PERGAMON

Atmospheric Environment 36 (2002) 5159–5175

ATMOSPHERIC ENVIRONMENT

www.elsevier.com/locate/atmosenv

The chemical and strontium isotope composition of atmospheric aerosols over Japan: the contribution of long-range-transported Asian dust (Kosa)

Shinji Kanayama[a,b,*], Sadayo Yabuki[b], Fumitaka Yanagisawa[a], Remi Motoyama[a]

[a] *Interactive Symbiosphere Science, Graduate School of Science and Engineering, Yamagata University, 1-4-12,Kojirakawa, Yamagata, 990-8560, Japan*

[b] *The Institute of Physical and Chemical Research (RIKEN), 2-1, Hirosawa, Wako, Saitama, 351-0198, Japan*

Received 22 November 2001; accepted 16 July 2002

Abstract

Atmospheric aerosols have been collected at four sites around Japan during 2000. From systematically monitoring the major (Na, Mg, Al, K, Ca, and Fe) and trace (Rb and Sr) elements, along with the Sr isotope composition, we have tried to estimate the contribution of long-range-transported Asian dust (“Kosa”) to the atmospheric aerosols.

The results are summarized as follows:

(1) The concentration of each element in the aerosols increased during the “Kosa” period. The increase was particularly obvious in samples collected on 8 April 2000, when the “Kosa Phenomenon” was observed at all the sampling sites in Japan, 2 days after a very heavy dust storm had occurred in China.

(2) The Rb–Sr isotopic diagram shows a two-component mixing relationship: one with a high ^{87}Sr/^{86}Sr ratio and a high ^{87}Rb/^{86}Sr ratio, and the other with a low ^{87}Sr/^{86}Sr ratio and a low ^{87}Rb/^{86}Sr ratio. There is a significant difference between that of the expected end member of the Asian dust and that of the reported Asian loess, which is thought to be the possible source of the components of the “Kosa”, although the lower component is consistent with the local component at Wako.

(3) Plots of the ^{87}Sr/^{86}Sr ratio vs the Ca/Al and Sr/Al ratios support a two-component mixing suggested by the Rb–Sr systematics, and they indicate that the contributing continental soil components to the “Kosa” aerosols should be composed of the silicate fraction of Asian loess.

(4) The discrepancy in the Rb–Sr systematics between the expected end member and the possible sources may be caused by the dissolution of the Ca-bearing minerals via long-range dust transport, or by a combination of source characteristics and grain size separation.

Keywords: Sr isotope; Asian dust; Loess; Anthropogenic components; Two-component mixing

1. Introduction

An Asian dust event is a common phenomenon that frequently occurs during spring in the arid and semi-arid regions of inland Asia. Surface soil particles are blown into the upper atmosphere, and transported over long distances to East Asia (Liu et al., 1981), Japan (Ishizaka and Ono, 1982), the Pacific regions (Uematsu et al., 1983), and even to North America (Husar et al., 2001) by strong westerly winds. In Japan, Asian dust is commonly known as “Kosa” (yellow sand) (KOSA, 1991). Recently, Asian dust has been attracting increased attention because it has a significant influence on the global climate (Tegen and Fung, 1995), and on

---

*Corresponding author. Fax: +81-48-462-4654.

E-mail address: kanayama@postman.riken.go.jp (S. Kanayama).

1352-2310/02/$ - see front matter © 2002 Elsevier Science Ltd. All rights reserved.
PII: S1352-2310(02)00587-3

geochemical mass cycles (Iwasaka et al., 1988). Therefore, it is important to identify in a systematic way the origin, transportation process, and flux of Asian dust.

There are many pioneering and successful studies using Sr isotopes as a tracer of long-range-transported materials, such as loess and desert sands (Liu et al., 1994; Gallet et al., 1996; Honda, 1999), deep-sea sediments (Biscaye and Dasch, 1971; Asahara et al., 1995), and ice-trapped materials (Biscaye et al., 1997; Svensson et al., 2000). This is because Sr isotopes are relatively unaltered by the processes of weathering, transportation, and deposition. As mentioned above, these studies were concerned with aeolian components in sedimentary material, but were not concerned with aeolian dust as one of the components of atmospheric aerosols.

According to an IPCC report (1996), an atmospheric aerosol is a mixture of various source materials—soils, sea salt, elemental carbon or organic material—and secondary particles such as sulphate or nitrate. Furthermore, each component is also a mixture of local and long-range-transported materials, or natural and anthropogenic materials. In the case of such a complicated mixture, it is difficult to determine the source materials by major and trace element chemistry alone. For example, Ca has been assumed to be one of the most important indicators of Asian dust, but construction materials (Zhang and Iwasaka, 1999) and road dust, which is caused by friction between wheels and road surfaces, (KOSA, 1991) are also important sources of Ca in atmospheric aerosols. By using an isotope composition, such as Sr, S, Pb, along with chemical composition, we can obtain further information regarding the source materials of an aerosol and the long-range-transported materials of Asian dust. Sr isotope studies of atmospheric aerosols have been carried out mainly for the soluble components of aerosols in rainwater (Herut et al., 1993; Nakano and Tanaka, 1997). Nakano and Tanaka (1997) described a mixing model of three components: Asian dust, sea salt, and local soil, for Sr isotopes in wet precipitation in Japan. However, Sr is a soil constituent element, and water-soluble Sr is only a part of the total Sr content of an aerosol. In particular, to discuss the contribution of Asian dust, mainly consisting of soil particles, to the aerosols over Japan, we need to perform bulk aerosol analysis, as well as analysis of the water-soluble components.

In this study, we present the Sr isotope composition of the bulk aerosols, as well as the major and trace element composition, and try to estimate the contribution of Asian dust to the atmospheric aerosol over Japan, particularly focusing on the "Kosa" event of 8 April 2000. For this purpose we introduce Ca/Al and Sr/Al ratios as an index of chemical alteration during a long-range dust transport.

2. Samples and experimental methods

2.1. Samples

Aerosol samples were collected at four sites around Japan during 2000: Shimonoseki, a seaside city in western Japan; Wako, an inland city in the Tokyo metropolitan area; and Tsuruoka and Yamagata, a seaside city and an inland city in northeastern Japan, respectively (see Fig. 1). All of the samples were collected on the roofs of buildings (about 20 m high) using high-volume air samplers (SIBATA; HV-1000F) with a $20 \times 25\,\mathrm{cm}^2$ SiO_2 or PTFE filter and a constant flow rate of 700 or $1000\,\mathrm{l\,min}^{-1}$. The aerosol sample descriptions are summarized in Table 1. One Wako aerosol sample, W000918, was collected near ground level besides a road subject to heavy traffic (Route 254), in order to evaluate the local aerosol components in an urban area. The collection period was during September, in a season when it is thought that the Asian dust does not affect the aerosols in Japan.

To discuss the local source components, and for comparison with the local aerosol components, four local soils were collected around the Wako sampling site: two natural soils near the W000918 sample point, and two road deposits collected beside Route 254. The road deposit is thought to be of mixed components, consisting of remote soils sticking to a tire, fragments of asphalt, tire chips, road dust, and so on. As another possible source component in the studied period, we collected and analysed a volcanic ash, ejected from a series of volcanic activity at Miyakejima Island in the summer of 2000. To discuss the Asian dust source and transportation process, a leaching experiment was carried out for the four loess samples collected at the Yili Basin located in northwest Xinjiang, China. All the soil sample descriptions are summarized in Table 2, and the locality of the Yili Basin is shown in Fig. 1.

2.2. Experimental

Appropriate fractions of the collected aerosol filters (1/4 or 1/8) were used for chemical and isotopic analyses by decomposition and by water extraction. For decomposition, the aerosol samples were decomposed in a screw cap Teflon container containing hydrofluoric, perchloric, and nitric acids at 220°C. To reduce the risk of contamination, all the acid used in the decomposition and in the preparation of the solutions used ultra-pure reagents from the Kanto Chemical Co. Inc., and Milli-Q water was used for preparation and dilution. For water extraction, duplicate aerosol samples were extracted using distilled water and ultrasonic cleaning to analyse the water-soluble elements (Tanaka et al., 1998).

