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Estimation of the concentration and chemical composition of kosa aerosols at their origin

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Abstract

Kosa aerosols are formed of mineral dust lifted into the atmosphere by dust storms generated over the Asian continent. An intense dust storm which lifted kosa particles into the atmosphere was generated over the Badain Jaran Desert, China from 14 to 15 April 1998. We sampled the kosa aerosol at seven locations in China and Japan along its transport route, and investigated aerosol mass concentrations and the concentrations of chemical components in the aerosol. The aerosol mass concentrations and concentrations of crustal elements in the aerosols decreased exponentially with the distance from the kosa aerosol source. The chemical composition of the kosa aerosol at source, estimated from regressions of aerosol crustal element concentrations on distance from the kosa source, closely coincided with those of simulated Asian mineral dust certified reference material (CJ-2) prepared from the desert surface soil. The estimated concentration of Al in the kosa source aerosol was $5.3 \times 10^3\,\mu g\,m^{-3}$ and the aerosol mass concentration was $9.0 \times 10^4\,\mu g\,m^{-3}$.

Keywords: Asian dust storm; Mineral dust; Long-range transport; Simulated Asian mineral dust certified reference material (CJ-2)

1. Introduction

Many researchers have pointed out the importance of climate forcing by mineral dust in the atmosphere (Duce, 1995; Li et al., 1996). However, large uncertainties in predicting the radiative impacts of atmospheric dust still exist because of deficiencies in our understanding of the physical and chemical processes involved and a lack of adequate data (Sokolik et al., 2001). China's inland arid regions are, like the Sahara desert area of North Africa, major sources of atmospheric mineral dust, known also as kosa aerosols. There are reports that kosa aerosols have been transported thousands of kilometres from their source to South Korea (Chung and Yoon, 1996; Kim et al., 2001), Japan (Mori et al., 1999), and the United States (Uematsu et al., 1983; Tratt et al., 2001). Although kosa aerosols have been observed in various locations, there are few reports on the concentration and chemical composition of kosa aerosols at their source (Zhang et al., 1993). Aerosol monitoring at kosa sources is nearly impossible because of topographical and meteorological difficulties.

In the work reported here we have estimated the aerosol mass concentration and the chemical composition of the aerosol at its source based on chemical analysis data. Kosa aerosols were collected at multiple locations during an intense and typical kosa event that occurred over the entire East Asia region from 14 to 22 April 1998.

2. Experimental

Between 1997 and 1999 we established a kosa aerosol sampling network comprising two locations in China

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and five in Japan (Fig. 1). Sample collection at each location was performed according to information from near the kosa source in China. Dust storm information from Yinchuan, the nearest of our sampling sites to the kosa source, was used to prepare for aerosol sampling in Beijing, and information from Beijing was used to prepare for sampling at the locations in Japan. Researchers at each sampling location decided the exact sampling period with regard to prevailing meteorological conditions. Aerosol samplers were either high-volume samplers with quartz fibre filters (2500QAT-UP, Pallflex), or low-volume samplers with membrane filters (FM-80, Fuji Photo Film) (Table 1). All samplers collected total suspended particles (TSP).

Aerosol concentrations were obtained by weighing the filters before and after sampling. The filters were left for at least 24 h in a balance chamber (20°C, 50% humidity) until a constant weight was obtained; this method

