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Transport and evolution of a winter-time Yellow sand observed in Korea

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Abstract

Long-range transport of mineral dust such as Yellow sand (YS) is not restricted to the springtime periods in Northeast Asia. A YS phenomenon was observed during 25–27 January 1999, which was a remarkably distinctive episode in the occurrence time and intensity that had ever observed in the wintertime in Korea. This YS event was traced to be originated from the arid region of central and eastern Asia; the Gobi desert and Loess plateau. The traveling speed of the dust storm was found to be about 70 km h^{-1} with it's horizontal size of larger than the whole Korean peninsula during this episode. Aerosol mass loadings changed by an order of magnitude within a few hours. The dominant ion components were SO_4^{2-}, NO_3^-, Ca^{2+} and Na^+ during the passage of YS. The mode diameter of these compounds of YS was around 4 μm, compared to 0.4–0.9 μm after the passage of YS. SO_4^{2-} and NO_3^- concentrations were found to be well correlated with Ca^{2+} concentration in the coarse mode during the YS event, whereas they were well correlated with NH_4^+ concentration during the non-YS period, indicating a significant amount of SO_4^{2-} and NO_3^- formations on the Ca^{2+}-rich coarse aerosol during the long-range transport of YS.

Keywords: Yellow sand; Long-range transport; SO_4^{2-} and NO_3^- formations

1. Introduction

Atmospheric aerosols are important components of the earth–atmosphere system, which play a major role both in the global climate and in many biogeochemical cycles. Aerosols also help control the concentration, lifetime and the physical and chemical behavior of many important tracer gases by providing reaction sites and serving as carriers and/or sinks for many atmospheric species (Zhang and Carmichael, 1999; Dentener et al., 1996).

Yellow sand (YS), in other words "Asian dust", observed in the Korean peninsula originates mainly from the Takla–Makan Desert, the Gobi Desert and the Loess Plateau of China. Central Asia is one of the world's largest dust sources, however, the magnitude of this source remains highly uncertain but is to be of the order of 800 Tg yr^{-1} (Zhang et al., 1997). The Yellow sand phenomena are generally observed 3–5 times in Korea during the spring season (MOE, 1999) with the maximum frequency in April. Kang and Sang (1991) found that the mean concentration of total suspended particles during the Yellow sand period exceeded 500 μg m^{-3} over a large geographical region.

In addition, the production and long-range transport of mineral aerosols from the Asian continent may exert significant radiative forcing over a large area of the North Pacific region (Uematsu et al., 1983; Prospero and Savoie, 1989; Gao et al., 1992; Jaff et al., 1999). Therefore, the interest of atmospheric chemists and aerosol

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scientists has recently turned to the Asia/Pacific region, because the aerosol loadings there already have been seriously perturbed by anthropogenic activities (ACE-Asia, 1999).

There have been many researches regarding the characteristics of mineral aerosol (Pye, 1987; Hirai et al., 1991; Nishikawa et al., 1991; Gao et al., 1992). Most measurements have been conducted downwind of the desert/loess areas in order to investigate the role of dust particles as carriers for anthropogenic chemical constituents, such as SO_2, NO_x, and HCl. Zhang et al. (1993) performed size-resolved factor analysis in five Chinese cities and they found both fine and coarse aerosol to be enriched in sulfur. Gao et al. (1991) and Nishikawa et al. (1991) measured a substantial fraction of sulfate in the coarse aerosol in Japan during the YS period. Iwasaka et al. (1988) and Parungo et al. (1996) discovered through a TEM imaginary experiment that the mineral aerosol (Yellow sand) were coated by sulfate- and nitrate-containing solutions.

However, the roles of YS on the long-range transport of the sulfur and nitrogen compounds are largely unquantified due to the lack of detailed observed data. In general, aerosol measurements are insufficient to understand the characteristics of YS especially for the case of the winter-time YS event. Therefore, the objective of this study is to examine the physical and chemical properties of YS through the analysis of aerosols measured during the winter-time YS event in Korea.

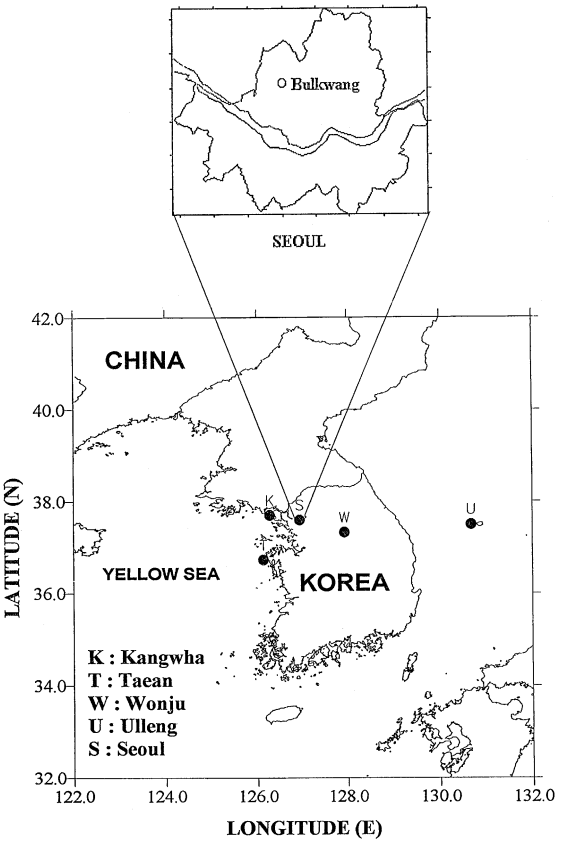


Fig. 1. Locations of monitoring sites in Korea. (K), Kangwha, (S), Seoul, (W), Wonju, (U), Ulleng, and (T), Taean.

2. Measurements

Four sites (Kangwha, Seoul (Bulkwang), Wonju, and Ulleng) of the routine air pollution monitoring network of Korea were chosen for the analysis (Fig. 1). Monitoring locations were selected to the direction of the dust storm propagation. Total suspended particle (TSP) was measured with Dasibi (7001) by the β-ray absorption method.

