chung and Yoon, 1996]. Hence the Asian dust is typically observed in east Asian countries, including China, Korea, and Japan in springtime.

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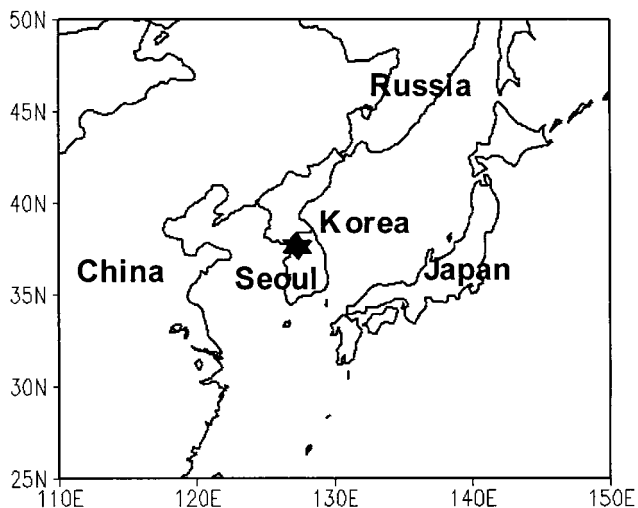


Figure 1. Map of study area showing eastern Asia, including the Korean peninsula and Seoul.

During Asian dust periods, several observations have been made in the eastern part of China [Zhou *et al.*, 1981; Wang *et al.*, 1982], Japan [Mizohata and Mamuro, 1978], and the North Pacific Ocean [Duce *et al.*, 1980; Uematsu *et al.*, 1983]. Cho [1980] and Kim *et al.* [1986] reported that the number concentration of aerosols in Korea was relatively higher in spring than other seasons. However, there are still very few sets of observed data in Korea, particularly for chemical composition [Pacyna and Ottar, 1985; Carmichael *et al.*, 1996, 1997]. Thus more research is required to investigate factors determining physical, optical, or chemical properties of aerosols, particularly during Asian dust events in Korea. Since the concentration of different species in the atmospheric aerosol has been significantly affected by human activities, the study of the elemental composition of the particles is crucial for environmental management on local, regional, and continental scales.

In this study, total suspended particulate (TSP) and particulate smaller than $10\ \mu\text{m}$ (PM_{10}) were collected in Seoul during March–May 1998. Through the chemical and statistical analysis of particulate composition, an attempt was made to define the chemical characteristics and contributing sources of atmospheric aerosols in Seoul during spring when Asian dust events frequently occurred.

2. Experiment

During March–May 1998, aerosols were collected on the roof of the old Meteorological Research Institute building (12 m height) in the northern and central Seoul. Seoul is the capital of South Korea and located in the central part of Korea, about 30 km east of the Yellow Sea (Figure 1) [Chun *et al.*, this issue, Figure 4]. North of the sampling site is a mountainous area and downtown is south of it. As of the end of 1997, the area of Seoul is $605.52\ \text{km}^2$ with a population of 10 million people, which accounts for about a quarter of the total national population. Samples were taken in spring when Asian dust is frequently observed over Korea. Details about meteorological conditions and synoptic features of this study area are provided in another paper [Chun *et al.*, this issue].

Total suspended particles (TSP) and particles with an aerodynamic diameter smaller than $10\ \mu\text{m}$ (PM_{10}) were collected

on cellulose membrane filters exposed for 24 hours (from 0010 to 0010 LT) using high-volume samplers (Andersen) at the average flow rate of $1.1\ \text{m}^3/\text{min}$. Filters were preweighed and then dried in a desiccator for at least 24 hours after being exposed to air. Before the final weighing, filters were stored in an environmental chamber (HOTPACK model 305502, Customer & Technical Service, United States) for 24 hours at a constant humidity (50% RH) and temperature between 20° and 23°C .

