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# Change in size distribution and chemical composition of kosa (Asian dust) aerosol during long-range transport

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## Abstract

Kosa (Asian dust) aerosol was sampled at eight locations in China and Japan along the route of the dust event in March 2001. Mass concentrations, concentrations of the chemical components in the aerosol, and the size distribution of the aerosol were examined. The mass concentration of the aerosol was  $6700 \mu\text{g m}^{-3}$  in the Chinese interior (in and around the Inner Mongolia Autonomous Region);  $1500 \mu\text{g m}^{-3}$  at Beijing, China; and  $230 \mu\text{g m}^{-3}$  at a remote island in Japan. The concentrations dropped by one order of magnitude as the dust was transported from the interior of China to Japan. As with the mass concentrations, the concentrations of crustal elements in the aerosol, e.g., Al, Fe, etc., followed this same trend and decreased by one order of magnitude or more as the dust made its way from the Chinese interior to Japan. Coefficients of determination ( $r^2$ ) were greater than 0.9 for all linear regressions of crustal element concentration on Al concentration in the size-separated coarse particles larger than  $2.1 \mu\text{m}$  in aerodynamic diameter, for samples from both China and Japan. This indicated that the concentrations of all the crustal elements in coarse particles were constant and did not depend on particle size. However, the concentration of  $\text{NO}_3^-$  was higher in Japan (e.g., a remote island sampling site:  $5.8 \mu\text{g m}^{-3}$ ) than in China (e.g., an inland sampling site:  $1.7 \mu\text{g m}^{-3}$ ). Coefficients of determination ( $r^2$ ) were greater than 0.9 for linear regressions of  $\text{NO}_3^-$  concentrations on the  $[\text{Al}_{\text{mass}}/D]$ , where  $\text{Al}_{\text{mass}}$  is the Al concentration and  $D$  is representative diameter, for size-separated particles larger than  $2.1 \mu\text{m}$  in diameter both in China and Japan. This indicated that  $\text{NO}_3^-$  was attached to the surface of the kosa aerosol particles.

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**Keywords:** Yellow sand; Dust storm; Nitrate; Sulphate; Heavy metal

## 1. Introduction

Kosa aerosol, generated when the surface soil in the arid region of the Asian continental landmass is lifted by winds, is the major mineral aerosol transported from east Asia to the Pacific region. The frequency and scale of dust events giving rise to kosa aerosols has increased rapidly in the east Asian region since 2000 (Nishikawa and Mori, 2003). This fact should not be ignored when the solar radiation balance for the region is calculated.

An understanding of the behaviour of these dust events will lead to the construction of a more accurate solar radiation balance model (Sokolik et al., 2001). In addition, since large amounts of kosa aerosol are transported over long distances, large amounts of crustal elements that comprise the aerosol are also being transported. It then becomes important to understand the behaviour of kosa aerosols from the viewpoint of geochemical cycles.

Research into kosa aerosols has been conducted by several research groups using the area from east Asia to the Pacific region as the field of study. Chemical composition and the concentrations of the components

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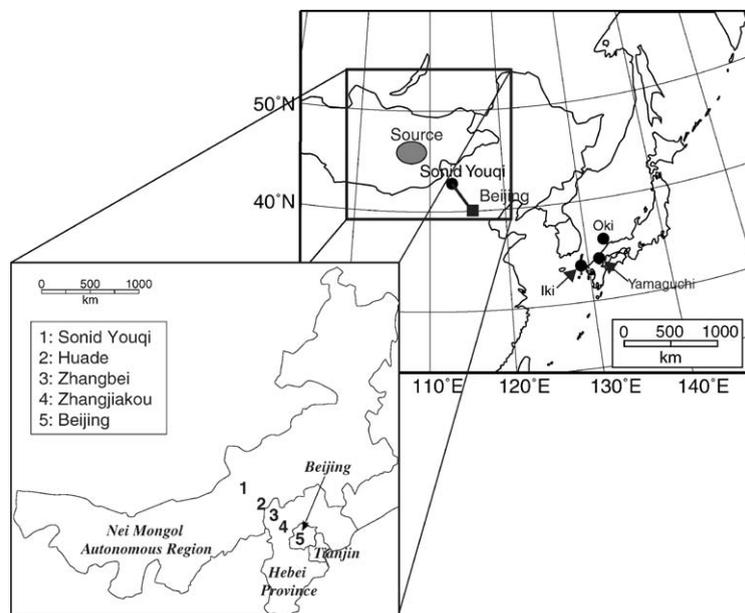


Fig. 1. Aerosol sampling locations.

for individual kosa events (Uematsu et al., 1983; Choi et al., 2001), the vertical distribution and concentration of aerosols (Murayama et al., 2001) and estimates of the dispersion of the kosa layer using model calculations (Uno et al., 2001) or satellite data (Husar et al., 2001) have been reported. Mori et al. (2002) investigated the concentration of chemical components in a kosa aerosol sampled along its transport route, and suggested that significant amounts of nitrate entered the air mass containing the kosa aerosols from outside the system during long-range transport from the source to Japan. However, it is not known whether this supplemental  $\text{NO}_3^-$  was: (a) attached to the kosa particles; (b) had reacted with the kosa particles; or (c) was merely suspended in the air mass where kosa aerosols were also suspended. Electron microscopy revealed soil particles coated by water or by a solution containing  $\text{SO}_4^{2-}$  (Iwasaka et al., 1988) and also showed that mineral aerosol particles initially mixed with sea salt (Fan et al., 1996). These results showed the possibility that the  $\text{NO}_3^-$  was attached to the kosa particles and/or had reacted with the kosa particles. If most kosa aerosol particles are covered with substances including  $\text{NO}_3^-$  or have reacted with substances including  $\text{NO}_3^-$ , it will be necessary to re-evaluate the influence of such a modified kosa aerosol on the radiation balance and the impact of deposited kosa aerosols on ecosystems.