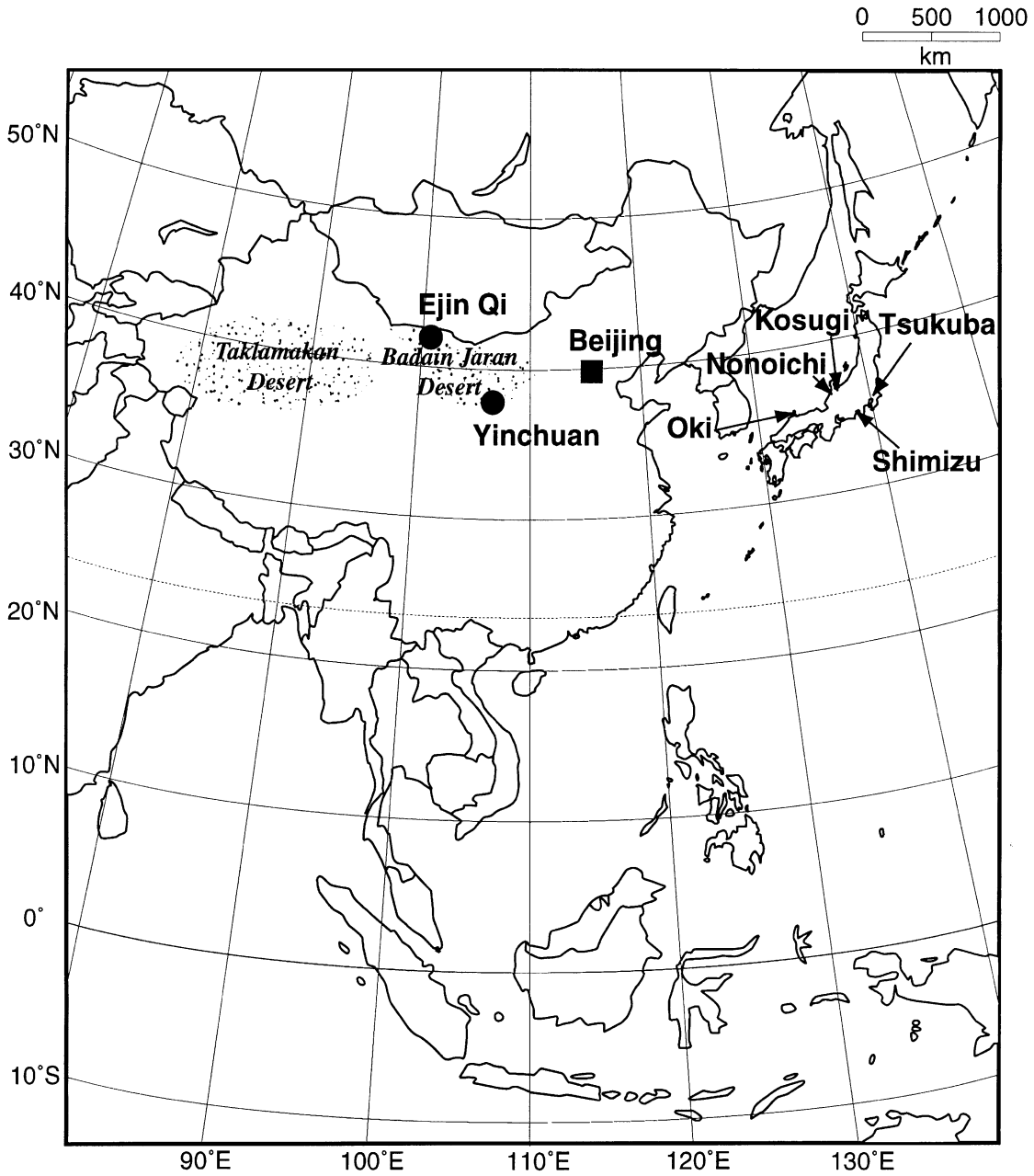

Fig. 1. Sampling locations.

Table 1
Details of sampling locations and methods

Station		Latitude	Longitude	Date	Sampler	Filter
Yinchuan	Desert, Urban	39°N	106°E	15 Apr. 1998	HV	Quartz
Beijing	Urban	40°N	116°E	17–18 Apr. 1998	HV	Quartz
Oki	Remote Island	36°N	133°E	20–22 Apr. 1998	LV	Membrane
Nonoichi	Rural	36°N	137°E	20–21 Apr. 1998	LV	Membrane
Kosugi	Rural	37°N	137°E	20–21 Apr. 1998	LV	Membrane
Shimizu	Urban	35°N	139°E	20–22 Apr. 1998	HV	Quartz
Tsukuba	Urban	36°N	140°E	20–22 Apr. 1998	LV	Membrane