A cascade impactor (Anderson, AN-200) was used to identify the particle size distribution of aerosol at the Bulkwang monitoring site in Seoul (Fig. 1). The complete unit consists of a sampler, flow meter and pump (Hitachi RC-20S, 200W) whose flow rate was controlled at $28.3\,l\,min^{-1}$. It has eight size-fractionated stages with the nominal cut size of 11.0, 7.0, 4.7, 3.3, 2.1, 0.65, 0.43 μm, and backup filter. The substrates used in the impactor were Teflon membrane filters 80 mm in diameter and 0.22 μm pores (Hi-Fil), but Quartz filters (Whatman, microfiber filters) for the backup filters. The sampling was carried out from 1500 LST 25 January to 1215 LST 27 January 1999 for the YS event and 1040 LST 28 January to 1020 LST 1 February 1999 after YS event (AYS).

A dichotomous air sampler (Sierra 244) was used for the measurement of the suspended particles with two size fractions of 2.5 – 10 μm (coarse) and less than 2.5 μm (fine) in the Taean site (Fig. 1). The Taean site is located 20 m above the mean sea level (msl). This site is the nearest monitoring station from the Shandong peninsula of China. The samples were collected uniformly on two 37 mm Teflon membrane filters with 2 μm pore size. The virtual impactor head has a single-stage design with a cut-point of 2.5 μm and a flow rate of $1\,m^3\,h^{-1}$ (16.7 lpm). The sampling module consisted of the aerosol inlet, virtual impactor head, two 37 mm filter holders, and tripod mount. The sampling was done for 24 h with 1-day period during the YS event, and with 6-day period during the non-YS period from 7 January to 31 February 1999.

Analyzed species of both samples were ion components such as SO_4^{2-}, NO_3^-, Cl^-, NH_4^+, K^+, Ca^{2+}, Na^+, and Mg^{2+}. Water-soluble species were extracted with 20 ml of distilled and deionized water for 60 min. Concentrations of SO_4^{2-}, NO_3^-, and Cl^- were determined by the ion chromatography (Dionex-500). Concentrations of K^+, Ca^{2+}, Na^+, and Mg^{2+} were analyzed by the atomic absorption spectroscopy (Varian, SpectraAA 800), while that of NH_4^+ was determined by indophenol colorimetric method (Perkin-Elmer, Lambda 20).

Backward trajectory analyses on the isentropic surface (290 K) had been carried out for air parcels arriving at Seoul. Global Data Assimilation and Prediction System (GDAPS) from Korean Meteorological Administration were used for the analysis. Meteorological data of 6-h interval and 1.25° of horizontal scale were used (KMA, 1999).

3. Results and discussion

3.1. Meteorological field

Synoptic analyses of weather maps showed that the strong low level wind was remarkably intensified on 24 January 1999 over the desert area in China, one day before the observation of Yellow Sand in Seoul. It could be attributed to the meteorological conditions of higher temperature about 3°C and less precipitation amount than the normals over Northern China during January, 1999 (KMA, 1999). Over the source region in China, the synoptic weather condition was favorable to produce much dust by strong pressure gradient and strong baroclinicity on the 850 hPa level. The surface and 850 hPa weather maps at 25 January in 1999 were shown in Fig. 2. The dominant feature on 850 hPa was strong continental outflow from northeast China to Korean peninsula, behind the intensified deep trough extending from Manchuria to the East Sea (Fig. 2b). This trough was associated with the surface low-pressure system centered at Manchuria accompanied with a cold front extending southeastward through the East Sea. The strong surface pressure gradient behind the front made it possible to bring Yellow sand ladden air over Korean peninsula (Fig. 2a).

Backward trajectories were carried out using RDAPS (Regional Data Assimilation and Prediction System) on the isentropic surface (Fig. 3). The air parcels were transported to Korea through the Gobi desert and the Loess plateau during 25–27 January, 1999. The YS event was detected in Korea after the passage of the moving low-pressure system over Korea with a little rainfall in the morning on 25 January. About 10 h later, the intensity of YS abruptly decreased. The first arriving short and strong dust pulse was followed by a weak and long-lasted second one. After the precipitation in the late afternoon of 27 January, the bi-modal type of the YS event disappeared over the whole country.

The sea-level pressure of 25 January at Seoul, Korea dropped to 1012 from 1019 hPa of the previous day in association with the passage of low-pressure system, and rose to 1020–1023 hPa (KMA, 1999). The visibility also remarkably decreased to 4–5 km during the YS event. The diurnal mean relative humidity was 57–66%, and temperature was 3.8–5.0°C. These values were a little higher than the monthly averages of January (58% and −0.8°C). Unexpectedly, the relative humidity (RH) was not low during the event. As explained later, it could be a favorable condition for the absorption and subsequent conversion of SO_2 and NO_x into sulfate and nitrate.