Soil samples collected around the Wako site were sieved, and only the fraction $<0.5\,\mathrm{mm}$ was decomposed and

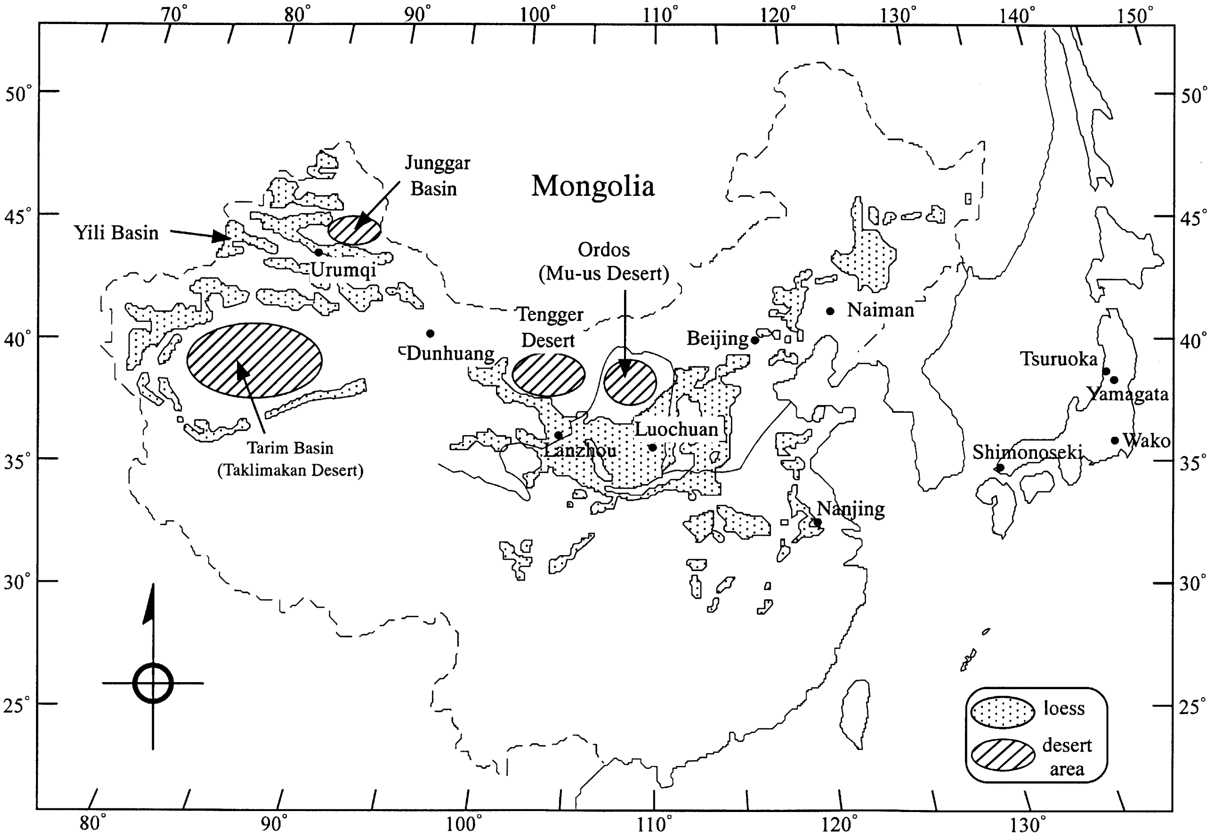


Fig. 1. The location of the sampling sites. This map is modified after Pye (1987).

analysed in the same manner as the aerosol decomposition. In order to estimate the insoluble (mainly silicate) fraction separately, four loess samples were leached using 25% acetic acid for 24 h. Both the bulk and the insoluble silicate fraction were decomposed and analysed.

For two aliquots of the prepared samples, the concentrations of the major and trace elements were determined using an inductively coupled plasma atomic emission spectrometer (Nippon Jarrell Ash TCAP-575II) and an inductively coupled plasma mass spectrometer (VG Elemental PQ-Ω) located at RIKEN. The uncertainties in the major and trace element analyses were estimated to be less than ±5%. Major element compositions of the bulk and silicate fractions of the loess samples were determined using an X-ray fluorescence spectrometer (Shimadzu SXF-1100). Fusion glass made from a mixture of the sample and $Li_2B_4O_7$ flux in a proportion of 1:10 was used for the measurement.

2.3. Isotope measurement

The sample fraction $>1\,\mu g$ for Sr was split and chemically purified using column separation with a cation exchange resin (DOWEX 50W-8X). The Sr was eluted with 2 N HCl twice on the column (∅ 4 mm and 7 cm height resin, about 0.9 ml in volume) to reduce any Rb interference in the isotopic measurement. The Sr recovery rate was estimated at about 90% in a single separation. The Sr was loaded as the nitrate on a Ta-single filament. Isotopic analysis was carried out using a thermal ionization mass spectrometer (TIMS: VG Elemental SECTOR 54) with multiple collectors. The samples were run in multi-dynamic mode for >100 ratios with a filament current 3.0–3.7 A, and ^{88}Sr beam of $1\text{–}2\times10^{-11}$ A. The Sr isotope ratios ($^{87}Sr/^{86}Sr$) were normalized to $^{86}Sr/^{88}Sr=0.1194$. The long-term mean and uncertainties (2σ) of the Sr standard (NBS987) were 0.71025 ± 0.00001 ($n=12$). The procedure blank for the chemical separation was estimated at 1–2 ng for Sr, which is $<0.2\%$ of the total Sr, and is negligible.

3. Results and discussion

3.1. Filter blank and sea-salt correction

Among the aerosol samples, those from Shimonoseki and Tsuruoka were collected on a quartz filter. In the

Table 1
Aerosol sample descriptions collected around Japan, in 2000

Sampling site	Sample	Period	Filter	Flow rate ($\mathrm{l\,min^{-1}}$)	Sampled air volume ($\mathrm{m^3}$)	Aerosol weight (g)	Filter fraction		2000 Kosa Phenomenon
							Water	Decompose	
Shimonoseki	S000327	27/3–31/3	SiO_2	1000	5810	n.m.	1/4	1/8	3, 2, 3, 24, 26/3
(Yamaguchi Pref.)	S000406	6/4–10/4	SiO_2	1000	5765	n.m.	1/4	1/8	8, 11, 14/4
	S000417	17/4–21/4	SiO_2	1000	6076	n.m.	1/4	1/8	
Wako (RIKEN)	W000324	24/3–31/3	PTFE[a](050)	1000	6841	0.3994	1/8	1/8	8/4, 14/4
(Saitama Pref.)	W000331	31/3–13/4	PTFE(050)	1000	17901	2.1217	1/8	1/8	
	W000413	13/4–21/4	PTFE(050)	1000	10423	0.7039	1/8	1/4	
	W000424	24/4–2/5	PTFE(050)	1000	11506	0.6777	1/8	1/4	
	W000512	12/5–29/5	PTFE(050)	1000	24351	1.1838	1/8	1/4	
	W000529	29/5–15/6	PTFE(050)	1000	14689	1.3306	1/4	1/4	
	W000615	15/6–29/6	PTFE(050)	1000	19888	1.1316	1/8	1/8	
	W000721	21/7–11/8	PTFE(050)	1000	29600	1.4299	1/8	1/4	
	W000814	14/8–29/8	PTFE(050)	1000	21407	0.9972	1/8	1/4	
Surface (Wako)	W000918	18/9–22/9	PTFE(050)	1000	5027	0.4962	1/8	1/4	
Yamagata	Y000323	23/3–28/3	PTFE(040)	700	5040	n.m.	1/4	1/8	24/3, 8/4
(Yamagata Pref.)	Y000330	30/3–4/4	PTFE(040)	700	5040	n.m.	1/4	1/8	
	Y000406	6/4–11/4	PTFE(040)	700	4620	n.m.	1/4	1/8	
	Y000413	13/4–18/4	PTFE(040)	700	5040	n.m.	1/4	1/8	
Tsuruoka	T000327	27/3–1/4	SiO_2	700	5040	n.m.	1/8	1/8	8/4[b]
(Yamagata Pref.)	T000410	10/4–15/4	SiO_2	700	5040	n.m.	1/8	1/8	
	T000424	24/4–29/4	SiO_2	700	5040	n.m.	1/8	1/8	

A numbering of samples mean a sampling site and a started date of the sampling (m/d/y).
For example, S000327 has collected at Shimonoseki and started the sampling on 27 March, 2000.

[a] PTFE filter (050):PF050 (ADVANTEC), collection efficiency (0.3 μmDOP%) = 85.0 and PF040 (ADVANTEC), collection efficiency (0.3 μmDOP%) = 95.0.

[b] "Kosa Phenomenon" observed in Sakata, located on 25 km north from Tsuruoka.

Table 2
Soil samples descriptions

Sample	Type	Place	Descriptions	Grain size (mm)	Note
WRD-1	Road deposit-1	Wako, Japan	Beside the heavy traffic road (R254)	$<\phi$ 0.5	
WRD-2	Road deposit-2	Wako, Japan	Another point of WRD-1	$<\phi$ 0.5	
WS-1	Surface soil-1	Wako, Japan	The flower bed soil beside WRD-2	$<\phi$ 0.5	
WS-2	Surface soil-2	Wako, Japan	Sampled at the same point of W000918	$<\phi$ 0.5	
MYK-Ash	Volcanic ash of Miyake-jima	Miyakejima, Japan	Sampled at August 2000 after the heavier explosion	No segregation	The eruption started from 8 July and continued intermittently
Z-3	Loess (typical)	Yili basin, China	Zeketai sequence	No segregation	Extracted by 25% acetic acid for 24 h. The bulk and silicate components had been analysed
Z-191	Loess (typical)	Yili basin, China	Zeketai sequence		
L-4	Loess (Ca rich)	Yili basin, China	Kuerdenengbulake sequence		
L-7	Loess (Ca poor)	Yili basin, China	Kuerdenengbulake sequence		

case of these samples, not only the sample materials but also the filter matrix were completely decomposed by the $HF–HClO_4–HNO_3$ mixed acid. To evaluate the quartz filter blank, we carried out four sets of blank filter decomposition and measurements. As shown in Table 3, a considerable amount of chemical constituents were detected as the quartz filter blank. Since each filter had a relatively uniform chemical and Sr isotope composition, the values of the samples collected on the quartz filter were corrected by the average value of the blanks. On the other hand, the contribution of the PTFE filter blank was small, because tetrafluoroethylene is stable against the $HF-HClO_4-HNO_3$ mixed acid. Except for K, the blank fraction of the total contents in all samples was less than 5%, which is within the analytical error for the ICP-AES and ICP-MS. Hence, we consider that it is also negligible for the correction of the PTFE filter blank to the sample constituents used in this study.