Chemical analysis was done for soluble ionic species such as NO_3^- , SO_4^{2-} , Na^+ , NH_4^+ , K^+ , Mg^{2+} , and Ca^{2+} and metallic elements such as Al, Ca, Mg, Na, Fe, Mn, Cr, Co, Ni, Cu, Zn, Cd, and Pb. The filters were shaken for 10 min in 40 mL deionized water, and an extract was filtered through a $0.45\ \mu\text{m}$ cellulose acetate membrane. The filtrate was then analyzed for water-soluble ionic species using ion chromatography (Dionex DX 500). Metal ions were extracted with 5 mL of mixed acids ($\text{HF}:\text{HNO}_3:\text{HClO}_4 = 4:4:1$) in 60 mL Teflon beaker. GR grade reagents were used after being refined, employing quartz and Teflon subboiling reflux system. Al, Fe, Ca, Mg, Na, and Mn were determined using inductively coupled plasma atomic emission spectrophotometer (ICP/AES, Shimadzu ICP-IV). Inductively coupled plasma mass spectrophotometer (ICP/MS, VG PQ II+) was used for the analysis of Cr, Co, Ni, Cu, Zn, Cd, and Pb [Broekaert *et al.*, 1983]. The ICP/AES system was calibrated for every 10 samples, and internal standards (^{115}In , ^{205}Tl) were added to both standard solutions and samples for the analysis using ICP/MS. The accuracy of ICP analysis checked using NIST SRM 1646 was within 5–10% for all elements. Meteorological parameters, including air temperature, relative humidity, wind speed and direction, barometric pressure, and solar radiation were concurrently measured.

3. Results and Discussion

3.1. Asian Dust Episodes

Asian dusts observed in Korea are classified into regular, heavy, or very heavy dust according to its intensity. The “regular dust” refers to the weakest dust event causing visibility to be slightly decreased with no recognizable amount of deposition. If there is a deposition on the surface along with considerable reduction in visibility, it is rated “heavy dust.” The dust event that turns the sky into brownish yellow and is accompanied by heavy deposition is classified as “very heavy dust.” In spring 1998, Asian dust occurred in Seoul during March 28–30 and April 14–22. Of these, heavy dust was detected on April 19 and 20 with daily averaged PM_{10} over $150\ \mu\text{g}/\text{m}^3$, and the heavy dust cloud was also captured in satellite [Husar *et al.*, this issue]. To characterize the chemical properties of spring aerosol in 1998, all data were sorted into three categories: nondust, regular dust, and heavy dust. There was a total of 35 aerosol samples taken in Seoul during the springtime of 1998 (March–April). Out of the 35 samples, seven collected March 28, 29, and 30 and April 14, 15, 18, and 21, and two collected April 19 and 20 fell into regular dust and heavy dust categories, respectively. The other 26 samples were considered nondust cases. Although Asian dust was observed April 17 and 22, samples collected on these two days were included in the nondust category due to rain.

3.2. Aerosol Mass Concentrations

Figure 2 shows the time series variation of daily concentrations of TSP and PM_{10} observed in Seoul during March–May

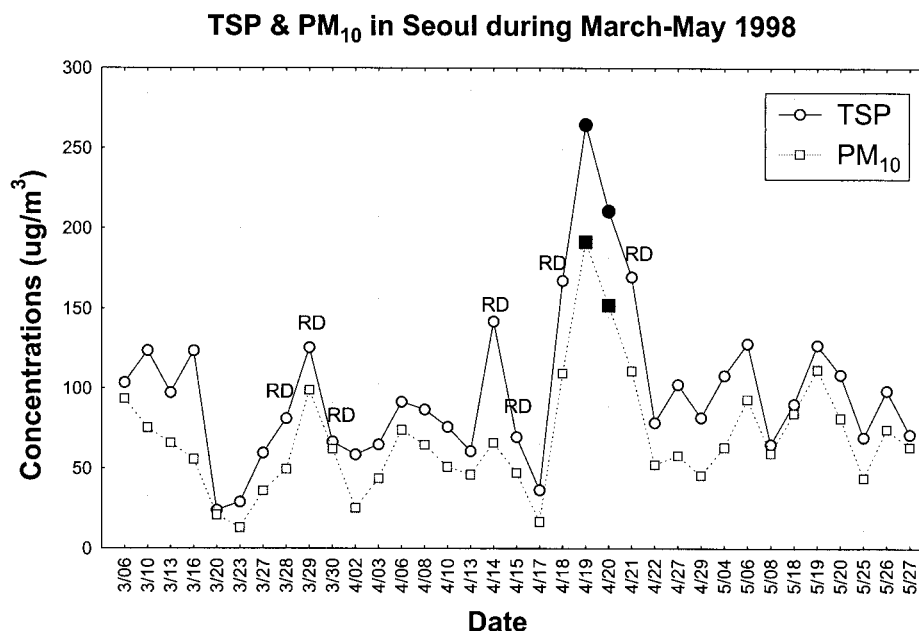


Figure 2. Time series variation of total suspended particles (open circles) and PM₁₀ (open squares) concentrations observed in Seoul during March–May 1998. Concentrations are in $\mu\text{g}/\text{m}^3$. Data for the heavy dust case were marked with solid circles and solid squares, and those for the regular dust case are labeled “RD.”