3.2. The transport behavior of Yellow sand

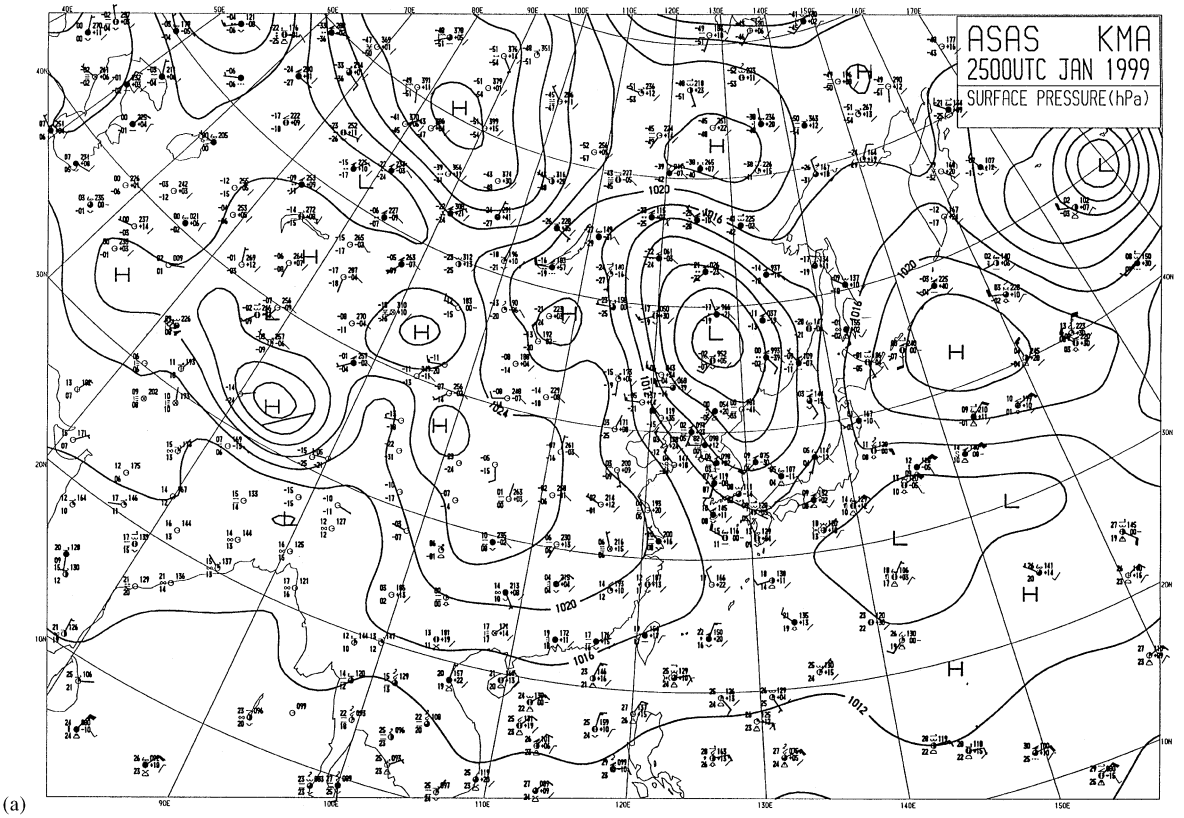
The temporal variations of hourly average TSP concentrations at four sites were shown in Fig. 4 from 25 to 27 January, 1999. The observed YS event had a bi-modal temporal structure. The first dust plume was detected on 25 January with the hourly averaged maximum concentration of almost $1000\,\mu g\,m^{-3}$, and the daily average concentration of $210–276\,\mu g\,m^{-3}$. It was followed by the wide-spreaded and long-lasted second one with the hourly average maximum concentration of $670\,\mu g\,m^{-3}$ and the daily average concentration of $232–349\,\mu g\,m^{-3}$. These daily average values were lower than, but comparable to those of some Chinese cities of $400\,\mu g\,m^{-3}$ (UNDP, 1996). The annual average concentration of TSP for all stations in Korea was $70\,\mu g\,m^{-3}$ in Korea (MOE, 1999).

The starting time of the YS event in Korea was assumed to be the time when hourly average concentration of TSP exceeded $150\,\mu g\,m^{-3}$, which is the national long-term criteria for the hourly average concentration. The first time when TSP concentrations exceeded more than $150\,\mu g\,m^{-3}$ was at 0900 LST 25 January at Kangwha island which is located in the west coast of peninsula (Fig. 4). This event lasted for 13 h until 21 LST. Two hours later, it was continuously observed in Seoul, which is located about 50 km east of the Kangwha site. The TSP concentration also increased at the Wonju site 2 h later and at Ulleng island 5 h later than at Kangwha site. Ulleng island is loacted in the middle of the East Sea between Korea and Japan (Table 1).

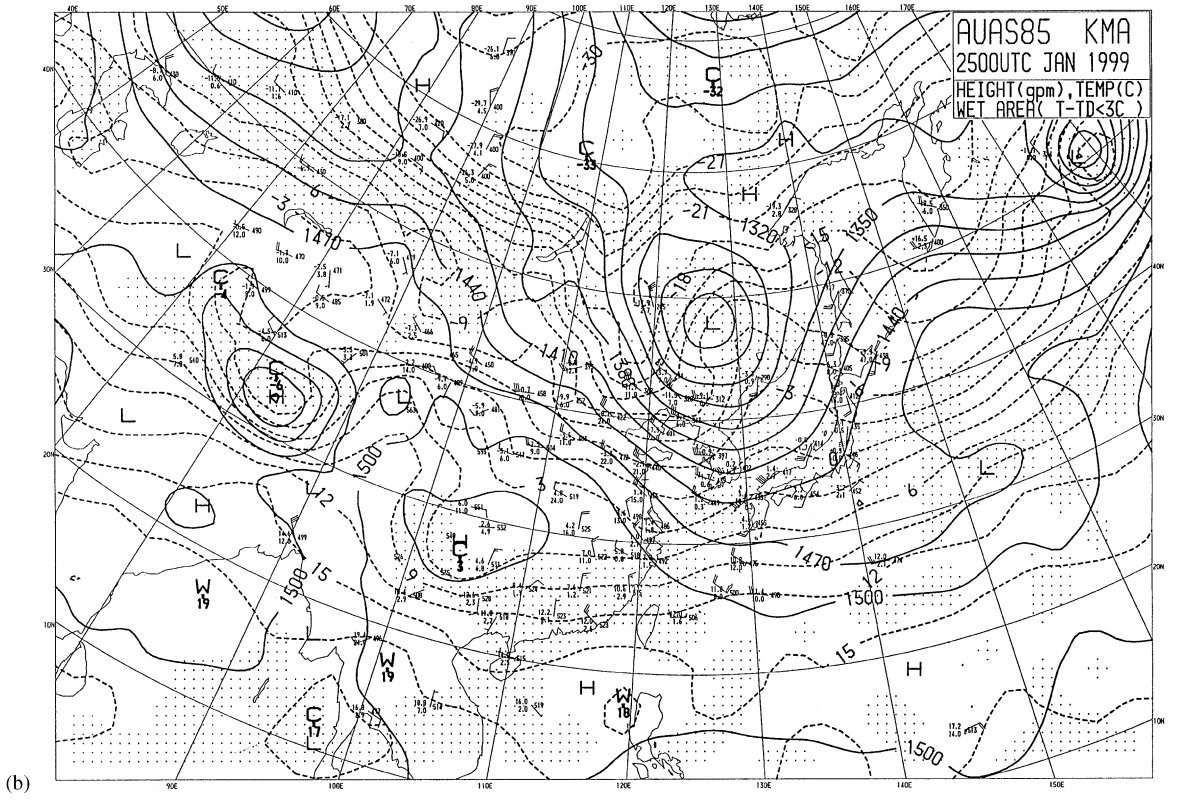
The propagation speed of the dust storm could be estimated using the maximum concentration occurring time. From Table 2, traveling speed of the first dust plume of this episode was roughly $70\,km\,h^{-1}$, that is comparable to $60\,km\,h^{-1}$ estimated by Nishikawa (1993). The horizontal scale of the first dust event was about 710–920 km estimated from the propagation speed and the duration time. It was followed by the wide-spreaded second one. However, the second dust plume occupied so large region over Korea that the maximum-concentration occurring time could not be identified clearly. The duration time of second dust event ($TSP > 150\,\mu g\,m^{-3}$) over Korea was approximately 24–30 h. During this episodic period, aerosol mass loadings changed by an order-of-magnitude within a few hours.