conforms to the official Japanese method (Nishikawa et al., 1983). Water-soluble components (NO_3^-, SO_4^{2-}, Na^+, NH_4^+, K^+, Mg^{2+}, Ca^{2+}, Na, Mg, Al, P, K, Ca, Ti, V, Mn, Fe, Co, Ni, Cu, Zn, Sr, Ba, Pb) in the aerosol samples were extracted by 10 min ultrasonication with ultrapure water (100 × sample weight; sample weights were typically 0.1–0.2 g). The concentration of each component was then measured by ion chromatography (DX-100, Dionex) or inductively coupled atomic emission spectrometry (ICP-AES; 61E Trace and ICP-750, Thermo Jarrell-Ash). To facilitate the measurement of the insoluble components (Na, Mg, Al, P, K, Ca, Ti, V, Mn, Fe, Co, Ni, Cu, Zn, Sr, Ba, Pb), the samples were digested in mixed acid (nitric, hydrofluoric, perchloric; 100 × sample weight) and the concentration of each component was measured by ICP-AES (Nishikawa et al., 1991).

Forward trajectory analyses were performed to investigate the kosa aerosol transport route. The route was computed by the fourth-order Runge–Kutta method on an isentropic surface disregarding the effects of topography and using ECMWF-Global 2.5° grid data (Hayashida-Amano et al., 1991; Fujinuma et al., 2000).

3. Results and discussion

3.1. Transport route of the kosa aerosol

The kosa event that we observed at all our monitoring sites from 15 to 22 April 1998 was intense and extensive. It was observed in Korea (Choi et al., 2001) and Japan (Murayama et al., 2001). The dust cloud was identified on satellite images (Husar et al., 2001). From these satellite images the source could be identified near Ejin Qi city in the Badain Jaran Desert on the southern side of the Gobi Desert. Weng (1998) reported that on 14 and 15 April 1998 Ejin Qi was hit by a very severe dust storm. As at that time there were no reports of major dust storms in the Taklamakan Desert area west of the Badain Jaran, this dust storm likely had its origin in the region near Ejin Qi. Therefore, we conducted forward trajectory analyses starting at Ejin Qi (42°N, 101°E). The results indicated that between 17:00 and 21:00 on 14 April 1998 (GMT) the air masses at 1000 m above sea level over Ejin Qi passed over Yinchuan, moved over the Beijing area one day later, and between 3 and 8 days later were over Japan (Fig. 2).

3.2. Relationship between distance from dust storm origin and aerosol concentration

In general, as the distance from the source increases, the atmospheric aerosol concentration decreases because of particle deposition and three-dimensional dispersion phenomena. When there is no obvious supplementation by other aerosols during the kosa aerosol transport process, it is perhaps possible to find a correlation between aerosol transport distance and aerosol concentration and between transport distance and the concentrations of the chemical components of the aerosol.

Fig. 3a shows that TSP concentrations decreased as the distance from the source increased. The TSP concentration at Yinchuan, a city on the eastern side of the Badain Jaran Desert, was 10 times that at Beijing, and 100 times that in Japan. We fitted an exponential relationship between the distance between the kosa aerosol source and the sampling location and the TSP concentration by the least-squares method.

$$y = a\exp(bx), \tag{1}$$

where x is the distance between the kosa source and the sampling location, y is the TSP concentration, and a and b are constants. The coefficient of determination r^2 was 0.96, showing that the TSP concentration was strongly dependent on the distance from the source (Table 2). It was possible to estimate the approximate concentration in relation to the distance from the kosa aerosol source, and thereby the decrease in concentration, using Eq. (1). This paper shall use the term *distance decrement function* for the function relating the concentration of the kosa aerosol to the distance from its source.

Similarly, we plotted the concentrations of each aerosol component in relation to the distance from the kosa source (Fig. 3b–f). Aerosols sampled in Japan always had a high proportion of sea salt (Fan et al.,