As the Japanese islands are surrounded by sea, the sea-salt contribution to the aerosols cannot be avoided, even in the inland area. Assuming that the whole of the water-soluble Na in the collected aerosols originates from sea salt, $(X)_{\mathrm{nss}}$, the concentration of non-sea-salt Mg, Ca, K, Rb and Sr was calculated from the following equations:

$$(X)_{\mathrm{nss}} = (X)_{\mathrm{mes}} - (X)_{\mathrm{sea}} - (X)_{\mathrm{filter}}, \tag{1}$$

$$(X)_{\mathrm{sea}} = (X/\mathrm{Na})_{\mathrm{sea}} \times (\mathrm{Na})_{\mathrm{mes}}, \tag{2}$$

where $(X)_{\mathrm{mes}}$ is the measured quantity of Mg, Ca, K, Rb and Sr. The sea-salt fraction of each element is denoted by $(X)_{\mathrm{sea}}$, and is calculated by using Eq. (2). The filter blank fraction of the elements, $(X)_{\mathrm{filter}}$, is cited in Table 4, and is only for the samples collected on the SiO_2 filter (at Shimonoseki and Tsuruoka). The quantity $(\mathrm{Na})_{\mathrm{mes}}$ is the measured Na for the water-extracted components. The concentration ratio of each elements in seawater, $(X/\mathrm{Na})_{\mathrm{sea}}$, had the values $(\mathrm{Mg/Na}) = 0.120$, $(\mathrm{Ca/Na}) = 0.038$, $(\mathrm{K/Na}) = 0.037$, $(\mathrm{Rb/Na}) = 0.011 \times 10^{-3}$, and $(\mathrm{Sr/Na}) = 0.725 \times 10^{-3}$, respectively, were calculated from Matsuo (1989) and were used in Eq. (2). The Sr isotope ratio of non-sea-salt strontium, $(^{87}\mathrm{Sr}/^{86}\mathrm{Sr})_{\mathrm{nss}}$, was calculated using the following equation:

$$\begin{aligned}(^{87}\mathrm{Sr}/^{86}\mathrm{Sr})_{\mathrm{nss}} = [&\{(^{87}\mathrm{Sr}/^{86}\mathrm{Sr})_{\mathrm{mes}} \times (\mathrm{Sr})_{\mathrm{mes}}\} \\ &- \{(^{87}\mathrm{Sr}/^{86}\mathrm{Sr})_{\mathrm{sea}} \times (\mathrm{Sr})_{\mathrm{sea}}\} \\ &- \{(^{87}\mathrm{Sr}/^{86}\mathrm{Sr})_{\mathrm{filter}} \times (\mathrm{Sr})_{\mathrm{filter}}\}]/(\mathrm{Sr})_{\mathrm{nss}},\end{aligned} \tag{3}$$

where $(^{87}\mathrm{Sr}/^{86}\mathrm{Sr})_{\mathrm{mes}}$ and $(\mathrm{Sr})_{\mathrm{mes}}$ are the measured values of the Sr isotope ratios and the measured quantity of Sr. The Sr isotope ratio of seawater, $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$, was taken as $^{87}\mathrm{Sr}/^{86}\mathrm{Sr} = 0.70906$ (Faure, 1986). The values of $(\mathrm{Sr})_{\mathrm{sea}}$ and $(\mathrm{Sr})_{\mathrm{nss}}$, the concentration of sea-salt and non-sea-salt strontium respectively, were calculated from Eqs. (1) and (2). The filter blank term is defined as

Table 3
Results of filter blank measurement. The value of major and trace elements show the quantity (mg or μg) by a piece of filter (20 × 25 cm)

Sample	No	(mg)						(μg)		$^{87}Sr/^{86}Sr$
		Na	Mg	Al	K	Ca	Fe	Rb	Sr	
SiO_2 filter	1	1.30	0.19	1.50	0.37	0.77	0.15	0.71	4.59	n.m.
	2	n.m.	n.m.	n.m.	n.m.	n.m.	n.m.	0.71	4.71	n.m.
	3	1.09	0.19	1.16	0.42	0.88	0.14	0.81	4.98	0.710324 ± 15
	4	1.22	0.19	1.13	0.38	0.82	0.14	0.87	5.01	0.710351 ± 17
Average		1.20	0.19	1.26	0.39	0.82	0.14	0.78	4.82	0.710338 ± 16
T000327[a]		7.06	1.72	3.44	2.60	3.83	2.38	10.39	32.09	
Blank (%)		17.1	11.2	36.7	15.0	21.5	5.9	7.5	15.0	
PTFE filter	1	0.08	<0.001	0.04	0.10	0.10	0.03	0.04	0.52	n.m.
	2	0.09	<0.001	0.05	0.10	0.11	0.04	0.05	0.61	n.m.
Average		0.08	0.00	0.05	0.10	0.10	0.03	0.05	0.57	—
Y000323[b]		4.37	1.60	4.99	1.77	2.92	2.96	7.06	19.05	
Blank (%)		1.9	0.0	0.9	5.8	3.5	1.1	0.6	3.0	

[a] T000327 has the minimum element concentration of all the samples collected on a SiO_2 filter in this study.

[b] Y000323 has the minimum element concentration of all the samples collected on a PTFE filter in this study.

$(Sr)_{filter} = 4.82\,\mu g$, and $(^{87}Sr/^{86}Sr) = 0.710336 \pm 16$ (2σ) (See Table 3). The sea-salt contribution to the Sr (%) concentration was estimated, and is shown in Table 4. The values of the seaside cities are higher than those of the inland cities.

3.2. The "Kosa Phenomenon" over spring 2000

"Kosa Phenomena" in Japan are closely related to the dust storm events of inland Asia. The potential source areas were analysed using SYNOP data compiled on geostationary meteorological satellite (GMS) images (Kurosaki, 2001, pers. commun.), and are summarized in Fig. 2. The major "Kosa Phenomena" in Japan during spring 2000, which had been observed at more than two of the sampling sites in Table 1, are documented below.

(1) "Kosa Phenomenon" on 24 March at Shimonoseki and Yamagata (Fig. 2a). Preceding this event, a dust storm had broken out in the southern parts of Mongolia and the western part of Inner Mongolia, China (40–45°N, 105°E–115°E) on 21 and 22 March, and a few dust storms and a "Kosa Phenomena" had been observed around the Korean Peninsula on 23 and 24 March.

(2) "Kosa Phenomenon" on 8 April all over Japan (Fig. 2b). A very strong dust storm resulting in the most intensive "Kosa Phenomenon" in 2000 had broken out in the southern parts of Mongolia (45–50°N, 100–115°E) on 5 April, and the air mass that caused this dust storm moved eastward to the western part of Inner Mongolia, China (40–50°N, 110–120°E) on 6 April. At the same period, the dust covered Beijing and the southern area on 6 and 7 April. After the "Kosa Phenomenon" had caused serious damage over the Korean Peninsula on 7 April, it arrived in Japan on 8 April.

(3) "Kosa Phenomenon" on 14 April at Shimonoseki and Wako (Fig. 2c). Although the dust storm related to this "Kosa Phenomenon" was sparser than the former two events, they had been observed around Gansu Province (35–40°N, 90°–105°E) on 12 April, and on 13 April, the dust covered the southern part of Gansu Province and the Ningxia Hui Autonomous Region, just located on the Central Loess Plateau. Although a "Kosa Phenomenon" had been observed in the southern parts of the Korean Peninsula, this event was obviously different to that in Japan from 14 to 15 April.

3.3. Characteristics of major and trace element composition

The results of the chemical and isotope analyses are summarized in Table 4. All the data are corrected for sea