3.3. The chemical properties and the particle size distributions of Yellow sand

The particle size distributions obtained from the cascade impactor were shown in Fig. 5 for the YS and AYS



(a)



(b)

Fig. 2. Weather maps on the surface (a) and on 850 hPa (b) 25 January, 1999.

periods. This comparison made it possible to investigate how the size distribution of dust particles could change under the different dust loading. The dominant ion components were SO_4^{2-}, NO_3^-, Ca^{2+} and Na^+ with the total sum of eight-stage cascade impactor filter concentrations of 11.3, 7.6, 6.1 and 4.2 $\mu g\,m^{-3}$ respectively during the YS period, compared to the corresponding concentrations of 4.1, 4.6, 0.4 and 1.2 $\mu g\,m^{-3}$ after the passage of YS (AYS) over Korea.

Most ion components increased for all sizes during the YS period, compared to the AYS period. The overall remarkable increase in the coarse mode from 2.0–11.0 μm was seen in Fig. 5. However, only the NH_4^+ concentration decreased significantly during the Yellow sand period (1.89 $\mu g\,m^{-3}$), compared to the AYS period (3.34 $\mu g\,m^{-3}$). The mode diameter of SO_4^{2-} and NO_3^- during the YS event was around 4 μm, compared to 0.4–0.9 μm of AYS, indicating a significant amount of SO_4^{2-} and NO_3^- formations on the coarse mode during the long-range transport of Yellow sand. However, the maximum concentration of NH_4^+ was found in the fine particle of diameter less than 2 μm for both YS and AYS samples. These observational findings are quite similar to the measurement results at Yaku island, Japan (Nishikawa et al., 1991) during the Yellow sand event except for the sulfate distribution. The size-resolved composition showed a main peak of sulfate in the coarse mode in this study, while there were bi-modal peaks, one of which was in the fine mode, and the other in the coarse mode at Yaku island.

In most cases, particles with diameters smaller than 1 μm did not increase during the YS event, indicating a lesser influence of dust storms on the masses of fine particles (Zhang and Carmichael, 1999). Parungo et al. (1996) observed that the sulfate distribution on dusty days shows a large fraction of sulfate in the coarse mode. It could be caused by the large amount of cations in the coarse mode providing larger available surface areas for the deposition of sulfate. The observed sulfate distribution of a main peak in the coarse mode in Seoul, Korea during the YS event was similar to that near source region in Beijing, China (Parungo et al., 1996) rather than at Yaku island, Japan (Nishikawa et al., 1991).

The analysis of the equivalent molar ratio for the major inorganic ions (Table 3) showed that non-sea-salt (nss)SO_4^{2-} and NO_3^- concentrations were well correlated with NH_4^+ concentration for the AYS period, whereas they were well correlated with NH_4^+ and Ca^{2+} concentrations during the YS event. The nss-sulfate are estimated based on sodium ion. NH_4^+ was relatively rich compared to nss-SO_4^{2-} in the fine mode, whereas nss-SO_4^{2-} was relatively rich compared to NH_4^+ in the coarse mode (Table 3). Therefore, Ca^{2+} should be added to satisfy the ion balances in the coarse mode. The equivalent ratios of $(NH_4^+)/(\text{nss-}SO_4^{2-} + NO_3^-)$ were 0.8–1.0 in the range of 0.43–1.1 μm of particle diameter, whereas the equivalent ratios of $(NH_4^+ + \text{nss-}Ca^{2+})/(\text{nss-}SO_4^{2-} + NO_3^-)$ were 0.8–1.4 in the range of 2.1–11.0 μm of particle diameter. It suggests that the compounds of these YS aerosols transported to Korea are mainly $(NH_4)_2SO_4$ and NH_4NO_3 in the fine mode, and $CaSO_4$ and $Ca(NO_3)_2$ in the coarse mode.

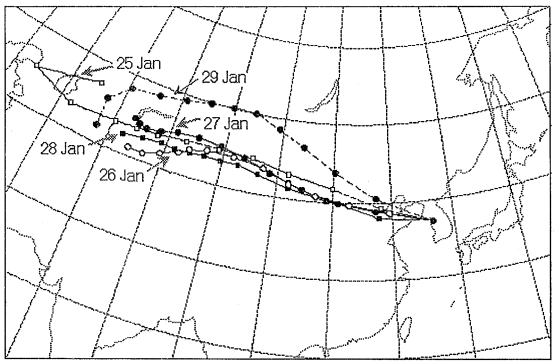


Fig. 3. Backward trajectories of the air mass arrived at Seoul at 0900 LST on 25–29 January 1999 on the isentropic surface of 290 K.

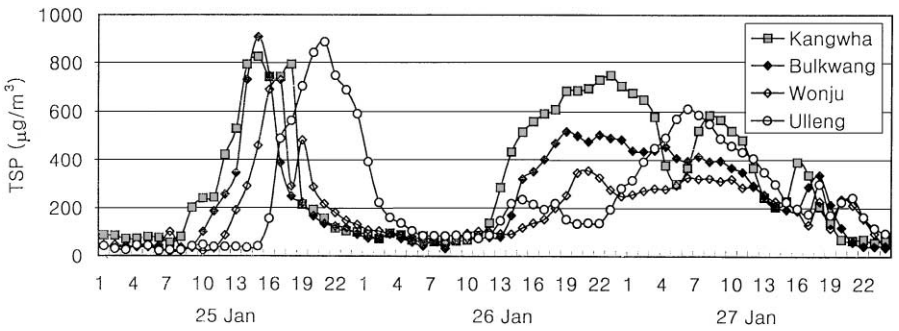


Fig. 4. Temporal variations of TSP concentrations at each site during 25–27 January 1999.

Table 1
Description of monitoring sites

Station	Latitude (N)	Longitude (E)	Analyzing compounds	Aerosol sampling		
				Measuring system	Principle	Sampling interval
Kangwha	37°42′	126°16′	TSP	β-gauge	β-ray absorption	25–27 Jan.[a]
Seoul (Bulkwang)	37°36′	126°56′	TSP Size distribution	β-gauge Cascade impactor	β-ray absorption Inertial impactor	25 Jan.–1 Feb.[b]
Wonju	37°20′	127°56′	TSP	β-gauge	β-ray absorption	25–27 Jan.[a]
Ulleng	37°30′	130°40′	TSP	β-gauge	β-ray absorption	25–27 Jan.[a]
Taean	36°44′	126°08′	TSP PM2.5, PM10	β-gauge Dichotomous AS	β-ray absorption Virtual impactor	25–27 Jan.[a] 7 Jan.–31 Feb.[c]

[a]Continuous monitoring.
[b]2–3 days sampling.
[c]Every six-day sampling.