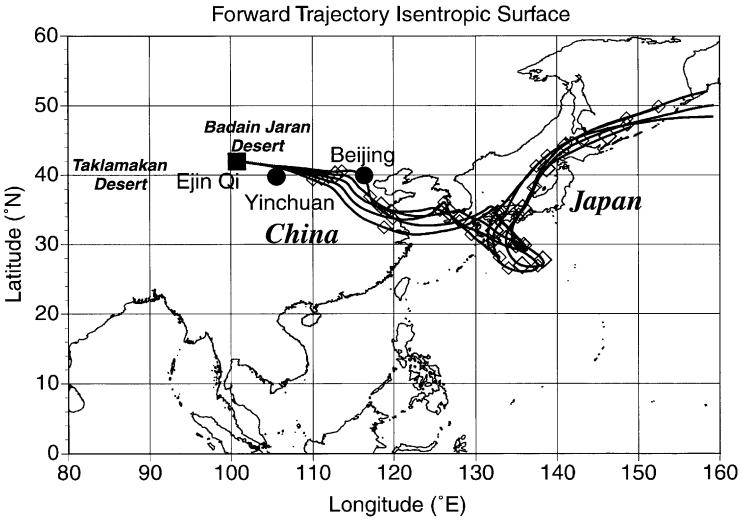

Fig. 2. Results of the forward trajectory analyses conducted every hour on the hour between 17:00 and 21:00 on 14 April 1998 (GMT) with the starting point 1000 m over Ejin Qi (l42°N, 101°E). Duration of the trajectory calculation is 5 days. The diamond symbols indicate the positions of the air masses at 12:00 (GMT) each day.

1996), which made it necessary to subtract the contribution of sea salt particles when estimating SO_4^{2-}, Ca, Mg, Na, K, and Sr. 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. (1991) to calculate the non-sea salt values of those six components.

Al, Ti, and Fe exist in large quantities in the Earth's crust, and are often considered to be indicator elements for mineral aerosols. We calculated the distance decrement functions of these crustal elements as we did TSP concentrations. The r^2 values for all the elements were at least 0.9 and there was thus a strong correlation between the concentration of each element and distance from the aerosol source. The distances at which the concentrations were reduced to one half (half-concentration distances) for these three elements coincided closely between 3.2×10^2 and 3.3×10^2 km. These values were smaller than the half-concentration distances (500–600 km) for atmospheric mineral dust calculated on the basis of the Al concentration in the aerosol by Tsunogai and co-workers (1985). This discrepancy is probably due to differences between their sampling conditions and ours; in particular, there is likely to have been a difference in particle sizes which depended on the transport distance from the source. The distance decrement functions of Na, Mg, K, Ca, Mn, Sr, and Ba were largely the same as those of Al, Ti, and Fe, with r^2 values of at least 0.9 and half-concentration distances between 2.8×10^2 and 3.4×10^2 km. Because the correlation between element concentration and distance from the kosa aerosol source was so strong, the supplementation by aerosols from other sources for these ten elements was likely to be so small as to be negligible. And because the half-concentration distances of the ten elements were all similar, it is likely that all behaved in similar manner since the generation of the aerosol at its source.

The distance decrement functions of the nitrate and ammonium in the aerosol had small r^2 values, and their half-concentration distances were long compared with the other components (Table 2). These observations suggested that significant amounts of these two components came from outside the system during transport from the source to Japan.

The distance decrement functions of eight components: SO_4^{2-}, P, V, Co, Ni, Cu, Zn, and Pb, had r^2 values of 0.81–0.89, and their half-concentration distances were between 3.4×10^2 and 4.9×10^2 km. Other sources of these components during transport appear to be significant. That the TSP half-concentration distance was 4.5×10^2 km is probably a reflection of the balance of all the aerosol components.

3.3. Chemical composition and concentrations of the aerosol at the kosa origin

Substituting $x = 0$ in the distance decrement function makes it possible to estimate the chemical component concentrations of the kosa aerosol at its source. We estimated values for only those components with half-concentration distances of 2.8×10^2 to 3.4×10^2 km and $r^2 \geqq 0.9$ (Table 3), the conditions under which it appeared possible to ignore the above-mentioned supplementation