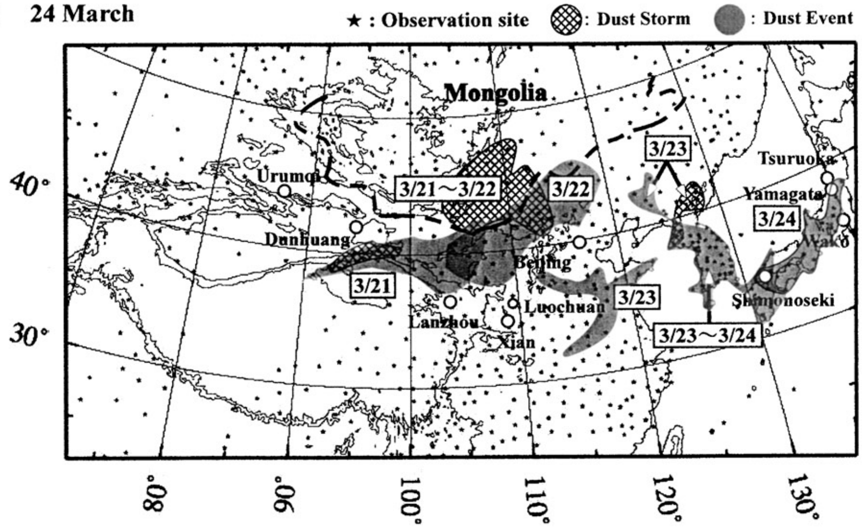
Table 4
Analytical results of aerosols

Site	No.	(μg m^{-3})								(ng m^{-3})		^{87}Sr/^{86}Sr$\pm 2\sigma$	^{87}Rb/^{86}Sr	Ca/Al	Sea Salt (%) for Sr
		Aerosol conc.	Na$-W^a$	Ca$_{nss}-W^a$	Mg$_{nss}$	Al	K$_{nss}$	Ca$_{nss}$	Fe	Rb$_{nss}$	Sr$_{nss}$				
Shimonoseki	S000327	n.m.	10.10	1.99	0.82	4.85	1.79	4.04	3.22	11.01	23.45	0.712627±24	1.36	0.83	23.80
	S000406	n.m.	3.66	2.21	1.19	6.66	1.86	2.86	4.27	11.52	20.82	0.712651±24	1.60	0.43	11.31
	S000417	n.m.	2.67	0.74	0.18	0.91	0.59	0.80	0.80	3.49	4.88	0.711123±21	2.07	0.87	28.36
Wako	W000324	58.4	0.66	1.59	0.61	2.03	0.60	1.71	1.45	2.32	7.87	0.710862±17	0.85	0.84	5.75
	W000331	118.5	1.88	1.37	1.36	5.72	1.59	3.62	3.35	8.69	18.85	0.711391±15	1.33	0.63	6.75
	W000413	67.5	0.61	1.83	0.38	1.63	0.81	1.61	1.36	3.86	8.00	0.711302±24	1.39	0.99	5.25
	W000424	58.9	0.42	2.13	0.31	1.39	0.53	3.05	1.28	2.32	8.85	0.709804±18	0.76	2.20	3.33
	W000512	48.6	0.88	1.31	0.17	1.04	0.36	1.34	1.15	1.83	5.21	0.709785±26	1.01	1.29	10.86
	W000529	90.6	1.19	4.47	1.74	2.03	0.59	3.60	1.88	2.18	9.10	0.708719±23	0.69	1.77	8.66
	W000615	56.9	0.61	1.08	0.14	0.72	0.43	1.04	0.97	1.50	4.95	0.709226±23	0.88	1.44	8.24
	W000721	48.3	0.96	1.34	0.25	1.49	0.43	1.71	1.41	1.30	8.25	0.708480±21	0.46	1.15	7.74
	W000814	46.6	1.06	1.03	0.09	0.65	0.31	1.04	1.13	1.00	6.85	0.708561±20	0.42	1.58	10.11
Surface (Wako)	W000918	98.7	1.22	1.61	0.35	1.49	0.57	1.72	2.01	2.17	8.80	0.709221±23	0.71	1.16	9.13
Yamagata	Y000323	n.m.	0.48	0.42	0.26	0.99	0.33	0.56	0.59	1.40	3.43	0.710662±111	1.18	0.57	9.27
	Y000330	n.m.	0.67	0.48	0.25	0.93	0.33	0.73	0.54	1.41	4.07	0.710085±27	1.00	0.78	10.63
	Y000406	n.m.	0.52	1.03	0.54	2.50	0.93	1.64	1.50	4.35	8.49	0.711366±27	1.48	0.65	4.27
	Y000413	n.m.	0.28	0.65	0.31	1.13	0.53	0.94	0.81	2.78	5.06	0.711661±73	1.59	0.83	3.86
Tsuruoka	T000327	n.m.	1.50	0.54	0.11	0.54	0.38	0.54	0.45	1.89	4.32	0.711253±23	1.27	1.01	20.08
	T000410	n.m.	2.16	1.11	0.56	2.48	1.13	1.18	1.73	6.27	10.63	0.712549±20	1.71	0.47	12.83
	T000413	n.m.	0.94	0.54	0.23	1.05	0.47	0.61	0.75	2.37	4.34	0.712320±21	1.58	0.58	13.61

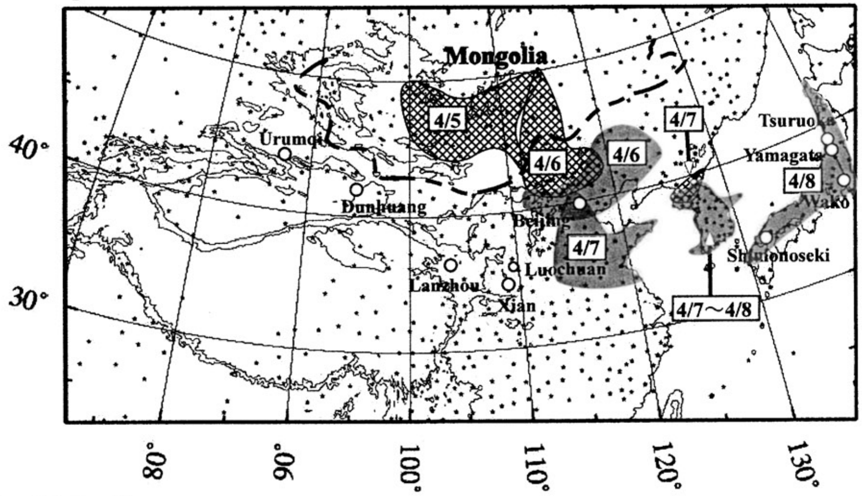
[a] The measured value of water-soluble component. The method of water extraction was referred by Tanaka et al. (1998).

Nss in footnote means non-sea-salt component, see text.

(a) **24 March**



(b) **8 April**



(c) **13 April**

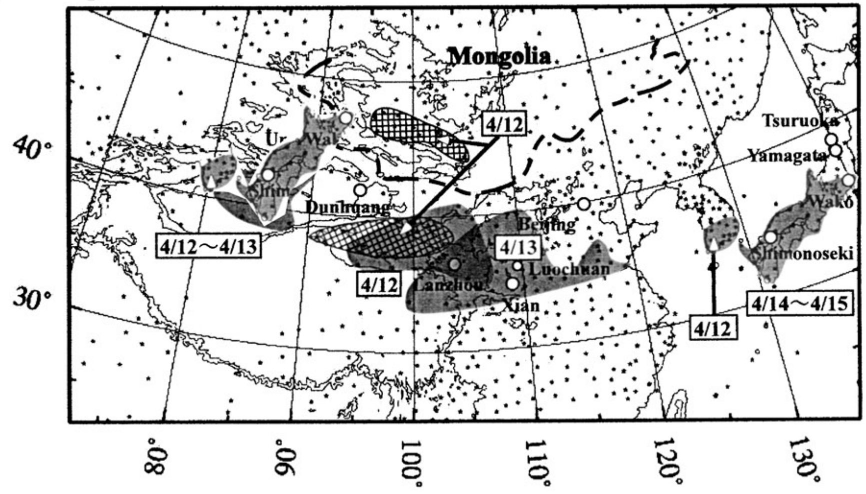


Fig. 2. Results of the potential source area determination for the major "Kosa Phenomenon" in 2000 from SYNOP data analysis. Basic map referred from Kurosaki (2001, pers. commun). (a) "Kosa Phenomenon" on 24 March, (b) on 8 April and (c) on 14 April.

salt and the contribution of the quartz filter blank. To obtain the total aerosol mass concentration, we measured the net weight of the aerosol samples collected at Wako. Among these, sample W000331 was collected from 31 March to 13 April, and shows the highest mass concentration, 5.72 $\mu g\,m^{-3}$. Although we did not measure the net weight of the total aerosol of the samples collected at the other sampling sites, the major and trace elements concentration in samples S000406, Y000406 and T000410, collected during almost the same period, clearly show the highest values. These high concentrations of total aerosol and major and trace elements are clearly related to the extensive "Kosa Phenomenon" on 8 April. Moreover, the sea-salt contributions (%) for Sr are relatively lower in the same period, even in the seaside cities, Shimonoseki (11.3%) and Tsuruoka (12.8%). This supports the idea that the contribution of the soil components is increased with the "Kosa Phenomenon". Although another "Kosa Phenomenon" was also observed at Wako on 14 April and Yamagata on 24 March, the major and trace element concentrations of the aerosols did not show any peculiar increase. This suggests that the "Kosa Phenomenon" does not necessarily lead to a significant change in aerosol composition.

The relationship between the soil components and the major and trace element concentrations are best discussed with reference to a plot of each element concentration against Al, which is one of the representative soil constitutive elements. In Fig. 3, the concentration of Mg, K, Fe, Rb, and Sr show a good linear correlation with Al, but this does not extend to Ca. This implies that the major and trace elements, with the exception of Ca, are mostly provided from soil particles with a similar element composition. The relatively low correlation between Ca and Al suggests that part of the Ca arises from a different source. We found that the aerosol from Wako contained more water-soluble Ca than those from the other sampling sites, and that the Ca from Wako during the non-"Kosa" period, was predominantly water-soluble. This suggests that a high degree of anthropogenic Ca exists in the urban atmosphere and that most of it is water-soluble. There are many possible sources of Ca in Japanese urban aerosols besides soil particles (KOSA, 1991): road dust, particulate matter derived from friction between asphalts and wheels; road deposits, mixed soil components from various local soils transported by automobiles with some anthropogenic materials, such as, road dust and asphalt; construction materials such as, concrete; and fly ash. The Ca component of these sources consists of soluble carbonate species, mainly soluble $CaCO_3$ (KOSA, 1991).