Here we discuss how kosa aerosols and supplemental substances behaved in the same air mass based on the size distributions of the chemical components in kosa aerosols sampled along their transport routes.

## 2. Experiment

In February 2001 we established a kosa aerosol monitoring network comprising 21 stations in China and four stations in Japan to monitor kosa aerosols along their transport routes from the Chinese interior to Japan. A typical kosa event was observed within the sampling network during 21–24 March 2001 (UT), and we succeeded in collecting kosa aerosol samples at eight stations along the transport route (Fig. 1). Aerosol was collected using a high-volume sampler with a quartz fibre filter (2500QAT-UP, Pallflex), and a low-volume sampler with a membrane filter (FM-80, Fuji Film). Both samplers collected the total suspended particles (TSP). An Andersen sampler with a quartz fibre filter (2500QAT-UP, Pallflex) was used to collect size-separated aerosol samples at both Beijing and Yamaguchi. The 50% cut-off points for the impaction stages were 11, 7.0, 4.7, 3.3, 2.1, 1.1, 0.65, and 0.43  $\mu\text{m}$  aerodynamic diameter. Details of the samples are summarised in Table 1. The procedures to determine the aerosol mass concentrations and the concentrations of chemical components in the aerosols, and the trajectory analysis have been described in Mori et al. (2002).

## 3. Results and discussion

### 3.1. Transport route of the kosa aerosol

The kosa event was observed at each monitoring station within the sampling network from 21 to

Table 1  
Aerosol sampling locations and methods

Station name and grouping	Latitude (°N)	Longitude (°E)	Date (UT)	Sampler
Sonid Youqi Inland	42.5	113	3/21/01 0:30–3/21/01 8:30	High-volume sampler
Huade Inland	42	114	3/21/01 1:20–3/21/01 9:36	High-volume sampler
Zhangbei Inland	41	114.5	3/21/01 3:40–3/21/01 9:44	High-volume sampler
Zhangjiakou Loess, Urban	41	115	3/21/01 3:40–3/21/01 10:15	High-volume sampler
Beijing Urban	40	116.5	3/21/01 10:15–3/21/01 16:15	High-volume sampler
			3/20/01 8:40–3/21/01 14:30	Andersen sampler
Iki Remote, Island	34	130	22/3/01 9:45–23/3/01 9:00	High-volume sampler
Oki Remote, Island	36	133	22/3/01 4:00–23/3/01 4:00	Low-volume sampler
Yamaguchi Urban	34	131.5	22/3/01 2:00–23/3/01 1:57	Low-volume sampler
			22/3/01 2:00–24/3/01 2:00	Andersen sampler

24 March 2001 (UT). Forward and backward trajectory analyses were conducted at 11:00 (UT) 21 March 2001 with the starting point 3000 m over Beijing (40°N, 116.5°E) (Fig. 2). The results of the backward trajectory analysis showed that the air mass present at 3000 m over Beijing on 11:00 (UT) 21 March 2001 came from Mongolia. The results of the forward trajectory analysis showed that the same air mass passed over the Korean peninsula on 22 March 2001, and then passed over Japan during 22–25 March 2001. It is possible to say that all aerosol samples shown in Table 1 were indeed collected along the course of this particular dust event.

### 3.2. Change in chemical composition of kosa aerosol during long-range transport

Table 2 shows the mass concentration and chemical component concentrations in the aerosol sampled during the dust event. The values in the “inland” column are the average values for three inland stations (Sonid Youqi, Huade, Zhangbei), and the values in the “Remote Island” column are the average values for two remote island stations (Iki, Oki). We adopted the non-sea salt value for  $F^-$ ,  $SO_4^{2-}$ , Na, Mg, K, Ca, and Sr in the aerosol collected in Japan since aerosols collected in Japan have always had a high proportion of sea salt (Fan et al., 1996). We assumed that all  $Na^+$  in the aerosol samples collected in Japan was derived from sea salt particles and used the method described by Nishikawa et al. (1991a) to calculate the non-sea salt concentrations of those seven components. The concentration of TSP in the interior of China decreased to 1/30 as the aerosol was transported to Japan (Table 2). The concentrations of Na, Mg, Al, P, K, Ca, Sc, Ti, V, Mn, Fe, Co, Ni, Sr, Y, Ba, La in the aerosol, as well as the mass concentration of the aerosol itself, decreased to 1/10–1/40 as the aerosol was transported from the interior of China to Japan. Those components for which the concentrations decreased by an order of magnitude or more as the aerosol was transported from the interior of China to Japan were all crustal elements. However,