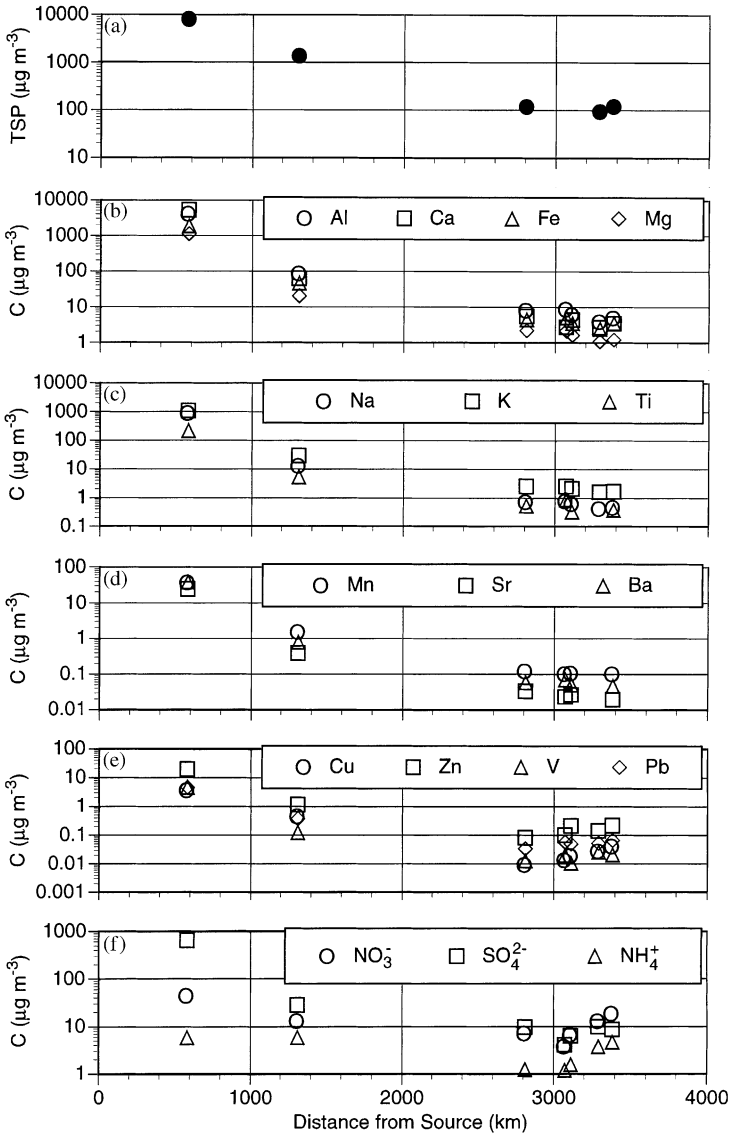

Fig. 3. Concentrations of TSP and aerosol chemical components in relation to distance from the kosa source. Values plotted for the Ca, Mg, Na, K, Sr, and SO_4^{2-} in the aerosol sampled in Japan are non-sea salt values from which the sea salt contributions have been subtracted.

during transport. The elements that met these conditions were crustal elements.

To test the validity of our chemical composition estimates, we calculated the correspondence factor (CF) of the kosa aerosol at its source in relation to the composition of CJ-2, the certified reference material for simulated Asian mineral dust prepared from desert surface soil (Nishikawa et al., 2000) (Eq. (2)):

$$(CF)_X = (X/Al)_{estimated}/(X/Al)_{CJ\text{-}2}, \quad (2)$$

where $(CF)_X$ is the CF of element X, $(X/Al)_{estimated}$ is the estimated ratio by weight of the concentration of element X to the concentration of Al in the aerosol at the source, $(X/Al)_{CJ-2}$ is the ratio of the concentration of element X to the concentration of Al in CJ-2. The CF values ranged between 0.7 and 1.5 (Table 3), and the estimated composition of the kosa aerosol at source was very similar to that of CJ-2.

The estimated concentration of Al in the aerosol at the dust storm source was $5.3 \times 10^3\,\mu g\,m^{-3}$, which was 830 times the average Al concentration ($6.4\,\mu g\,m^{-3}$) that we measured in the aerosol in Japan during the kosa event of April 1998. Because, the content of Al in the kosa aerosol solids was $5.88 \pm 0.16\%$ (Nishikawa et al.,