Conversely, the Ca concentration of the samples taken during the "Kosa" period (S000327, W000331, Y000406, and T000410) contained a significant quantity of insoluble Ca. In addition, the concentration of soluble Ca also increased with the insoluble Ca content during the "Kosa" period. This means that a considerable amount of Ca is transported along with "Kosa" as insoluble soil particles or as soluble salts.

3.4. Characteristics of Sr isotope composition

From March to April, the ratio of $^{87}Sr/^{86}Sr$ was higher in the seaside areas of Shimonoseki and Tsuruoka (0.7111–0.7126) than inland at Wako and Yamagata (0.7098–0.7116). This was the same as the characteristics observed in the chemical compositions: the highest $^{87}Sr/^{86}Sr$ ratio was observed in the aerosols that included the "Kosa Phenomenon" on 8 April, except for Yamagata (the highest value was 0.7117 in Y000413). From Table 4, we can see a positive relationship between the $^{87}Sr/^{86}Sr$ ratio and the major and trace element concentrations of each aerosol.

The $^{87}Sr/^{86}Sr$ ratio of the loess in China, which is thought to be the source of "Kosa" particles, is mainly higher than 0.710 (Gallet et al., 1996; Liu et al., 1994; Honda, 1999). Svensson et al. (2000) reported in their study for dust particles from a Greenland ice core that the possible source materials from Asia have $^{87}Sr/^{86}Sr = 0.715$–0.727. They used an extracted silicate fraction and the size-segregated fraction, $< 5\,\mu m$ (diameter), for the source identification in their study. The higher $^{87}Sr/^{86}Sr$ ratio during the "Kosa" period in this study also suggests the contribution of long-range-transported Asian dust particles.

To clarify the contribution of local components, we collected and analysed the four local soils and one aerosol. In Table 5, one can see that the $^{87}Sr/^{86}Sr$ ratios of the Wako surface soils are 0.7093–0.7095. Although the Ca and Sr concentrations of the road deposits are 2–4 times greater than those of the surface soils, there is no deference in the $^{87}Sr/^{86}Sr$ ratios for each. In addition, a typical local aerosol sample at Wako, W000918, has a similar value to them, $^{87}Sr/^{86}Sr = 0.7092$. Tanaka (1998) reported that the $^{87}Sr/^{86}Sr$ ratios of road deposits and surface soils from Yamagata and Tsuruoka, collected near the aerosol sampling points in this study, were 0.7059–0.7085. These results support the idea that the contribution of local sources cannot explain the increase of the $^{87}Sr/^{86}Sr$ ratio in the aerosols.

The three aerosol samples collected from July (W000721) to August (W000814) have a lower $^{87}Sr/^{86}Sr$ ratio (0.7085–0.7086) than the local aerosol sample from Wako (W000918). This feature may be explained by a contribution from volcanogenic materials, which erupted on Miyakejima Island in the Pacific Ocean, located 200 km south of Wako. The eruption had been intermittent since 8 July, and large-scale eruptions had occurred on 10 and 18 August, during the sampling period of samples W000721 and W000814. A contribution of volcanic material with a low $^{87}Sr/^{86}Sr$ ratio

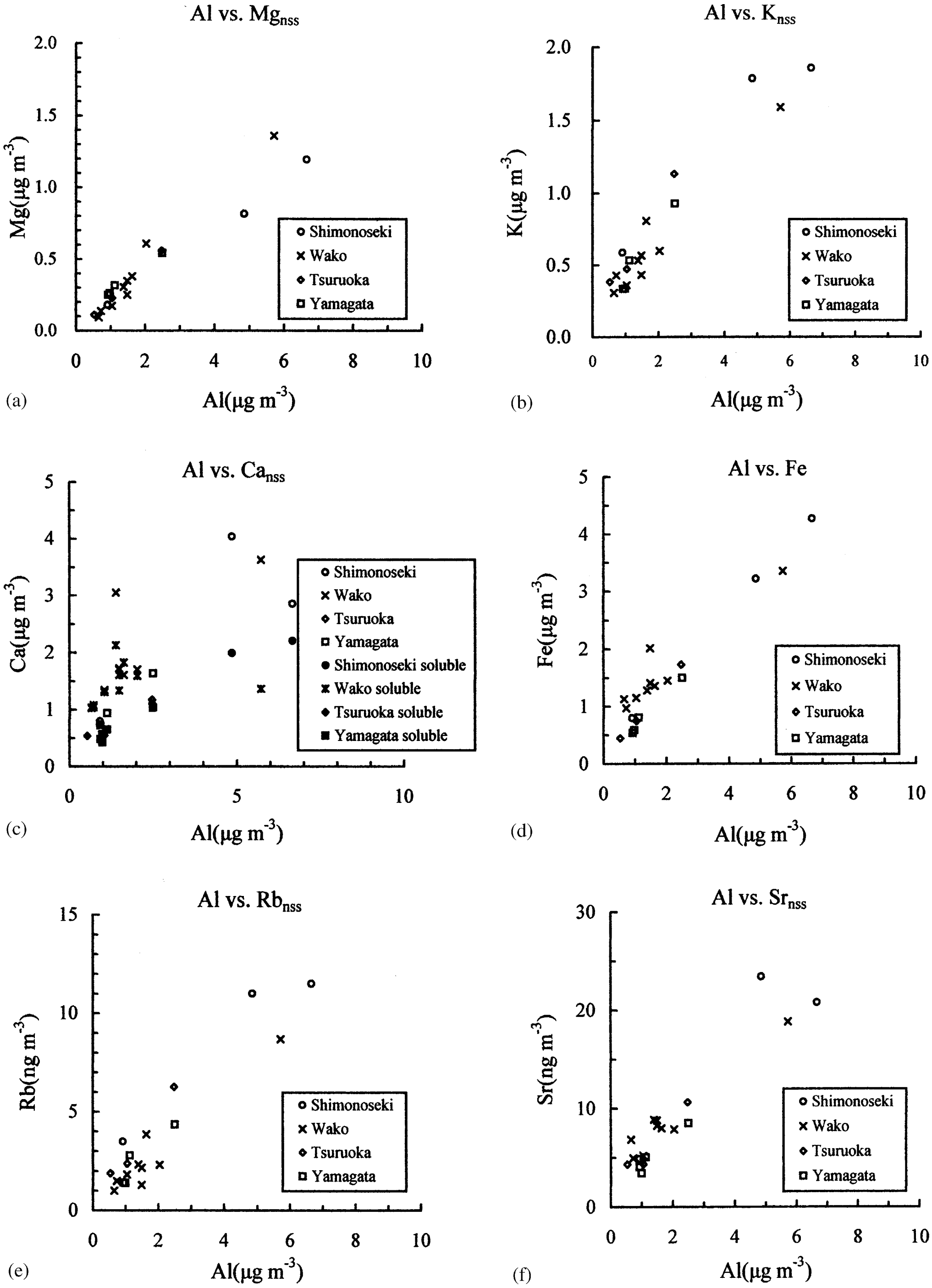


Fig. 3. Plots of the major and trace elements vs Al concentration for the aerosols collected over Japan in 2000: (a) Mg vs Al; (b) K vs Al; (c) Ca vs Al; (d) Fe vs Al; (e) Rb vs Al; and (f) Sr vs Al.

Table 5
The description and analytical results of the possible source components, mainly local soils

No.	Grain size (mm)	(%)						(ppm)		$^{87}Sr/^{86}Sr \pm 2\sigma$	$^{87}Rb/^{86}Sr$	Ca/Al
		Na	Mg	Al	K	Ca	Fe	Rb	Sr			
WRD-1	$<\phi$ 0.5	1.42	1.23	5.97	1.39	4.10	3.82	57.15	205.6	0.709509±24	0.80	0.69
WRD-2	$<\phi$ 0.5	1.36	1.30	5.36	1.28	3.48	4.12	52.57	189.1	0.709384±26	0.80	0.65
WS-1	$<\phi$ 0.5	0.67	0.93	7.69	1.14	1.24	5.21	22.17	101.2	0.709296±47	0.63	0.16
WS-2	$<\phi$ 0.5	0.62	1.30	8.67	0.95	0.96	6.12	41.16	90.1	0.709508±26	1.32	0.11
MYK-ash		1.66	2.03	8.03	0.42	6.50	7.88	4.51	249.3	0.703514±18	0.05	0.81