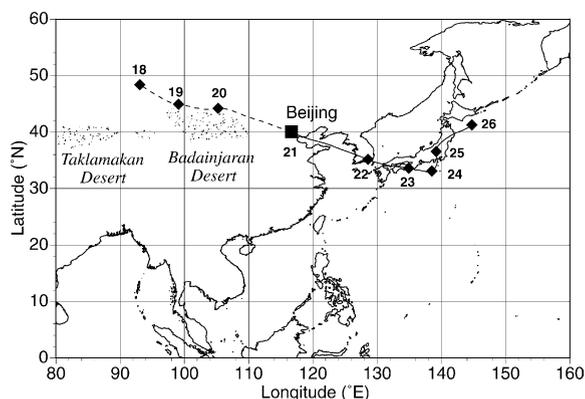


Fig. 2. Results of the backward (dashed line) and forward (solid line) trajectory analyses conducted for 11:00 UT 21 March 2001 with the starting point 3000 m over Beijing (40°N, 116.5°E). The diamond symbols indicate the positions of the air masses at 11:00 (UT) each day.

concentrations of  $NO_3^-$  in the aerosol were greater at the various sampling stations in Japan than in China. Although the concentrations of  $F^-$ ,  $SO_4^{2-}$ ,  $NH_4^+$ , Zn, and Pb decreased as the kosa aerosol was transported from China to Japan, the reduction was a mere  $\frac{1}{2}$  to  $\frac{1}{6}$ .

We calculated the ratios of the concentrations of each chemical component (X) to the concentration of Al which is often used as an indicator for a mineral aerosol (Uematsu et al., 1983; Duce, 1995) (Fig. 3). The ratio  $X/Al$  of Al, Sc, La, Fe, Co, Mn, Ba, Ti, K, P, Y, Mg, Sr, V, Ni, Ca, Na, taken at all the stations (or average values for the “inland” and “remote island” stations), to the average value for the five station or groups of stations (shown in Table 1) ( $[X/Al]_{av}$ ) was always between 0.5 and 1.5, i.e.,  $0.5 < [X/Al]/[X/Al]_{av} < 1.5$ , showing  $[X/Al]$  was nearly constant regardless of the station at which the aerosol was sampled (Fig. 4). The result indicated that supplementation to the aerosol from other sources for these 17 crustal elements during long-range transport was likely to be negligibly small. This conclusion also agreed with the findings of Mori et al. (2002) for a dust

Table 2

Mass concentrations ( $\mu\text{g m}^{-3}$ ) and chemical component concentrations ( $\mu\text{g m}^{-3}$ ) in the aerosol collected along the route of the dust event in March 2001

	China			Japan	
	Inland	Loess/Urban (Zhangjiakou)	Urban China (Beijing)	Remote island	Urban Japan (Yamaguchi)
TSP	6700	4500	1500	230	200
F <sup>-a</sup>	0.219	0.263	0.219	0.056	0.076
SO <sub>4</sub> <sup>2--a</sup>	30.5	40.8	18.4	10.3	16.0
NO <sub>3</sub> <sup>-</sup>	1.7	1.3	1.4	5.8	10.8
Na <sup>+</sup>	8.5	21.7	5.8	5.2	2.3
NH <sub>4</sub> <sup>+</sup>	4.76	2.82	2.54	0.79	2.29
Na <sup>a</sup>	56.6	40.3	14.7	1.4	1.5
Mg <sup>a</sup>	84.0	41.2	21.5	3.2	4.5
Al	412	183	95	12	16
P	5.74	2.75	1.35	0.22	0.21
K <sup>a</sup>	122	54	28	4	6
Ca <sup>a</sup>	142	66.9	43.6	8.9	10.3
Sc	0.0757	0.0360	0.0180	0.0023	0.0028
Ti	26.7	12.1	6.1	1.0	1.1
V	0.555	0.254	0.130	0.020	0.033
Mn	4.99	2.25	1.21	0.18	0.21
Fe	216	98	51	7	9
Co	0.110	0.052	0.026	0.003	0.005
Ni	0.221	0.103	0.051	0.009	0.016
Zn	0.733	0.372	0.274	0.116	0.309
Sr <sup>a</sup>	1.23	0.59	0.32	0.05	0.06
Y	0.164	0.076	0.038	0.004	0.005
Ba	3.26	1.57	0.87	0.11	0.15
La	0.220	0.096	0.055	0.007	0.008
Pb	0.149	0.207	0.046	0.070	0.173

<sup>a</sup>The sea salt contribution was subtracted for the Japan sample concentrations only.

event in April 1998, which showed that the supplementation of the crustal elements during the long-range transport of the aerosol could be negligibly small. However, the ratio of the concentration of NO<sub>3</sub><sup>-</sup> to Al was two orders of magnitude greater in Japan (0.46–0.69) than in China (0.004–0.015) (Fig. 3), and the [NO<sub>3</sub><sup>-</sup>/Al]/[NO<sub>3</sub><sup>-</sup>/Al]<sub>av</sub> was 0.018–2.9 (Fig. 4). In contrast to the crustal element to Al concentration ratios, the NO<sub>3</sub><sup>-</sup>/Al ratio varied greatly depending on the sampling station. The ratios of the concentrations of F<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup>, Zn, and Pb to that of Al were one order of magnitude larger in Japan than in China (Fig. 3). The range for the [X/Al]/[X/Al]<sub>av</sub> values was also larger than the crustal element range (Fig. 4), and the ratios of the concentration of F<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup>, Zn, Pb to that of Al differed depending on the aerosol sampling stations. From these results it can be deduced that there were different sources of NO<sub>3</sub><sup>-</sup>, F<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, NH<sub>4</sub><sup>+</sup>, Zn, and Pb, and that these were different from the source of the crustal elements in the aerosol along its transport route.