Table 2
TSP concentrations at Kangwha, Seoul, Wonju, and Ulleng sites and the characters of Yellow sand transport

Contents	Kangwha	Seoul (Bulkwang)	Wonju	Ulleng
Distance from Gobi (km)	2150	2200	2300	2500
Distance from Kangwha (km)	0	50	150	350
First dust plume				
Maximum hourly concentration ($\mu g\,m^{-3}$)	826	908	730	888
Duration (LST) (TSP > 150 $\mu g\,m^{-3}$)	0900–2100 25 Jan.	1100–2000 25 Jan.	1300–2300 25 Jan.	1600 25 Jan.–0300 26 Jan.
Mode occurrence time (LST)	1500 25 Jan.	1500 25 Jan.	1700 25 Jan.	2100 25 Jan.
Plume travelling speed ($km\,h^{-1}$)	—	—	75	67
Dust plume scale[a] (km)	920	710	780	850
Second dust plume				
Maximum hourly concentration ($\mu g\,m^{-3}$)	748	517	355	612
Duration (LST) (TSP >150 $\mu g\,m^{-3}$)	1300 26 Jan.–1800 27 Jan.	1400 26 Jan.–1900 27 Jan.	1700 26 Jan.–2200 27 Jan.	2300 27 Jan.–2200 27 Jan.

[a]Estimated from the average travel speed.

Na^+ and nss-Ca^{2+} were used as tracers of sea-salt and soil components for comparing the relative abundance of sea-salt and soil particles. The equivalent molar ratio of nss-Ca^{2+}/Na^+ during the YS period (1.61) was 5 times larger than that for the AYS period (Table 3). It remarkably increased to 1.5–2.3 especially in the range of 2.1–7.0 μm of particle diameter for the YS event, but less than 0.5 for the AYS period. The increase in the ratio was due to mainly an increase in nss-Ca^{2+}. Ca^{2+}-rich mineral aerosol existed in the coarse mode during the YS event. These high ratios were similar to the result of Zhuang et al. (1999) under the influence of a dust episode in Hong Kong.

3.4. Distribution characteristics of fine and coarse particles

The coarse particle (2.5–10 μm) and the fine particle (less than 2.5 μm) were measured at the Taean site. The mass concentrations of fine and coarse particles on dust days were 46.1 (42.3–50.0) and 221.9 (145.6–298.2) $\mu g\,m^{-3}$ respectively which were significantly increased amounts compared to 15.8 (9.7–21.2) and 24.3 (9.9–49.9) $\mu g\,m^{-3}$ on non-dust days (Table 4 and Fig. 6). The overall mean nss-sulfate concentration in the fine and coarse modes were 5.9 and 3.8 $\mu g\,m^{-3}$, respectively on dust days. Nss-sulfate, nitrate, and calcium concentration in the coarse

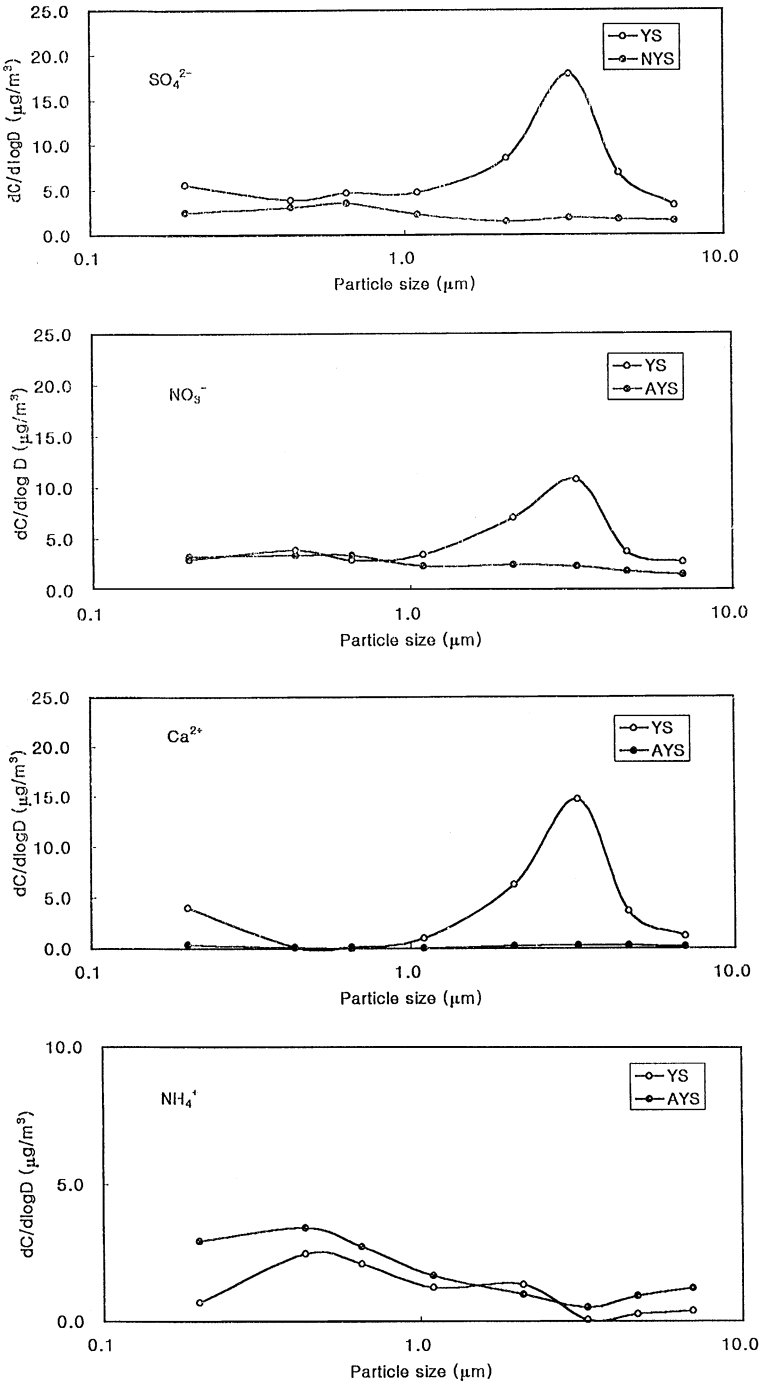


Fig. 5. Particle size distributions of the major ion components (SO_4^{2-}, NO_3^-, Ca^{2+}, and NH_4^+) during the Yellow sand event (25–27 January) and after the YS period (28 January–1 February) at Seoul.

particles increased by a factor of 10.6, 3.8, and 5.2, respectively during the YS period (Table 4). The Na^+ and Cl^- concentrations were very high (3.7 and 5.3 $\mu g\,m^{-3}$, respectively) especially in the coarse mode during this period. In other words, the contribution to coarse particle mass from the sea-salt source could be great. The sea-salt contribution was also very high during the NYS period.