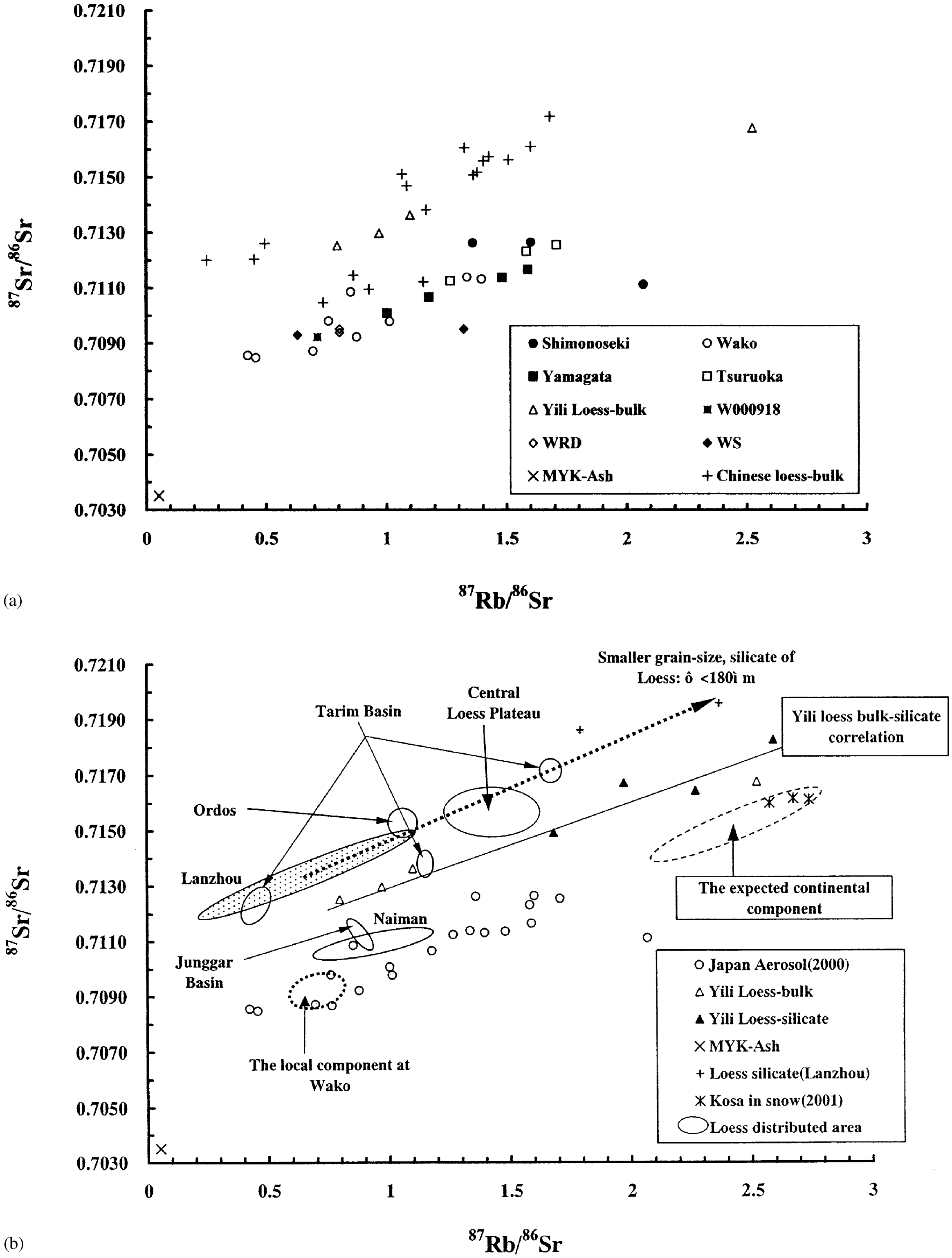
(0.7035) from Miyakejima ash (MYK-ash) would cause a decrease in the $^{87}Sr/^{86}Sr$ ratio. In addition, the contribution of the volcanic materials to the aerosols would result in an increase in the element concentrations of Al, Ca, Fe and Sr, as recognized in sample W000721. As a rough estimation for Sr isotopes, the decrease in the $^{87}Sr/^{86}Sr$ value (W000918 to W000721) can be explained by a 10% contribution of MYK-ash to the quantity of aerosol Sr.

3.5. Rb–Sr isotope system

The $^{87}Sr/^{86}Sr$ and $^{87}Rb/^{86}Sr$ ratios of the aerosols and soil analysed in this study are plotted in Fig. 4a, together with those of the loess from several areas in China, the possible source materials of the Asian dust. The linear relationship of the Rb–Sr isotope system shows the apparent age, and the apparent initial $^{87}Sr/^{86}Sr$ ratio. However, if some components with a different Rb–Sr isotope composition were mixed through the weathering and transportation processes, the relationship would show a pseudo-isochron, without any time significance, and the straight line would represent the mixing line of the two end members (Faure, 1986). At Wako, the Rb–Sr isotope compositions of the aerosols show a roughly linear relationship, and at the other three sites almost all the data fall on the line. This means that the Rb–Sr systematics of the aerosols over Japan can be interpreted as arising from the mixing of two components with different Rb–Sr isotope compositions: a low $^{87}Sr/^{86}Sr$ and $^{87}Rb/^{86}Sr$ ratio, and a high $^{87}Sr/^{86}Sr$ and $^{87}Rb/^{86}Sr$ ratio.

The possible lower end member of the aerosol mixing line is compatible with the local soils and the surface aerosol (W000918) at Wako, except for WS-2. In Fig. 4a, there is no difference between the road deposits and the surface soil, WS-1. The surface soil, WS-2, is clearly different from the surface aerosol (W000918), the surface soil, WS-1, and both of the load deposits, WRD-1 and WRD-2, even though WS-1 was sampled at the same point as the W000918 sampling point. This suggests that the composition of the local aerosols is less affected by the surface soils.

In Fig. 4a, all the data of Shimonoseki, Yamagata, and Tsuruoka only collected during the "Kosa" period is plotted on the corresponding area of the higher part of the mixing line. This result supports the idea that a higher end member is the Asian dust component. The expected end member of the Asian dust must be located in the area extended from the mixing line to the higher side of both isotopic ratios. However, the reported Rb–Sr isotopic ratios of Chinese loess tend to have lower $^{87}Rb/^{86}Sr$ ratios than the values of the expected end member in Fig. 4. Asahara et al. (1995) have demonstrated that an increase in the silicate fraction (the insoluble components in diluted HCl) of the loess results



(a)

(b)

Fig. 4. (a) A plot of the ^{87}Sr/^{86}Sr vs ^{87}Rb/^{86}Sr ratios for aerosol components collected over Japan in 2000. Mixing on this diagram is showed as a linear array. (b) A plot of ^{87}Sr/^{86}Sr vs ^{87}Rb/^{86}Sr ratios for aerosols and some source components from China. Data source: Loess in China: Luochuan, Gallet et al. (1996); Lanzhou (bulk and silicate), Asahara et al. (1999); Tarim basin, Ordos, Naiman and Junggar basin (the location is shown in Fig. 1), Honda (1999), Kosa in snow (2001); Kanayama et al. (2002).

in higher $^{87}Sr/^{86}Sr$ and $^{87}Rb/^{86}Sr$ ratios than the bulk components, because of the dissolution of Ca-bearing minerals contained in the Chinese loess. They also showed that a grain-size dependency of the silicate fraction of the loess on $^{87}Sr/^{86}Sr$ and $^{87}Rb/^{86}Sr$ ratios exists: the smaller grain size has higher ratios of both. The data of a silicate fraction of Yili loess (Table 6, to be discussed later) and Lanzhou loess (Asahara et al., 1999) is plotted on Fig. 4b, together with previously reported loess data. The data arrangement of aerosols and loess in Fig. 4b may suggest that the continental soil components, contributing to the end member of the Asian dust, consists of a silicate fraction, or a small-sized silicate fraction ($<180\,\mu m$) of Chinese loess, rather than a bulk component, although there is no reference data for the corresponding higher end member.

Recently, Kanayama et al. (2002) have reported that the Rb–Sr isotopic data for long-range-transported “Kosa” particles extracted from snow on 2 January, 2001, are $^{87}Sr/^{86}Sr = 0.7160–0.7162$ and $^{87}Rb/^{86}Sr = 2.58–2.74$. These values just correspond to the higher end member in this study, as plotted in Fig. 4b. By SYNOP data analysis for occurrence of dust storms, the sources of these “Kosa” particles were located at Mongolia and/or Inner Mongolia, China. As for the “Kosa” of 2000, the potential source areas also were mainly in Mongolia and/or Inner Mongolia, China, northward of 40°N. As shown in Fig. 4b, among the Chinese loess, we can see some differences in Rb–Sr isotopes from their distribution areas. Although the chemical and Sr isotopic data for surface soils in the northern areas are very scarce, the loess of the Naiman and Junggar Basin (Honda (1999): bulk and no size-segregated fraction) are plotted near the area of the Japanese aerosol, distinct from the loess of Central Loess Plateau or the Tarim Basin (around 40°N). If loess, distributed northward of 40°N, such as the Naiman and Junggar Basin (See Fig. 1), were the main source of “Kosa” in 2000, the expected end member of the continental material should be explained as containing some components of them.