### 3.3. Size distribution of crustal elements in the aerosol collected during the kosa event

The mass size distribution of the aerosol collected in Beijing during the dust event had one major peak in the coarse particle size range at 4.7–7.0  $\mu\text{m}$  (Table 3, Fig. 5a). Ninety-three per cent of the total mass originated from coarse particles larger than 2.1  $\mu\text{m}$  in aerodynamic diameter (Table 4). This characteristic of the size distribution was similar to that of an aerosol observed during a previous dust event in Japan (Nishikawa et al., 1991b). The mass size distribution for the aerosol collected at Yamaguchi during the dust event had two peaks, one at 0.43–0.65  $\mu\text{m}$  and the other at 3.3–4.7  $\mu\text{m}$  (Table 3, Fig. 5b). Sixty-four per cent of the total mass originated from coarse particles larger than 2.1  $\mu\text{m}$  in diameter (Table 4). This was also similar to an aerosol that had been observed during a previous dust event in Japan (Kadowaki, 1979; Mizohata and Ito, 1995). The diameter of particles with the highest mass concentration was smaller in the aerosol sampled at

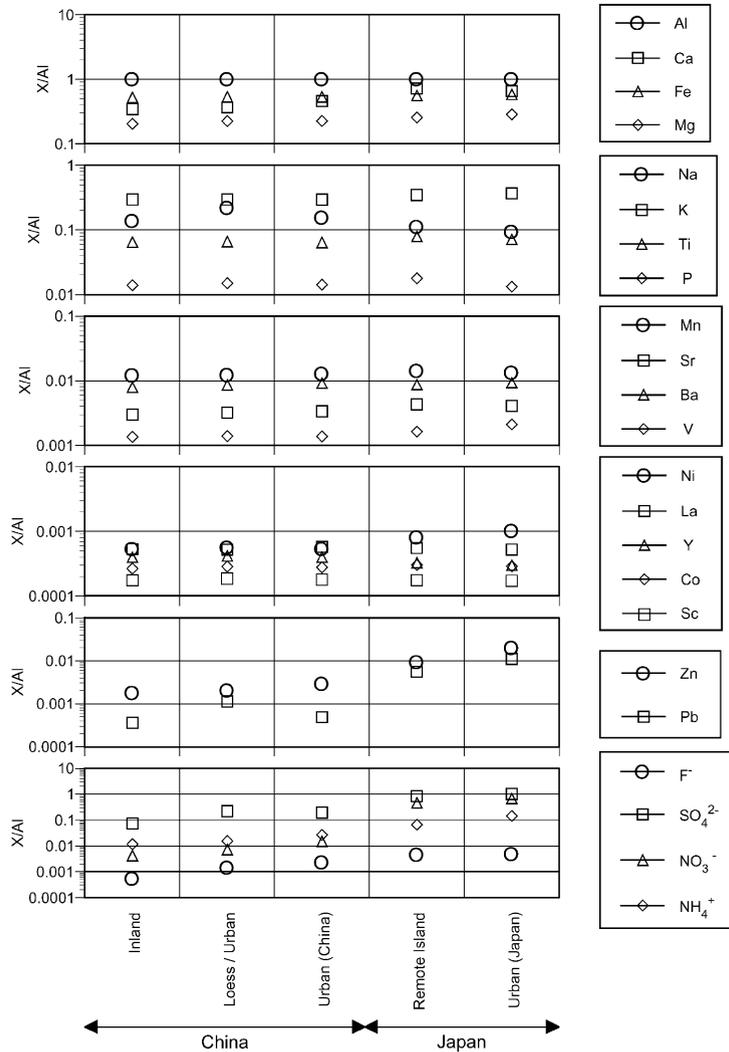


Fig. 3. Ratios of the concentrations of various chemical components to the concentration of Al in the aerosol collected in China and Japan during the dust event in March 2001.

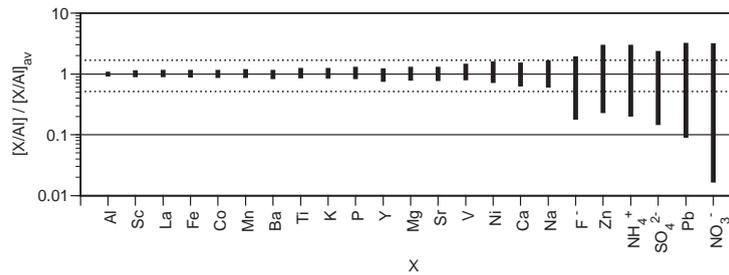


Fig. 4. Range of the  $[X/AI]/[X/AI]_{av}$  ratio (averaged over a total of five areas in China and Japan). The dotted line in the figure indicates the values for  $[X/AI]/[X/AI]_{av}$  is 0.5 or 1.5.

Yamaguchi, far from the source, than at Beijing, much closer to the source. These results accorded with the description in Duce (1995).