Table 3
Concentrations of major inorganic ions and their equivalent ratios for the YS[a] and AYS[b] periods observed at Seoul

Period	Particle size (μm)	Cl^-	NO_3^-	nss-SO_4^{2-}	Na^+	NH_4^+	nss-Ca^{2+}	nss-Ca^{2+}/Na	NH_4^+/nss-SO_4^{2-}	($NH_4^+ + Ca^{2+}$)/(nss-$SO_4^{2-} + NO_3^-$)
		($\mu g\,m^{-3}$)						Equivalent molar ratio		
YS[a]	− 0.43	0.86	1.00	1.64	0.96	0.23	1.30	1.55	0.38	1.54 (0.25)[c]
	0.43–0.65	0.15	0.70	0.66	0.16	0.44	0.02	0.15	1.79	1.02 (0.98)
	0.65–1.1	0.16	0.68	1.01	0.26	0.48	0.03	0.11	1.27	0.87 (0.83)
	1.1–2.1	0.28	0.97	1.27	0.31	0.35	0.28	1.04	0.73	0.79 (0.46)
	2.1–3.3	0.53	1.38	1.53	0.60	0.26	1.20	2.31	0.46	1.38 (0.27)
	3.3–4.7	0.68	1.65	2.47	1.09	0.01	2.22	2.33	0.01	1.43 (0.01)
	4.7–7.0	0.27	0.64	1.07	0.46	0.05	0.62	1.54	0.11	1.02 (0.08)
	7.0–11.0	0.17	0.53	0.55	0.37	0.07	0.23	0.69	0.36	0.77 (0.21)
	Sum	3.11	7.55	10.20	4.21	1.89	5.90	1.61	0.49	1.19 (0.31)
AYS[b]	− 0.43	0.30	1.10	0.76	0.32	0.97	0.12	0.42	3.40	1.78 (1.60)
	0.43–0.65	0.20	0.61	0.53	0.12	0.62	0.02	0.17	3.06	1.67 (1.63)
	0.65–1.1	0.18	0.78	0.80	0.11	0.63	ND	ND	2.09	1.19 (1.19)
	1.1–2.1	0.15	0.67	0.63	0.12	0.46	0.01	0.12	1.98	1.11 (1.08)
	2.1–3.3	0.12	0.48	0.26	0.15	0.19	0.04	0.30	1.91	0.95 (0.80)
	3.3–4.7	0.11	0.36	0.27	0.12	0.08	0.04	0.41	0.77	0.58 (0.38)
	4.7–7.0	0.12	0.31	0.27	0.11	0.16	0.04	0.48	1.55	1.04 (0.83)
	7.0–11.0	0.10	0.29	0.28	0.13	0.24	0.03	0.30	2.25	1.42 (1.26)
	Sum	1.27	4.59	3.80	1.19	3.34	0.31	0.30	2.34	1.31 (1.21)

[a](YS) 1500 LST 25 January to 1215 LST 27 January 1999.
[b](AYS) 1040 LST 28 January to 1020 LST 1 February 1999.
[c]The values in the parentheses denote the equivalent molar ratios of NH_4^+/(nss-$SO_4^{2-} + NO_3^-$).

Table 4
Comparisons of fine and coarse particles between Yellow sand (YS)[a] and non-Yellow sand (NYS)[b] periods observed at Taean (unit $\mu g\,m^{-3}$)

Size	Content	Mass	Cl^-	NO_3^-	nss-SO_4^{2-}	Na^+	NH_4^+	K^+	Mg^{2+}	Ca^{2+}
Fine	YS	46.1 (42.3–50.0)[c]	0.2 (0.1–0.2)	1.2 (1.0–1.5)	5.9 (4.6–7.4)	0.6 (0.4–0.7)	1.3 (0.5–2.1)	0.5 (0.4–0.6)	0.1 (0.1–0.1)	1.0 (0.5–1.6)
	NYS	15.8 (9.7–21.2)	0.3 (0.1–0.9)	0.5 (0.2–1.4)	2.4 (1.2–4.0)	0.4 (0.1–0.8)	0.9 (0.4–1.8)	0.1 (0.1–0.2)	0.1 (0.0–0.1)	0.1 (0.1–0.2)
	YS/NYS	2.9	0.5	2.6	2.5	1.3	1.5	3.4	1.4	8.5
Coarse	YS	221.9 (145.6–298.2)	5.3 (2.3–8.3)	2.2 (2.0–2.4)	3.8 (2.0–5.7)	3.7 (3.2–4.2)	0.01 (0.01–0.02)	0.2 (0.2–0.3)	0.5 (0.5–0.6)	2.9 (2.8–3.1)
	NYS	24.3 (9.9–49.9)	3.0 (0.2–5.7)	0.6 (0.2–1.0)	0.4 (0.1–1.4)	1.6 (0.3–2.9)	0.02 (0.0–0.09)	0.1 (0.1–0.4)	0.2 (0.0–0.4)	0.6 (0.2–1.8)
	YS/NYS	9.1	1.7	3.8	10.6	2.3	0.5	1.8	2.5	5.2

[a](YS) 25 to 27 January (2 samples).
[b](NYS) 7 January to 18 February six-day period sampling (8 samples).
[c]The values in the parentheses denote minimum and maximum concentrations.

The ratio for the YS period to NYS period of nss-sulfate concentrations was 2.5 in the fine mode and 10.6 in the coarse mode (Table 4). The concentration ratio for all compounds for the period of YS and NYS was larger than 1, except for NH_4^+ ion. Most of NH_4^+ existed in the fine mode, regardless of the periods of YS or NYS. However, maximum concentrations of Cl^- and NO_3^- were mainly distributed in the coarse mode on dusty days.