1998. For the regular dust case, mass concentration was increased but not significantly. Mass concentrations of both TSP and PM₁₀ show a strong peak between April 18 and 21. During the heavy dust incursion (April 19–20), mass concentrations of both TSP and PM₁₀ increased substantially and reached to 238 and 172 $\mu\text{g}/\text{m}^3$, respectively. Particularly, the aerosol concentration of PM₁₀ was far above the Korean air quality standard of 150 $\mu\text{g}/\text{m}^3$ (24 hour average). The mass concentrations averaged for three cases are given in Table 1 with their maximum and minimum values.

3.3. Aerosol Composition

3.3.1. Water-soluble ions. In Table 2, average concentrations of water-soluble ions in PM₁₀ are given for three cases. Concentrations of NO_3^- , non-sea-salt (nss) SO_4^{2-} , and NH_4^+ were the highest during the regular dust period, whereas Na^+ , K^+ , Mg^{2+} , and Ca^{2+} concentrations were the highest during the heavy dust period. Concentrations of nss SO_4^{2-} and NO_3^- were considerably increased during the regular dust period. Particularly, the concentration of NO_3^- was extremely high compared with that measured in Cheju Island, which is located in the southern part of Korea, during the spring for 3 years from 1992 to 1995 [Charmichael *et al.*, 1996, 1997]. During the heavy dust incidence, the concentration of nss SO_4^{2-} was also

elevated, while NO_3^- concentrations remained unchanged. Accordingly, the ratio of nss SO_4^{2-} to NO_3^- was close to 1 for both the nondust and the regular dust periods and a little higher (1.37) for the heavy dust period. Higher concentrations of nitrate and nss sulfate during the regular dust events may suggest an anthropogenic influence in the transport process and a catalytic effect of dust particles in the oxidation processes of nitrogen and sulfur compounds [Charmichael *et al.*, 1996; Dentmer *et al.*, 1996]. However, the nss SO_4^{2-} to NO_3^- ratio during the heavy dust period remains to be explained. The enhancement of Ca^{2+} and Mg^{2+} concentrations during the heavy dust event was remarkable compared to those of the nondust period. These results revealed that Ca^{2+} is a good indicator for Asian dust events and the intensity of dust events.

The weighed percentage of soluble ions against mass concentrations of PM₁₀ was calculated and listed in Table 3, which shows that the major components of PM₁₀ were NO_3^- , nss SO_4^{2-} , and NH_4^+ . These species were the most dominant during the regular dust event and amount to about 40% of the particle mass in PM₁₀. On the other hand, the fraction of these soluble ions decreased to 18% during the heavy dust event, which is about half of that for the nondust period (37%). This was mostly due to decrease in the concentrations of NO_3^- and nss SO_4^{2-} against the total mass in spite of an increase in concentrations of Mg^{2+} and Ca^{2+} . The sum of water-soluble ion concentrations in PM₁₀ increased slightly from the nondust to the dust periods but substantially decreased from the nondust to the heavy dust periods. This characteristic of chemical composition should be related to the source and chemical and physical processes during the transport of aerosols, which will be discussed in the following section.

3.3.2. Elemental metals. Metal concentrations averaged for each case are listed in Table 4. For crustal elements, including Na, Mg, Al, Ca, Fe, and Mn, concentrations were

Table 1. Mass Concentrations of TSP and PM-10

Case	Number of Samples	Mass Concentrations, $\mu\text{g}/\text{m}^3$	
		TSP	PM-10
Nondust	26	83 (24–128)	58 (13–112)
Regular dust	7	117 (67–170)	78 (47–111)
Heavy dust	2	238 (211–265)	172 (152–192)

Mean and range in parentheses.