The size distributions of the crustal elements (Mg, Al, K, Ca, Ti, V, Mn, Fe, Sr, Ba) in the aerosol collected at Beijing were one-modal. They had a peak between 4.7

Table 3

Summary of size distribution for the mass concentration and each chemical component concentration in the aerosol sampled at Beijing and Yamaguchi during the dust event in March 2001

Size range	Fine				Coarse				
	50% cutoff diameter ( $\mu\text{m}$ )	0.43	0.65	1.1	2.1	3.3	4.7	7.0	11
Stage no.	BF	8	7	6	5	4	3	2	1
Beijing									
Mass							P		
F <sup>-</sup>									P
SO <sub>4</sub> <sup>2-</sup>		P			<				P
NO <sub>3</sub> <sup>-</sup>			P		<	P			
NH <sub>4</sub> <sup>+</sup>		P			>	P			
Mg	—	—	—	—					P
Al	—	—	—	—					P
K	—	—	—	—					P
Ca	—	—	—	—					P
Ti	—	—	—	—					P
V	—	—	—	—					P
Mn	—	—	—	—					P
Fe	—	—	—	—					P
Zn	—	—	P	—	>				P
Sr	—	—	—	—					P
Ba	—	—	—	—					P
Pb	—	—	P	—		—	—	—	—
Yamaguchi									
Mass		P			<	P			
F <sup>-a</sup>	—	—	—	—		—	—	—	—
SO <sub>4</sub> <sup>2-a</sup>	—	P	—	—	>	—	P	—	—
NO <sub>3</sub> <sup>-</sup>	—	—	—	—		—	P	—	—
NH <sub>4</sub> <sup>+</sup>	—	P	—	—		—	—	—	—
Mg <sup>a</sup>	—	—	—	—		—	P	—	—
Al	—	—	—	—		—	P	—	—
K <sup>a</sup>	—	—	—	—		—	—	—	—
Ca <sup>a</sup>	—	—	—	—		—	P	—	—
Ti	—	—	—	—		—	P	—	—
V	—	—	—	—		—	—	—	—
Mn	—	—	—	—		—	P	—	—
Fe	—	—	—	—		—	P	—	—
Zn	—	—	P	—		—	—	—	—
Sr <sup>a</sup>	—	—	—	—		—	P	—	—
Ba	—	—	—	—		—	—	—	—
Pb	—	—	—	—		—	—	—	—

P: Stage within that size range (fine or coarse) containing the highest concentration of that chemical species.

—: Stage within that size range (fine or coarse) in which the concentration of the chemical species was below the limit of determination. The “<” or “>” indicates which of two peak concentrations for that chemical species (in the fine or in the coarse range) is the greater.

<sup>a</sup>The sea salt contribution was subtracted.

and 7.0  $\mu\text{m}$  aerodynamic diameter (Table 3). The concentrations of the crustal elements [X] in size-separated coarse particles larger than 2.1  $\mu\text{m}$  were plotted against the concentration of Al [Al<sub>mass</sub>] in particles of the same size range, and straight lines fitted

by the least-squares method (Fig. 6). The  $r^2$  values (coefficients of determination) for these linear relationships ( $[X] = a + b[\text{Al}_{\text{mass}}]$ ) were always more than 0.9 (Table 5). This showed that the chemical composition of all the crustal elements in particles larger than 2.1  $\mu\text{m}$

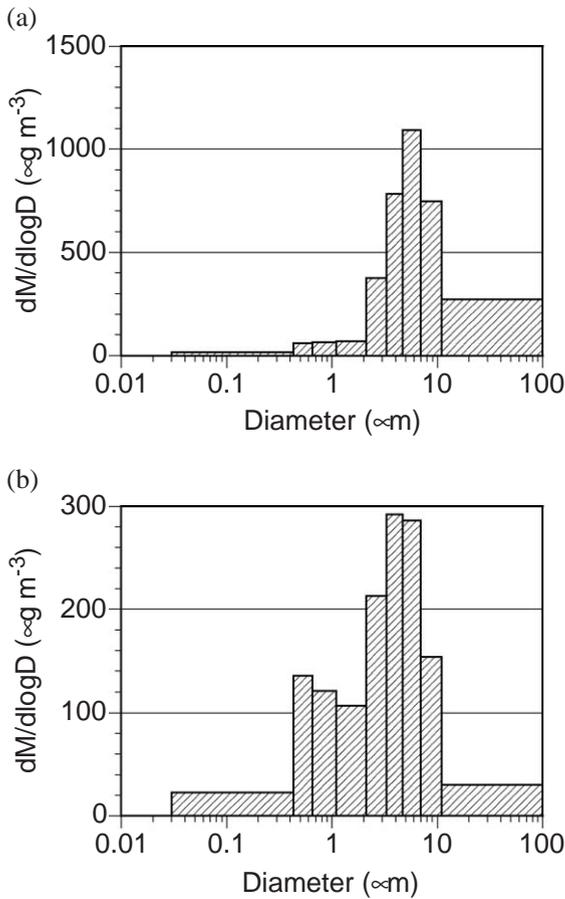


Fig. 5. Mass size distribution of the aerosol collected at Beijing (a) and Yamaguchi (b) during the March 2001 dust event.

was constant and did not depend on particle size. This strong uniformity in the coarse particle range indicated that all the crustal elements originated from the same source.

The size distribution for the crustal element concentrations in the aerosol collected at Yamaguchi had one peak between 3.3 and 4.7  $\mu\text{m}$  (Table 3). The particle diameter with the maximum concentrations of the crustal elements in the aerosol collected at Yamaguchi was smaller than for that collected at Beijing. The  $r^2$  values for  $[X] = a + b[\text{Al}_{\text{mass}}]$  for all the crustal elements were 0.9 (Table 5). Just as was the case for the Beijing sample, the composition of all the crustal elements in particles larger than 2.1  $\mu\text{m}$  in aerodynamic diameter was constant and did not depend on particle size. This again strongly indicated that these elements came from the same source.