The fine particle measured at the Taean site on dust days (Table 4) showed that nss-sulfate was relatively rich compared to NH_4^+ so that the equivalent ratio of

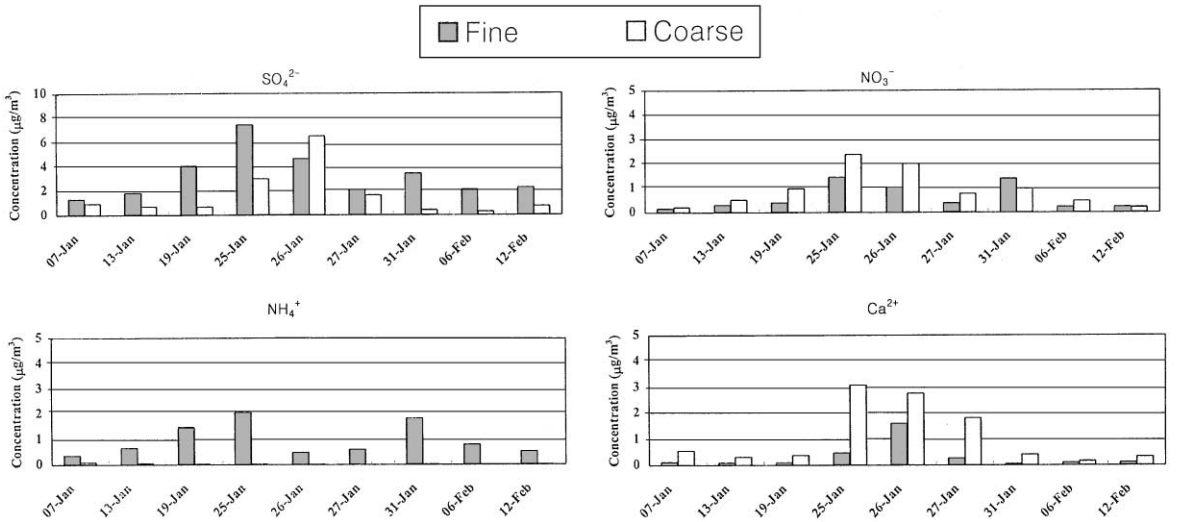


Fig. 6. Temporal variations of SO_4^{2-}, NO_3^-, NH_4^+, and Ca^{2+} concentrations in the fine and coarse modes observed at Taean during January and February 1999.

NH_4^+/nss-SO_4^{2-} was about 0.6. NH_4^+ concentration was so low (0.01 μg m^{-3}) in the coarse mode that Ca^{2+} ion was highly correlated with nss-SO_4^{2-} and NO_3^-. However, nss-SO_4^{2-} and NH_4^+ showed good correlation on non-dusty days. The equivalent molar ratio of NH_4^+/nss-SO_4^{2-} was 1.2 in Taean (KIST, 1999) and 0.9 in Kosan of Cheju island (Kim et al., 1998). It could imply that state of aerosol transported to Taean during the YS period was mainly in the form of $(NH_4)_2SO_4$ in the fine mode, and $CaSO_4$ and $Ca(NO_3)_2$ in the coarse mode. However, the aerosol during the NYS period was generally $(NH_4)_2SO_4$.

3.5. Discussion

Chemical analyses have shown that the observed elevated sulfate and nitrate during Asian dust storms are not associated with dust at the source region (Carmichael et al., 1996; Zhang and Iwasaka, 1999). The dust originated from source regions is likely to be alkaline and may act as an effective sink for acidic gases. SO_4^{2-} and NO_3^- are absorbed and/or formed on the surfaces of wetted dust particles and displace the carbonates during the long-range transport processes (Hirai et al., 1991). Additionally, analyses of precipitation sampled in Korea and Japan show that about 75% of the carbonate is displaced by SO_4^{2-} and NO_3^- by the time when the particles reach Korea and Japan (Nishikawa et al., 1991). Similar features were found in the measurements of cascade impactor in Seoul and Dichotomous sampler in Taean. The YS aerosols mainly consisted of $CaSO_4$ and $Ca(NO_3)_2$ in the coarse mode, which could support the possibility that a significant amount of SO_4^{2-} and NO_3^- were accumulated through the absorption and subsequent conversion of gas phase and substituted for carbonate on the Ca^{2+}-rich mineral dust during the long-range transport. Heterogeneous reactions of SO_2 and HNO_3 occur through fast neutralization reactions with alkaline material such as calcium in dust particles (Okada et al., 1990; Dentener et al., 1996). In fact, the oxidation of S(IV) on wetted dust is essentially limited by gas-phase diffusion in case of pH > 8 (Dentener et al., 1996). The SO_4^{2-} formation due to surface uptake becomes dominant and its mass exhibits a single-mode distribution at a larger dust loading and a larger surface area. The formation of SO_4^{2-} and NO_3^- are mainly determined by the overall heterogeneous loss rate constant, which is largest within the size range of 1.5–10.0 μm (Zhang and Carmichael, 1999).

However, Zhang and Iwasaka (1999) suggested that the sulfate and nitrate were hardly formed on the surface of dust particles during their transport from source region to Beijing. This might be due to very low relative humidity (about 22–27%) and the short traveling distance of dust particles over the low sulfur emission region. This is quite contrast to the present episodic event, in which the relative humidity was relatively high (about 57–66%). and the traveling distance of dust particles over the high sulfur emission region was long.

4. Conclusions

The physical and chemical properties of a wintertime YS event observed during 25–27 January 1999 were investigated. It was a remarkably distinctive episode in the

occurrence time and intensity that had ever been observed in Korea. The physical and chemical characteristics of the winter time YS was not much different from those of the springtime YS. The observed YS event consisted of two types of high-dust plume; one was small but relatively strong intensity and the other was less strong with large horizontal dimension.