Table 2. Concentrations of Water-Soluble Ions in PM-10, $\mu\text{g}/\text{m}^3$

Case	NO_3^-	SO_4^{2-}	NH_4	Na^+	K^+	Mg^+	Ca^+
Nondust							
mean	8.04	7.28	4.50	0.77	0.36	0.08	0.68
minimum	1.09	1.55	1.06	0.41	0.11	0.02	0.14
maximum	26.56	25.26	11.90	1.44	1.02	0.14	1.24
Dust							
mean	13.14	12.76	5.51	0.88	0.56	0.13	1.32
minimum	6.99	7.98	3.79	0.64	0.30	0.04	0.56
maximum	20.37	22.11	7.13	1.14	1.09	0.27	3.43
Heavy dust							
mean	8.16	11.16	3.87	1.29	0.63	0.33	4.66
minimum	7.27	11.01	3.19	1.08	0.56	0.30	4.44
maximum	9.05	11.32	4.55	1.50	0.70	0.37	4.88
			<i>Ratios</i>				
Dust/nondust	1.63	1.75	1.22	0.15	1.54	1.54	1.93
Heavy dust/nondust	1.02	1.53	0.86	1.68	1.72	3.95	6.85
Heavy dust/dust	0.62	0.87	0.70	1.46	1.12	2.57	3.55
			<i>Detection Limits^a</i>				
	0.003	0.002	0.007	0.008	0.003	0.002	0.019

^aDetection limits ($\mu\text{g}/\text{m}^3$) were determined by 3 times standard deviation of six blank filters.

slightly increased during the regular dust period and then substantially increased during the heavy dust event compared to those of the nondust period. The enhancement of these elements during the heavy dust period was more than a factor of 4 relative to the period of nondust. The mean concentration of Al during the heavy dust event was $10.5 \mu\text{g}/\text{m}^3$, which is close to that observed in the dust source region ($10 \mu\text{g}/\text{cm}^3$ at Xi'an) in the spring 1992 [Gao *et al.*, 1997]. Gao *et al.* [1997] also reported that Al concentrations decreased toward the east from Xi'an (located within the Loess Plateau region). Aluminum accounts for $\sim 8\%$ of crustal rock and is usually used as a dust indicator. Liu *et al.* [1985] showed that the Al content amounts to $\sim 7\%$ of the total mass of loess. During the heavy dust period in 1998 the mass of Al was about 6% of the total mass of PM_{10} .

For other elements, such as Ni, Cu, Zn, Cd, and Pb, the increase in concentrations was the most noticeable during the regular dust period. For these elements, concentrations were elevated about 30–90% during the regular dust period compared to the nondust period. For the heavy dust period, concentrations were comparable to those of the nondust event. The mass fraction of each element relative to the total mass of PM_{10} was the highest during the heavy dust event for the crustal element (Table 5). However, the mass fraction of other trace metals was apparently reduced during the heavy dust period.

The elemental ratios in the samples can give qualitative information on the source of materials [Duce *et al.*, 1983]. For this purpose the enrichment factor (EF) of an element (X) in aerosol relative to the crustal material was calculated as

$$EF_{Crust(X)} = \frac{(X/Al)_{aerosol}}{(X/Al)_{crust}},$$

where X is the concentration of the element of interest. Aluminum was used as a reference element. The elemental composition of the average crustal material was adopted from Taylor and McLennan [1985]. If EF approaches unity, the crust is probably the predominant source of the element. The average EF of trace elements for each case are presented in Figure 3. The calculations of enrichment factors indicate that continental crust was the dominant source for elements such as Na, Mg, Ca, Fe, and Mn for spring aerosols in Seoul. Since the elemental composition of the actual aerosol precursor may be different from that of average crustal rock, EF of 2–5 is usually considered as crustal origin. The enrichment factors of heavy metals derived from anthropogenic activity were, however, known to be much higher than 10. The enrichment factors for all metals were not distinguishable between nondust and regular dust periods but substantially lower during the heavy dust period. It implies that anthropogenic influence was comparatively reduced during heavy dust events. These results should be associated with different sources of aerosols and meteorological or chemical processes during the transport of aerosols, which was further examined using statistical and trajectory analyses.

3.4. Factor Analysis

To determine the characteristic source and the contributing factors of spring aerosol observed in Seoul, factor analysis was

Table 3. Mass Fraction of Soluble Ions Against Mass Concentration of PM_{10} , %

Case	NO_3^-	SO_4^{2-}	NH_4^+	Na^+	K^+	Mg^+	Ca^+
Nondust	13.83	12.52	7.74	1.32	0.63	0.14	1.17
Dust	16.92	16.44	7.09	1.13	0.72	0.17	1.69
Heavy dust	4.75	6.50	2.26	0.75	0.36	0.19	2.72