Although the particle diameter of maximum concentrations of the crustal elements in the aerosol collected at Yamaguchi was smaller than that collected at Beijing, the slope values ( $b$ ) for the  $[X] = a + b[\text{Al}_{\text{mass}}]$  relation-

ships for both Beijing and Yamaguchi were almost identical. The average values of  $X/\text{Al}$  for the bulk aerosol collected at each sampling station in China and Japan were nearly the same in each case (Table 5). These results indicated that, although the particle sizes changed during long-range transport from China to Japan, the chemical composition of the crustal elements in the coarse particles remained nearly constant, and was also nearly constant between the bulk aerosol (TSP) samples and aerosol particles larger than 2.1  $\mu\text{m}$  only.

### 3.4. Size distribution of $\text{NO}_3^-$ , $\text{SO}_4^{2-}$ , $\text{F}^-$ , $\text{NH}_4^+$ , $\text{Zn}$ , and $\text{Pb}$ in the aerosol collected during the kosa event

The concentration of  $\text{NO}_3^-$  in the bulk aerosol increased during its long-range transport from the interior of China to Japan as mentioned in the Section 3.2. The concentration detected in coarse particles greater than 2.1  $\mu\text{m}$  in the Beijing sample was 52%, which increased to 86% in the Yamaguchi sample (Table 4). The  $r^2$  value for  $[X] = a + b[\text{Al}_{\text{mass}}]$  (with X, in this case, the  $\text{NO}_3^-$  concentration) was smaller than those for the crustal elements (Table 5, Fig. 7). This indicated that the  $\text{NO}_3^-$  in the coarse particles was, in contrast to the crustal elements, not uniformly distributed within the aerosol. The  $r^2$  values for  $[\text{NO}_3^-] = a + b[\text{Al}_{\text{mass}}/D]$ , however, was high for both China and Japan (Fig. 7). The  $[\text{Al}_{\text{mass}}/D]$  value is proportional to the surface area of the kosa particles, assuming that a kosa particle is a spherical particle, as shown below:

$$\begin{aligned} [\text{Kos}_{\text{SA}}]_i &= \pi D_i^2 \\ &= \frac{6}{D_i} [\text{Kos}_{\text{v}}]_i \\ &= \frac{6}{D_i} \frac{[\text{Kos}_{\text{mass}}]_i}{\rho} \\ &= \frac{6}{D_i} \frac{C[\text{Al}_{\text{mass}}]_i}{\rho} \\ &= \frac{6C}{\rho} \frac{[\text{Al}_{\text{mass}}]_i}{D_i}, \end{aligned}$$

$$\frac{6C}{\rho} = \text{constant} \text{ (g}^{-1} \text{ m}^3\text{)}.$$

$$\text{Therefore } [\text{Kos}_{\text{SA}}]_i \propto \frac{[\text{Al}_{\text{mass}}]_i}{D_i},$$

where  $[\text{Kos}_{\text{SA}}]$ : surface area of kosa particles, subscript  $i$ : particle size range  $i$ ,  $D$ : representative diameter of kosa aerosols,  $[\text{Kos}_{\text{v}}]$ : volume of kosa particles,  $[\text{Kos}_{\text{mass}}]$ : mass concentration of kosa particles,  $\rho$ : specific gravity of kosa particles,  $C$ : Al content of a kosa particle,  $[\text{Al}_{\text{mass}}]$ : Al concentration. Therefore, the high  $r^2$  value for the linear regression ( $[\text{NO}_3^-] = a + b[\text{Al}_{\text{mass}}/D]$ ) for both China and Japan (Fig. 7) means a good correlation between  $\text{NO}_3^-$  and the surface area of the kosa particle. This indicated that  $\text{NO}_3^-$  was attached to the surface of

Table 4

Summary of the mass concentrations ( $\mu\text{g m}^{-3}$ ) and the concentrations ( $\mu\text{g m}^{-3}$ ) of chemical components in the size-separated aerosols collected at Beijing and Yamaguchi during the March 2001 dust event

	Beijing		Yamaguchi	
	Fine ( $\leq 2.1 \mu\text{m}$ )	Coarse ( $\geq 2.1 \mu\text{m}$ )	Fine ( $\leq 2.1 \mu\text{m}$ )	Coarse ( $\geq 2.1 \mu\text{m}$ )
Mass	60	790	109	195
F <sup>-a</sup>	0.074	0.475	—	—
SO <sub>4</sub> <sup>2--a</sup>	6.2	12.5	8.9	4.4
NO <sub>3</sub> <sup>-</sup>	3.62	3.92	0.95	6.06
NH <sub>4</sub> <sup>+</sup>	2.97	1.39	2.69	—
Mg <sup>a</sup>	—	11.9	—	2.0
Al	—	43.7	—	6.3
K <sup>a</sup>	—	11.1	—	—
Ca <sup>a</sup>	0.6	31.5	—	5.7
Ti	—	2.91	—	0.40
V	—	0.060	—	—
Mn	0.067	0.588	—	0.088
Fe	0.8	25.4	0.4	3.9
Zn	0.24	0.17	0.10	—
Sr <sup>a</sup>	—	0.199	—	0.031
Ba	—	0.289	—	—
Pb	0.150	—	—	—

—: The concentrations for certain size ranges were below the limit of determination.

<sup>a</sup>Sea salt contributions were subtracted from Yamaguchi values.