SO_4^{2-}, NO_3^-, Ca^{2+} and Na^+ were the dominant ion components with the concentration of 11.3, 7.6, 6.1 and 4.2 $\mu g\,m^{-3}$ during the passage of YS, compared to the corresponding concentrations of 4.1, 4.6, 0.4, and 1.1 $\mu g\,m^{-3}$ after the passage of YS over Korea. The size-resolved composition in Seoul Korea during the YS event showed a main peak of sulfate in the coarse mode. The equivalent molar ratio of nss-Ca^{2+}/Na^+ remarkably increased to 1.5–2.3 especially in the range of 2.1–7.0 μm of particle diameter during the YS event. Ca^{2+}-rich mineral aerosol existed in the coarse mode during the YS event. The YS aerosols transported to Korea were mainly composed of $CaSO_4$ and $Ca(NO_3)_2$ in the coarse mode. A significant amount of SO_4^{2-} and NO_3^- could be formed through the absorption and subsequent conversion of SO_2 and NO_x on the mineral dust during the long-range transport of Yellow sand.

The advanced program for the well-coordinated measurements will be necessary to obtain a consistent data set for elaborate physical and chemical properties of YS at targeted locations such as in the dust source region, in the downstream region, and in the remote marine environment, which could consequently help to understand the characteristics of YS.

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References

ACE-ASIA, 1999 Radiative forcing due to anthropogenic aerosols over the Asian Pacific region. Science and Implementation Plan for the ACE-ASIA Network Studies (Draft).

Carmichael, G.R., Zhang, Y., Chen, L.-L., Hong, M.S., Ueda, H., 1996. Seasonal variation of aerosol composition at Cheju island, Korea. Atmospheric Environment 30, 2407–2416.

Dentener, F.J., Carmichael, G.R., Zhang, Y., Lelieveld, J., Crutzen, P.J., 1996. Role of mineral aerosol as a reactive surface in the global troposphere. Journal of Geophysical Research 101 (D17), 22869–22889.

Gao, Y., Arimoto, R., Duce, R.A., Lee, D.S., Zhou, M.Y., 1992. Input of atmospheric trace elements and the mineral matter to the Yellow sea during the spring of a low-dust year. Journal of Geophysical Research 97, 3767–3777.

Hirai, E., Miyazaki, M., Chohji, T., Lee, M., Kitamura, M., Yamaguchi, K., 1991. Effect of Kosa aerosol on inorganic components in rainwater collected from Circum-Pan-Japan-Sea Area. Proceedings of the Second IUAPPA Regional Conference on Air Pollution, Seoul, Korea, Vol II, pp. 27–34.

Iwasaka, Y., Yamato, M., Imasu, R., Ono, A., 1988. Transport of Asian dust(KOSA) particles: importance of weak KOSA events on the geochemical cycle of soil particles. Tellus 40B, 494–503.

Jaff, D.A., Anderson, T., Covert, D., Kotchenruther, R., Trost, B., Danielson, J., Simpson, W., Berntsen, T., Karlsdottir, S., Blake, D., Harris, J., Carmichael, G., Uno, I., 1999. Transport of Asian air pollution to North America. Geophysical Research Letters 26, 711–714.

Kang, K.H., and Sang, S.E., 1991. Influence of Yellow sand on TSP in Seoul. Proceedings of the Second IUAPPA Regional Conference on Air Pollution, Seoul, Korea, Vol 2, International Union of Applied and Pure Physics, Seoul Korea, pp. 1–6.

Kim, Y.P., Shim, S.G., Moon, K.C., Hu, C.G., Kang, C.H., Park, K.Y., 1998. Monitoring of air pollutants at Kosan, Cheju island, Korea, during March–April 1994. Journal of Applied Meteorology 37, 1117–1126.

Korean Institute of Science and Technology, 1999. A survey on the long-range transport pollutants and environmental conservation in Northeast Asia (IV). 494pp.

Korean Meteorological Agency (KMA), Korea, 1999. Monthly Meteorological Data Report of January, 1999 (in Korean).

Ministry of Environment (MOE) Korea, 1999. Monthly Air Pollution Data Report of January, 1999 (in Korean).

Nishikawa, M., 1993. Environmental effects of Kosa aerosol. Journal of Environmental Chemistry 3, 673–682.

Nishikawa, M., Kanamori, S., Kanamori, N., Misoguchi, T., 1991. Environmental significance of Kosa aerosol (yellow sand dust) collected in Japan. Proceedings of the Second IUAPPA Regional Conference on Air Pollution, Seoul, Korea, 4 September, pp. 35–41.

Okada, K., Narus, H., Tanaka, T., Nemoto, O., 1990. X-ray spectrometry of individual Asian dust-storm particles over the Japanese islands and the North Pacific Ocean. Atmospheric Environment 23A, 1369–1378.

Parungo, F., Kim, Y., Zhu, C., Harris, J., Schnell, R., Li, X., Yang, D., Fang, X., Yan, P., Yu, X., Zhou, M., Chen, Z., Qian, F., Park, K., 1996. Asian dust storms and their effects on radiation and climate Part II. STC Technical Report 2959, 34pp.

Prospero, J.M., Savoie, D.L., 1989. Nitrate concentrations over the Pacific: oceanic background and continental impacts. Nature 339, 687–689.

Pye, K., 1987. Aeolian Dust and Dust Deposits. Academic Press, New York, 334pp.

Uematsu, M., Duce, R.A., Prospero, J.M., Chen, L., Merrill, J., Macdonald, R.L., 1983. Transport of mineral aerosol from Asia over the North Pacific Ocean. Journal of Geophysical Research 88, 5343–5352.

UNDP, 1996. United nations Development Program, Air Pollution Control Program, CPR/96/304, China International

Center for Economic and Technical Exchanges, Beijing, P.R. China.

Zhang, X., Arimoto, R., An, Z., Chen, T., Zhang, G., Zhu, G., Wang, X., 1993. Atmospheric trace elements over source regions for chinese dust: concentrations, sources and atmospheric deposition on the loess plateau. Atmospheric Environment 27, 2051–2067.

Zhang, X.Y., Arimoto, R., An, Z.S., 1997. Dust emissions from Chinese desert sources linked to variations in atmospheric circulation. Journal of Geophysical Research 102, 28041–28047.

Zhang, Y., Carmichael, G.R., 1999. The role of mineral aerosol in tropospheric chemistry in East Asia - a model study. Journal of Applied Meteorology 38, 353–366.

Zhuang, H., Chan, C.K., Fang, M., Wexler, A.S., 1999. Formation of nitrate and non-sea-salt surface on coarse particles. Atmospheric Environment 33, 4223–4233.

Zhang, D., Iwasaka, Y., 1999. Nitrate and sulfate in individual Asian dust-storm particles in Beijing, China in spring of 1995 and 1996. Atmospheric Environment 33, 3213–3223.