3.6. *Ca/Al and Sr/Al ratios as chemical indices*

In the preceding studies of “Kosa”, it has been reported that the Ca content of aerosols collected over Japan during the “Kosa” period showed a remarkable increase, and Ca-bearing minerals such as $CaSO_4$ and $CaCO_3$ were recognized by X-ray diffraction (Nishikawa et al., 1991; KOSA, 1991). Many researchers have used soluble Ca as a chemical index for the “Kosa Phenomenon” (KOSA, 1991), as well as soluble Sr (Mori et al., 1996), because loess and desert sands in northwestern China are characterized by their high carbonate content (Hseung and Jackson, 1952). As for the dissolution of Ca-bearing minerals, many

Table 6
Analytical results of chemical and Sr-isotope compositions of bulk and silicate fraction of loess from Yili, China, by leaching experiment

Sample no.	Loess component	(%) Al	(%) Ca	(ppm) Rb	(ppm) Sr	$^{87}Sr/^{86}Sr \pm 2\sigma$	$^{87}Rb/^{86}Sr$	Ca/Al	Sr/Al ($\times 10^{-3}$)	Silicate/bulk Ca (%)	Silicate/bulk Sr (%)
Z-3	Bulk	5.67	8.31	77.1	280.3	0.712522±24	0.80	1.47	4.94	15.0	70.1
	Silicate	7.02	1.25	114.3	196.6	0.714935±17	1.68	0.18	2.80		
Z-191	Bulk	6.37	7.09	100.3	264.0	0.713628±21	1.10	1.11	4.14	16.9	58.3
	Silicate	7.74	1.20	104.9	153.9	0.716730±16	1.97	0.16	1.99		
L-4	Bulk	5.27	12.11	82.9	247.2	0.712973±21	0.97	2.30	4.69	9.9	63.0
	Silicate	7.82	1.20	122.2	155.8	0.716442±13	2.27	0.15	1.99		
L-7	Bulk	7.70	1.29	131.2	150.5	0.716678±17	2.52	0.17	1.96	62.2	97.8
	Silicate	7.82	0.80	131.8	147.2	0.718266±16	2.59	0.10	1.88		

scientists have suggested a possibility of chemical alteration during long-range transport (Levin et al., 1990; Chang et al., 1996). As a result of such a dissolution process during long-range transport via chemical reaction with acidic materials such as sulphate and nitrate, it has been proposed that the contribution of continental carbonate species may prevent the acidification of the atmosphere and precipitation (Nishikawa et al., 1991). Here, we will discuss the dissolution of Ca-bearing minerals.

In the current "Kosa" studies, Ca has been assumed as one of the soil constituent elements. However, Kadowaki (in KOSA, 1991) suggests that the high Ca/Al ratios of urban aerosols in Japan (0.55–2.2) are more affected by anthropogenic sources, such as road deposits (0.82–1.1) or asphalts (0.92–1.0), than by natural local soils (0.13–0.27). In this study, Ca/Al ratios also show higher values, 0.43–2.2, than natural local soil (Table 4). A relatively lower value, from 0.43 to 0.63, was obtained for the samples collected during the "Kosa" period (see Table 4). This means that a high Ca/Al ratio of an aerosol is not necessarily caused by Ca-rich "Kosa" from the arid and semi-arid regions of inland Asia. If anything, it is quite probable that the dissolution of Ca-bearing minerals has occurred during long-range transport, and that anthropogenic Ca was added to the aerosol. As Sr belongs to the alkaline earth elements (the same as Ca) and they have similar chemical properties, we tried to use the relationship of the ^{87}Sr/^{86}Sr isotope ratio and the Ca/Al or Sr/Al ratios to discuss the mixing of Asian dust and anthropogenic local components. For this discussion, typical loess samples from the Yili Basin, NW China, were leached with acetic acid. The ^{87}Sr/^{86}Sr isotope ratio and the Al and Ca concentrations of the silicate fraction (leached residue) and of the bulk samples are summarized in Table 6.

Fig. 5a shows the ^{87}Sr/^{86}Sr ratio vs the Ca/Al ratio, and Fig. 5b shows the ^{87}Sr/^{86}Sr ratio vs the Sr/Al ratio of the aerosol samples of this study together with the Chinese loess samples. In Fig. 5a, the data arrays of the aerosols seem to be arranged regularly, and they are plotted in a separate area of the bulk loess component from China, although it is difficult to find a clear relationship between the aerosols and the loess in Fig. 5b. In Figs. 5a and b, we supposed that there were two different end-member components, and drew a calculated mixing curve with the L-7 silicate as the higher end member of the continental component, and the surface aerosol at Wako, W000918, as the lower end member of the local aerosol component. In Fig. 5a, the data of the Japanese aerosols is plotted along the calculated mixing curve. This suggests that the contributed continental soil components of these "Kosa" aerosols in Japan are silicate fractions of loess rather than bulk fractions. In Fig. 5b, the relationship between the aerosols and both components of the loess is not so clear. The uncertainty of the profile of the Sr/Al ratios relative to that of the Ca/Al ratios can be caused by the difference in the chemical mobility of Ca and Sr. For the silicate/bulk (%) term in Table 6, we can see that Sr is more stable than Ca against acetic acid.

In Fig. 5, for the local source components at Wako, the ground level aerosol (W000918), road deposit (WRD), and surface soil (WS) are clearly separated, although there was no distinctive feature in the Rb–Sr isochron of Fig. 4. This also suggests that the local aerosols consist of not only surface soils but also some additional components, mainly anthropogenic materials in urban areas in Japan. This hypothesis is supported by the WRD samples, which are strongly affected by some anthropogenic components originating from asphalts and other sources. This is closer to W000918 of the local aerosol for the Ca/Al ratio rather than the WS samples.

Additionally, it is found from Fig. 5 that Wako aerosols in non-"Kosa" periods have higher Ca/Al ratios than the tentative lower end-member, W000918, in spite of there being no significant difference in ^{87}Sr/^{86}Sr ratios. This may be attributed to the collection altitude of both aerosols: W000918 was collected at ground level, and the others at about a height of 20 m, i.e. there is a preferential classification of Ca-minerals at a higher level. Alternatively, it is possible for a contribution of anthropogenic Ca-abundant particles such as construction materials, reported for Chinese aerosols by Zhang and Iwasaka (1999) and Zhang et al. (2000), from the environs. At least, it is obvious that there is some contribution of Ca-rich components (Ca/Al$>$1), having a value of around ^{87}Sr/^{86}Sr$=$0.709, to the urban aerosols in Japan.

3.7. An estimation of the mixing rate for aerosols in 2000

By considering the Sr isotope characteristics of the bulk components of the Japanese aerosols in 2000, we suggested a contribution of the Asian dust to the local aerosols during the "Kosa" period along with two other major components that contribute to the urban aerosol. One is the continental soil component, such as the Asian dust, the silicate fraction of the loess, and the other is the local aerosol component, which is affected largely by anthropogenic sources and has different chemical properties from surface soils. Here, we tried to approximate the contribution of the continental soil by using the ^{87}Sr/^{86}Sr ratio.

In Fig. 5, we show the mixing curves between a silicate fraction of the Asian loess (^{87}Sr/^{86}Sr$=$0.7180, Ca/Al$=$0.1 and Sr/Al$=$2) and the local aerosol components, W000918 (^{87}Sr/^{86}Sr$=$0.7092, Ca/Al$=$1.2 and Sr/Al$=$7). The ^{87}Sr/^{86}Sr ratio of the mixture is calculated