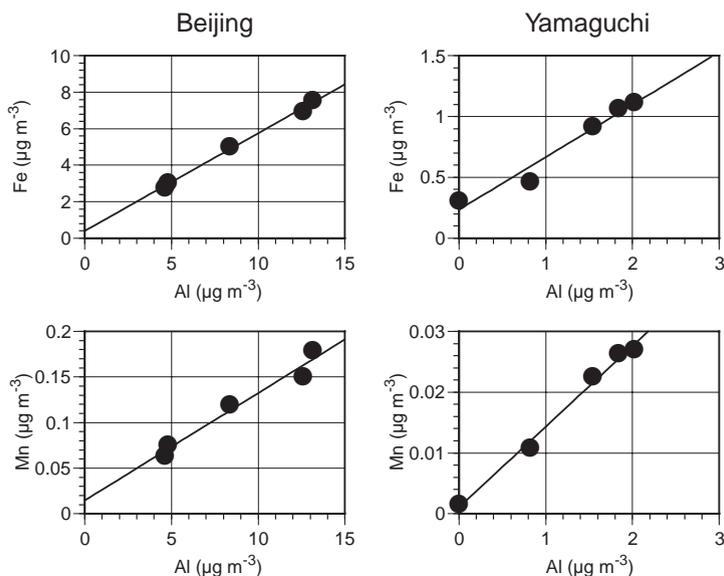


Fig. 6. Concentrations of Fe and Mn in the size-separated aerosol larger than  $2.1 \mu\text{m}$  in aerodynamic diameter plotted against Al concentration in the similar size range aerosol. Straight lines were fitted by the least-squares method.

the kosa particles and was not uniformly distributed within them. Moreover, these slope values ( $b$ ) showed that the coarse particles collected at Yamaguchi had 8.4 times the NO<sub>3</sub><sup>-</sup> on their surface than the coarse particles collected at Beijing (Fig. 7).

The concentration of SO<sub>4</sub><sup>2-</sup> in the bulk aerosol decreased by  $\frac{1}{3}$  during long-range transport from the interior of China to Japan as mentioned in the Section 3.2. The  $r^2$  value (0.67) of  $[\text{SO}_4^{2-}] = a + b[\text{Al}_{\text{mass}}]$  for the aerosol sample collected at Beijing was larger than the  $r^2$

Table 5

Slope and  $r^2$  values for linear regressions of Al concentrations and the concentrations of crustal elements in same particle size range, and the average ratio of crustal element concentrations to the Al concentrations in the bulk aerosol (TSP) over five groups of stations in China and Japan

X	Beijing $D \geq 2.1 \mu\text{m}$		Yamaguchi $D \geq 2.1 \mu\text{m}$		Average TSP
	$r^2$	Slope	$r^2$	Slope	X/Al
Mg <sup>a</sup>	0.99	0.3	0.95	0.2	0.2
Al	1.00	1.0	1.00	1.0	1.0
K <sup>a</sup>	0.99	0.3	—	—	0.3
Ca <sup>a</sup>	0.99	0.7	0.86	0.6	0.5
Ti	0.98	0.07	0.95	0.04	0.07
V	0.99	0.001	—	—	0.002
Mn	0.96	0.01	0.99	0.01	0.01
Fe	0.99	0.5	0.95	0.4	0.5
Sr <sup>a</sup>	0.99	0.004	0.90	0.005	0.004
Ba	0.98	0.01	—	—	0.009

<sup>a</sup>Sea salt contributions were subtracted from Yamaguchi values.

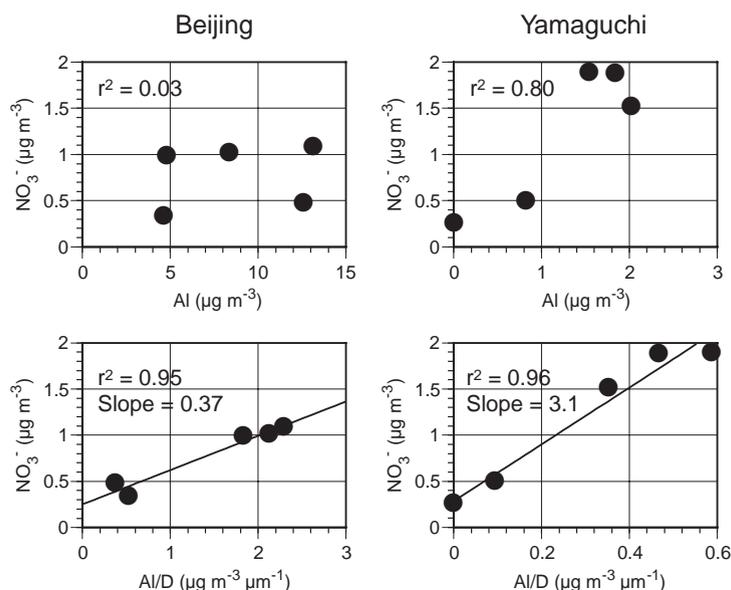


Fig. 7.  $\text{NO}_3^-$  concentrations in the size-separated aerosol larger than  $2.1 \mu\text{m}$  plotted against Al concentrations and Al/D for similar size ranges. Straight lines were fitted by the least-squares method.