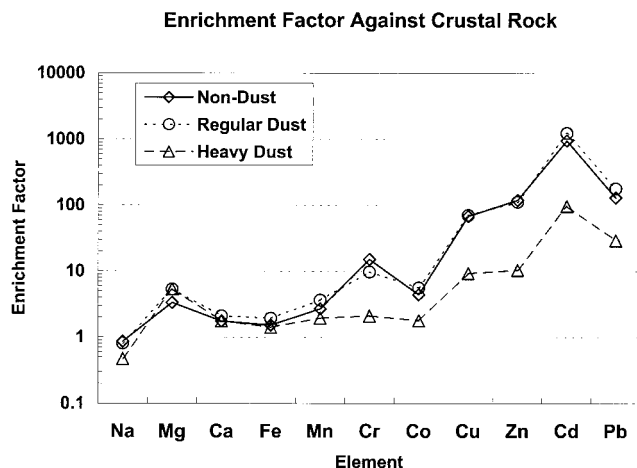


Figure 3. Enrichment factors of selected elements against crustal rock using Al as a reference element averaged for each case.

the other hand, the air originated from the ground level of desert regions and was uplifted during transport, reaching a 4 km level over Seoul in 4 days (Figure 5). The air mass shown in Figure 4 passed over heavily industrialized areas in north-eastern China, which is possibly linked with highly elevated nss SO_2 , NO_3 , and some heavy metals such as Pb, but low Ca during the regular dust period.

These trajectories are good evidence for the influence of Asian dust on spring aerosols over Seoul through long-range transport. Merrill *et al.* [1985] already demonstrated using the trajectory analysis that Asian dust was transported to the remote North Pacific at ~ 5 km above sea level. Satellite observation clearly shows the impact of soil dust on aerosol in the northwestern Pacific Ocean during springtime [Husar *et al.*, 1997]. Husar *et al.* [this issue] established that a big dust storm was generated in Asian desert regions on April 15 and a dust cloud was transported across the Korean peninsula. In the source regions of dust, dust swirls and storms were also ob-

Table 6. Factor Analysis of All Samples

Variable	Factor 1	Factor 2	Factor 3
NO_3^-	0.08	0.44	0.81
SO_4^{2-}	0.12	0.11	0.89
Na^+	0.56	-0.14	0.19
NH_4^+	-0.02	0.39	0.85
K^+	0.23	0.07	0.70
Mg^+	0.90	-0.14	0.14
Ca^+	0.95	-0.01	0.09
Na	0.88	0.18	-0.07
Mg	0.98	0.07	-0.01
Al	0.95	0.11	0.06
Ca	0.96	0.16	0.09
Fe	0.95	0.25	0.05
Mn	0.90	0.37	0.11
Cr	0.04	0.11	0.14
Co	0.57	0.42	<0.05
Ni	0.25	0.76	0.40
Cu	0.07	0.74	0.50
Zn	-0.03	0.66	0.41
Cd	0.08	0.91	0.05
Pb	0.28	0.85	0.17
Explained	7.87	4.04	3.59
Percent total	36	18	16

Table 7. Factor Analysis of Nondust Samples

Variable	Factor 1	Factor 2
NO_3^-	0.16	0.88
SO_4^{2-}	0.12	0.85
Na^+	<0.05	0.09
NH_4^+	0.20	0.93
K^+	0.20	0.63
Mg^+	0.41	-0.02
Ca^+	0.68	0.14
Na	0.57	-0.18
Mg	0.90	0.01
Al	0.94	0.13
Ca	0.93	0.22
Fe	0.93	0.23
Mn	0.83	0.29
Cr	0.13	0.04
Co	0.52	-0.02
Ni	0.62	0.54
Cu	0.69	0.56
Zn	0.74	0.42
Cd	0.19	0.50
Pb	0.25	0.62
Explained	7.09	4.67
Percent total	32	21

served 3–5 days ahead of the heavy dust incidence in Korea [Chun *et al.*, this issue]. The result of factor analysis already indicated that soil dust was the most important factor determining chemical composition of spring aerosol. It is therefore