Table 2
The values of the constants a and b, the coefficient of determination (r^2), and the distance at which concentrations diminish by half (the half-concentration distance) when $y = a\exp(bx)$ is used to approximate the relationship between the TSP concentration or the concentration of chemical components in the aerosol and the distance from the kosa source

	a	b	r^2	Half-concentration distance (km)
TSP	1.4×10^4	−0.0015	0.96	4.5×10^2
NO_3^-	36	−0.0005	0.40	1.5×10^3
SO_4^{2-}	5.7×10^2	−0.0014	0.81	4.9×10^2
NH_4^+	7.2	−0.0004	0.32	1.8×10^3
Na	1.2×10^3	−0.0025	0.93	2.8×10^2
Mg	1.4×10^3	−0.0022	0.92	3.2×10^2
Al	5.3×10^3	−0.0022	0.92	3.2×10^2
P	74	−0.0021	0.89	3.4×10^2
K	1.5×10^3	−0.0021	0.93	3.3×10^2
Ca	6.5×10^3	−0.0024	0.91	2.9×10^2
Ti	2.7×10^2	−0.0021	0.90	3.3×10^2
V	4.3	−0.0018	0.83	3.9×10^2
Mn	56	−0.0020	0.93	3.4×10^2
Fe	2.4×10^3	−0.0021	0.92	3.3×10^2
Co	0.88	−0.0019	0.89	3.7×10^2
Ni	2.5	−0.0019	0.86	3.7×10^2
Cu	6.2	−0.0018	0.85	3.8×10^2
Zn	21	−0.0016	0.84	4.3×10^2
Sr	33	−0.0023	0.92	3.0×10^2
Ba	51	−0.0022	0.91	3.1×10^2
Pb	5.3	−0.0015	0.88	4.7×10^2

Table 3
Estimated values for the concentrations of chemical components in the aerosol at the kosa source, and their correspondence factors (CFs) in relation to the simulated Asian mineral dust certified reference material (CJ-2)

Chemical components	Concentration ($\mu g\,m^{-3}$)	CF
Na	1.2×10^3	1.0
Mg	1.4×10^3	1.0
Al	5.3×10^3	1.0
K	1.5×10^3	0.9
Ca	6.5×10^3	1.4
Ti	2.7×10^2	0.7
Mn	56	1.0
Fe	2.4×10^3	0.9
Sr	33	1.5
Ba	51	1.2

$CF = (X/Al)_{estimated}/(X/Al)_{CJ\text{-}2}$.

2000), it was possible to calculate a value of $9.0\times10^4\,\mu g\,m^{-3}$ with an error of up to $\pm30\%$ for the aerosol concentration at the kosa source. This estimated concentration is solely that of aerosols that are subject to long-range transport, and does not refer to the concentration of particles that creep or saltate around the source during a dust storm. This value corresponds to 820 times the average TSP concentration ($1.1\times10^2\,\mu g\,m^{-3}$) that we observed in Japan during the kosa event, 100 times the model-estimated aerosol concentration (exceeding $900\,\mu g\,m^{-3}$) at the surface in this kosa event (Uno et al., 2001), and 90 times the mineral aerosol concentration ($\sim1\,mg\,m^{-3}$) calculated on the basis of the Al concentration in material collected during a dust storm in Shapuotou, China (Zhang et al., 1993).

4. Summary

We collected and analysed samples of a typical kosa aerosol that originated in China in April 1998 at seven locations in succession along its transport route from China to Japan. Our results led to the following conclusions:

(1) Ten elements in the kosa aerosol, Al, Ti, Fe, Na, Mg, K, Ca, Mn, Sr, and Ba, had estimated half-concentration distances between 2.8×10^2 and 3.4×10^2 km.
(2) The chemical composition of the kosa aerosol at the dust storm source was estimated as follows: Al: $5.3\times10^3\,\mu g\,m^{-3}$; Fe: $2.4\times10^3\,\mu g\,m^{-3}$; Ti: $2.7\times10^2\,\mu g\,m^{-3}$; Na: $1.2\times10^3\,\mu g\,m^{-3}$; Mg: $1.4\times10^3\,\mu g\,m^{-3}$; K: $1.5\times10^3\,\mu g\,m^{-3}$; Ca: $6.5\times10^3\,\mu g\,m^{-3}$; Mn: $56\,\mu g\,m^{-3}$; Sr: $33\,\mu g\,m^{-3}$; and Ba: $51\,\mu g\,m^{-3}$.
(3) The estimated concentration of the kosa aerosol was calculated to be $9.0\times10^4\,\mu g\,m^{-3}$ on the basis of the Al concentration in the aerosol at the dust storm source.

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