value (0.35) for  $[\text{SO}_4^{2-}] = a + b[\text{Al}_{\text{mass}}/D]$  (Fig. 8). This indicated that  $\text{SO}_4^{2-}$  in the coarse aerosol over Beijing existed uniformly within the kosa particles rather than being attached to particle surfaces. For the aerosol collected at Yamaguchi, the  $r^2$  value (0.87) of  $[\text{SO}_4^{2-}] = a + b[\text{Al}_{\text{mass}}]$  was almost identical to that (0.93) for  $[\text{SO}_4^{2-}] = a + b[\text{Al}_{\text{mass}}/D]$  (Fig. 8). This indicated that  $\text{SO}_4^{2-}$  in the Yamaguchi coarse aerosol existed uniformly within all the coarse particles as for the crustal elements, and that  $\text{SO}_4^{2-}$  was also attached to the surface just as for the  $\text{NO}_3^-$ . In addition, this result is evidence that more  $\text{SO}_4^{2-}$  was attached to the aerosol particle surfaces in the Yamaguchi aerosol sample than in the Beijing

sample. The slope values ( $b$ ) showed that the coarse aerosol sampled at Yamaguchi contained a higher proportion of  $\text{SO}_4^{2-}$  than the coarse aerosol sampled at Beijing. Thirty-three per cent  $\text{SO}_4^{2-}$  in the Beijing aerosol and 67%  $\text{SO}_4^{2-}$  in the Yamaguchi aerosol were detected in particles smaller than  $2.1 \mu\text{m}$ . This suggested that  $\text{NH}_4\text{HSO}_4$  and/or  $(\text{NH}_4)_2\text{SO}_4$ , that were found in the fine particles in urban air (Tani et al., 1983), existed in the same air mass.

Fluoride was detected in particles greater than  $2.1 \mu\text{m}$  at both Beijing and Yamaguchi. The  $r^2$  value for  $[\text{F}^-] = a + b[\text{Al}_{\text{mass}}/D]$  for aerosol sample collected at Beijing was 0.95, although the  $r^2$  value for  $[\text{F}^-] = a + b[\text{Al}_{\text{mass}}]$

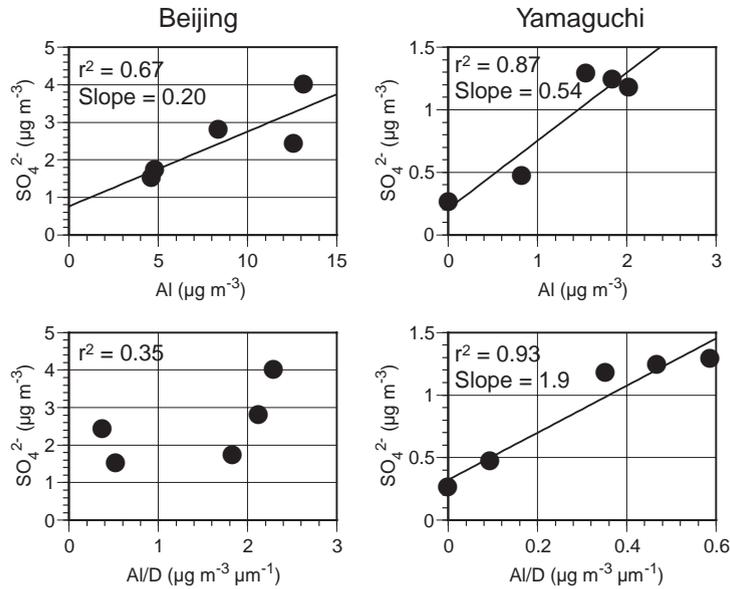


Fig. 8.  $\text{SO}_4^{2-}$  concentrations in the size-separated aerosol larger than  $2.1\ \mu\text{m}$  were plotted against Al concentrations and Al/D for similar size ranges. Straight lines were fitted by the least-squares method.

for aerosol sample collected at Beijing was 0.10. This indicated that  $\text{F}^-$  was also attached to the surface of the particles just as for  $\text{NO}_3^-$ . Since the concentrations of  $\text{F}^-$  in the Yamaguchi aerosol particles larger than  $2.1\ \mu\text{m}$  were measured in only three size ranges ( $2.1\text{--}3.3$ ,  $3.3\text{--}4.7$ , and  $4.7\text{--}7.0\ \mu\text{m}$ ), a comparison between the Al concentrations and the  $\text{Al}_{\text{mass}}/D$  values was not done. The  $r^2$  value for  $[\text{Zn}] = a + b[\text{Al}_{\text{mass}}]$  for particles greater than  $2.1\ \mu\text{m}$  collected at Beijing was less than 0.01, and the  $r^2$  value for  $[\text{Zn}] = a + b[\text{Al}_{\text{mass}}/D]$  was 0.37. This suggested that there was a tendency for Zn to be attached to the surface of coarse particles, although the  $r^2$  value was not high.

More than 50% of the  $\text{NH}_4^+$  and Pb were detected in aerosol particles smaller than  $2.1\ \mu\text{m}$  in aerodynamic diameter (Table 4). It is thus evident that an overwhelming abundance of particles containing  $\text{NH}_4^+$  and Pb were completely different from particles containing crustal elements, such as kosa aerosol particles, even though they were in the same air mass.

#### 4. Summary

The following conclusions were obtained from chemical analyses of the size-separated aerosol sampled along the route of the dust event in March 2001.

1. Nitrate ions in the aerosol collected along the transport route of the kosa event were shown to be attached to the surface of kosa particles. The  $\text{NO}_3^-$  attached to the aerosol surface was nearly 10 times

larger in Japan (after long-range transport) than in China (near the source).

2. Sulphate ions in the aerosol collected along the transport route of the kosa event existed in three categories: namely, attached to the surface of kosa particles, as a component inside coarse particles, in fine particles other than the coarse ones. Moreover, the surface accumulation of sulphate ions was greater in Japan than in China.
3. The relative concentrations of crustal elements in the aerosol samples collected in China and Japan for the same dust event did not depend on particle size or sampling location, and were almost constant.

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