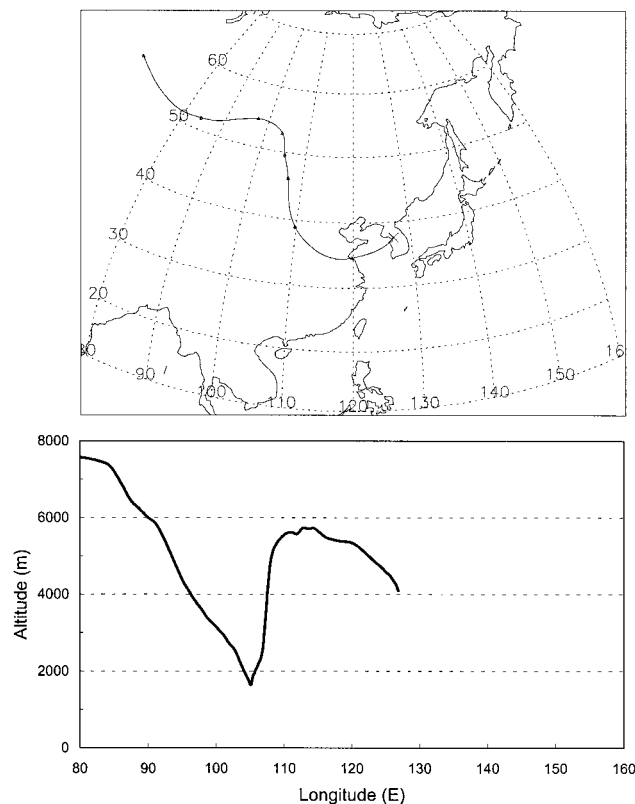


Figure 4. Isentropic trajectory back in time from Seoul (4 km altitude) for April 14, 1998, at 0600 UTC. Each triangle denotes a trajectory every 12 hours, and trajectories are shown for 4 days. The bottom panel shows the vertical motion of air mass along isentropic surface.

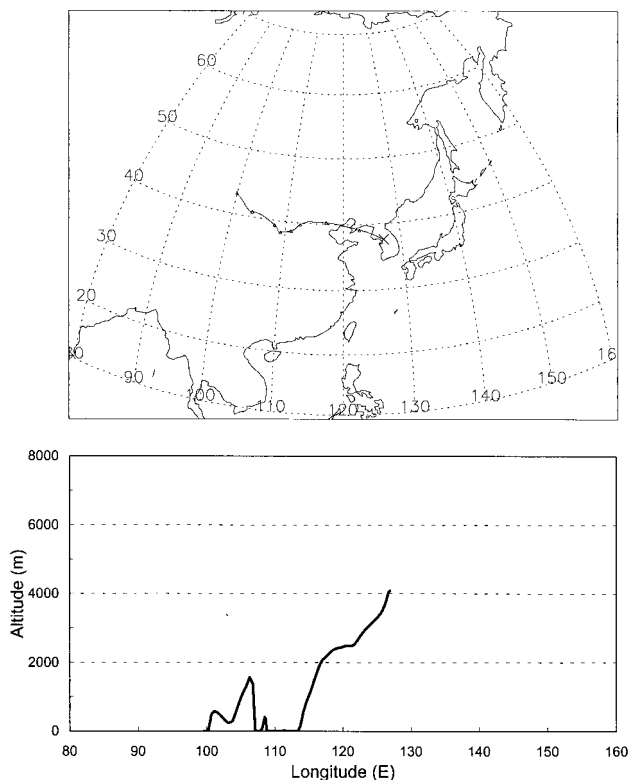


Figure 5. Isentropic trajectory back in time from Seoul (4 km altitude) for April 19, 1998. Others are the same as Figure 4.

evident that Asian dust transported for a long distance is the primary source of aerosols over Seoul during spring.

4. Conclusion

The chemical characteristics of Asian dust were well captured in aerosols collected in Seoul during April 19–20, 1998. Particle concentrations were directly affected by dust events. The average concentrations of both TSP and PM_{10} increased from the nondust to dust period and reached the maximum on the heavy dust incidence, when PM_{10} concentrations were well above the Korean air quality standard of $150 \mu\text{g}/\text{m}^3$. Major water-soluble ions were NO_3^- , SO_4^{2-} , and NH_4^+ , which suggests anthropogenic influence was significant on spring aerosols collected in Seoul. The enhancement of NO_3^- was remarkable only during the regular dust events, whereas that of SO_4^{2-} was considerable for both regular and heavy dust events. The difference in enhancement between SO_4^{2-} and NO_3^- during dust events was not unambiguously explained, and it needs further study. In comparison, Ca^{2+} and Mg^{2+} were clearly shown to be good indicators for dust incursions. While the concentrations of crustal elements were highly elevated during the heavy dust period, their enrichment factors against aluminum remained nearly constant through the whole course of the experiment. The contribution from soil dust was extracted as a pronounced factor, determining aerosol composition in spring. These results indicate that mineral dust is a main source of spring aerosols collected in Seoul. Its signature was the most evident during the heavy dust period when the air mass originated from right above the dust source regions in Asian deserts. It is

also implied that the chemical composition of aerosol is closely linked to air mass trajectories during long-range transport.

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