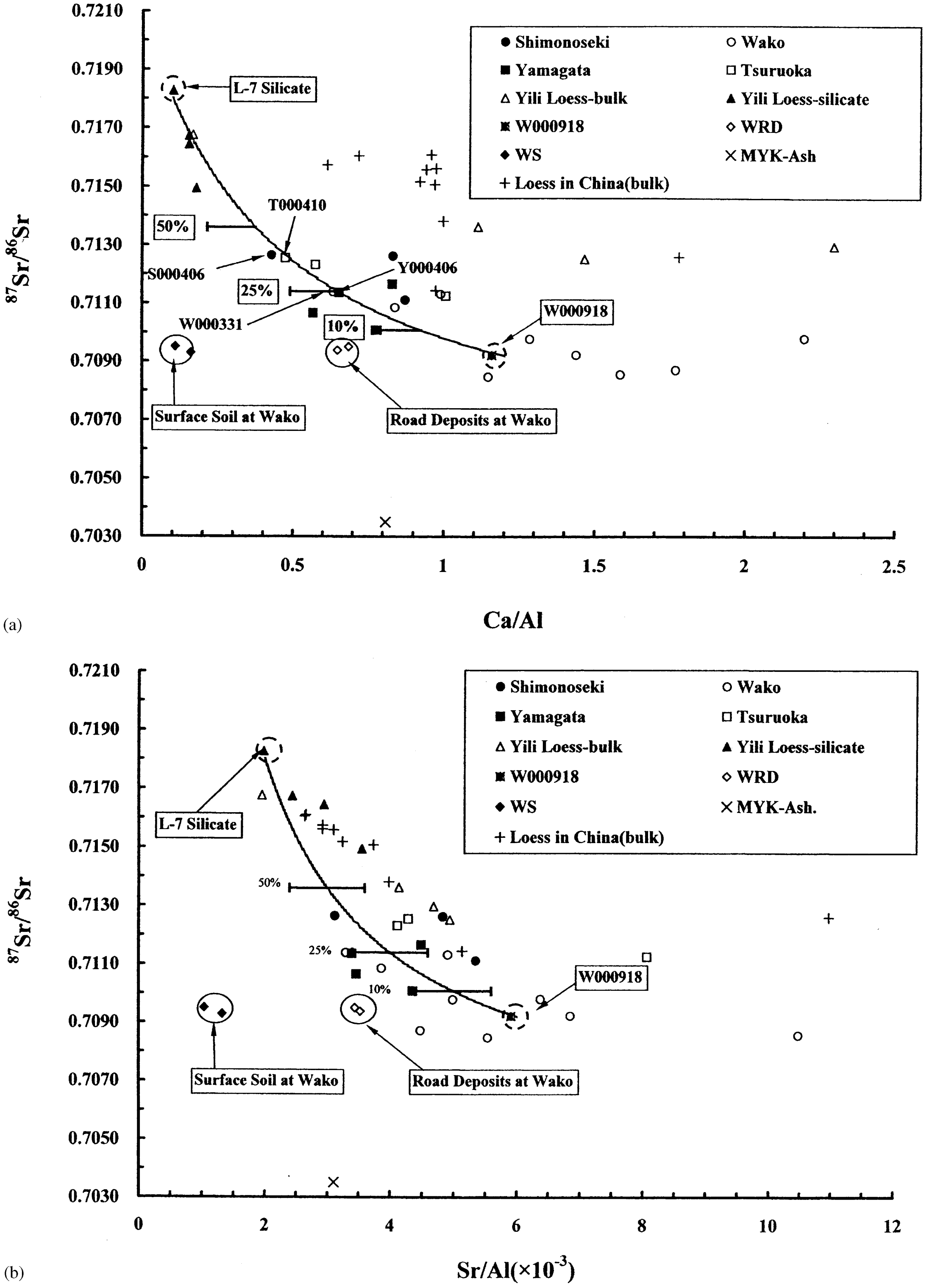


Fig. 5. (a) The ^{87}Sr/^{86}Sr vs Ca/Al ratio, and (b) the ^{87}Sr/^{86}Sr vs Sr/Al ratio for aerosol components over Japan. The end-member components are assumed to be as follows: a higher end member: ^{87}Sr/^{86}Sr = 0.718, Ca/Al = 0.1 and Sr/Al = 2, and a lower end member: ^{87}Sr/^{86}Sr = 0.7092, Ca/Al = 1.2 and Sr/Al = 6. Data source: same as for Fig. 3.

using the following equation:

$$\begin{aligned}({}^{87}\mathrm{Sr}/{}^{86}\mathrm{Sr})_{\mathrm{mixture}} = [\{({}^{87}\mathrm{Sr}/{}^{86}\mathrm{Sr})_{\mathrm{loess}} \times (\mathrm{Sr})_{\mathrm{loess}}\} \\ + \{({}^{87}\mathrm{Sr}/{}^{86}\mathrm{Sr})_{\mathrm{local}} \times (\mathrm{Sr})_{\mathrm{local}}\}] \\ \times 1/(\mathrm{Sr})_{\mathrm{mixture}}, \qquad (4)\end{aligned}$$

where $({}^{87}\mathrm{Sr}/{}^{86}\mathrm{Sr})_{\mathrm{mixture}}$ denotes the ^{87}Sr/^{86}Sr ratio of the mixing component. The end members for the Asian loess and local aerosol are given by $({}^{87}\mathrm{Sr}/{}^{86}\mathrm{Sr})_{\mathrm{loess}}$ and $({}^{87}\mathrm{Sr}/{}^{86}\mathrm{Sr})_{\mathrm{local}}$, respectively. The quantity $(\mathrm{Sr})_{\mathrm{mixture}}$ is the sum of $(\mathrm{Sr})_{\mathrm{loess}}$ and $(\mathrm{Sr})_{\mathrm{local}}$, and $(\mathrm{Sr})_{\mathrm{loess}}$ and $(\mathrm{Sr})_{\mathrm{local}}$ are quantities of Sr for each of the components and are variable parameters. Assuming the $(\mathrm{Sr})_{\mathrm{loess}}$ and $(\mathrm{Sr})_{\mathrm{local}}$, the quantity of Al and Ca for both components is defined by the Sr/Al and Ca/Al ratios.

As a result, a 25% contribution of the silicate fraction of the continental soil to the local aerosol gives $^{87}\mathrm{Sr}/^{86}\mathrm{Sr} = 0.7114$, $\mathrm{Ca/Al} = 0.65$, and $\mathrm{Sr/Al} = 4 \times 10^{-3}$. These values are consistent with the observed values of the sample collected during the "Kosa" period, W000331 ($\mathrm{Ca/Al} = 0.6$, and $\mathrm{Sr/Al} = 3.3 \times 10^{-3}$).

4. Conclusions

(1) The major and trace element concentrations and the ^{87}Sr/^{86}Sr ratios of the bulk component of aerosols increased when the "Kosa Phenomenon" was observed over Japan in 2000. This reflects the contribution of long-range-transported Asian dust.

(2) The Rb–Sr isotope systematics, and the profiles of ^{87}Sr/^{86}Sr vs the Ca/Al and Sr/Al ratios show a two-component mixing relationship: an Asian dust component with high ^{87}Rb/^{86}Sr and ^{87}Sr/^{86}Sr ratios, and low Ca/Al and Sr/Al ratios; and a local component in the Japanese urban areas with low ^{87}Rb/^{86}Sr and ^{87}Sr/^{86}Sr ratios, and high Ca/Al and Sr/Al ratios. Although the end member of the local component is roughly compatible with local aerosols at Wako, it is significantly affected by anthropogenic materials. The end member of the Asian dust component is inconsistent with the reported and referenced loess data.

(3) The Rb–Sr isotope systematics of the Asian dust component may reflect the source characteristics. This hypothesis is supported by the definitive Rb–Sr isotope systematics of the loess, distributed to the north of 40°N, which is a possible source area of the dust storms related to the "Kosa Phenomenon" in 2000, and the contribution of the silicate and/or smaller-size fraction rather than the bulk loess.

(4) The combination of ^{87}Sr/^{86}Sr and Ca/Al ratios suggests that the silicate fraction of the Chinese loess had contributed to the "Kosa" aerosols over Japan in 2000 as the Asian dust component. If the usually reported loess, containing a high concentration of these soluble minerals, had contributed to the "Kosa" aerosols, the Ca-bearing minerals should have been dissolved by some acidic materials during long-range transport.

(5) Our estimation using ^{87}Sr/^{86}Sr, Ca/Al, and Sr/Al ratios showed about a 25% contribution of the continental soil to the "Kosa" aerosol in April 2000.

(6) We propose that the Sr-isotope systematics in bulk aerosols reflects the source characteristics, and that the combination of this with chemical composition can explain the transportation process of Asian dust.

Acknowledgements

The authors appreciate the support of S. Sugihara (National Fisheries University) during the aerosol sampling, M. Maeda (Tokyo University of Fisheries) for providing the volcanic ash samples of Miyakejima and W. Ye (Zhejiang Normal University) for the loess samples. We are also grateful to M. Honda (Kyoto University) for his helpful discussions, and for help with the TIMS analysis. This study was partly supported through the Special Coordination Funds for promoting Science and Technology of MEXT, the Japanese Government.

References

Asahara, Y., Tanaka, T., Kamioka, H., Nishimura, A., 1995. Asian continental nature of ^{87}Sr/^{86}Sr ratios in north central Pacific sediments. Earth and Planetary Science Letters 133, 105–116.

Asahara, Y., Tanaka, T., Kamioka, H., Nishimura, A., Yamazaki, T., 1999. Provenance of the north Pacific sediments and process of source material transport as derived from Rb-Sr isotopic systematics. Chemical Geology 158, 271–291.

Biscaye, P.E., Dasch, E.J., 1971. The rubidium, strontium, strontium isotope system in deep-sea sediments: Argentine basin. Journal of Geophysical Research 76, 5087–5096.

Biscaye, P.E., Grousset, F.E., Revel, M., Van der Gaast, S., Zielinski, G.A., Vaars, A., Kukla, G., 1997. Asian provenance of glacial dust (stage 2) in the Greenland Ice Sheet Project 2 Ice Core, Summit, Greenland. Journal of Geophysical Research 102 (C12), 26765–26781.

Chang, Y.-S., Arndt, R.L., Carmichael, G.R., 1996. Mineral base-cation deposition in Asia. Atmospheric Environment 30 (13), 2417–2427.

Faure, G., 1986. Principles of Isotope Geology, 2nd Edition. Wiley, New York.

Gallet, S., Jahn, B.-M., Torii, M., 1996. Geochemical characterization of the Luochuan loess-paleosol sequence, China, and paleoclimatic implications. Chemical Geology 133, 67–88.

Herut, B., Starinsky, A., Katz, A., 1993. Strontium in rainwater from Israel: Sources, isotopes and chemistry. Earth and Planetary Science Letters 120, 77–84.

Honda, M., 1999. Geochemical, mineralogical and sedimentological studies on the desert deposits and loess around China. Ph.D. Thesis, Kumamoto University (in Japanese).

Hseung, Y., Jackson, M.L., 1952. Mineral composition of the clay fraction of some main soil groups of China. Soil Science Society of America, Proceedings 16, 294–297.

Husar, R.B., Tratt, D.M., Schichtel, B.A., Falke, S.R., Li, F., Jaffe, D., Gasso, S., Gill, T., Laulainen, N.S., Lu, F., Reheis, M.C., Chun, Y., Westpal, D., Holben, B.N., Gueymard, C., McKendry, I., Kuring, N., Feldman, G.C., McClain, C., Frouin, R.J., Merrill, J., DuBois, D., Vignola, F., Murayama, T., Nickovic, S., Wilson, W.E., Sassen, K., Sugimoto, N., Malm, W.C., 2001. Asian dust events of April 1998. Journal of Geophysical Research 106, 18317–18330.

IPCC, 1996. Climate change 1995. The science of climate change, Edited by J.T. Houghton, L.G. Meria Filho, B.A. Callander, N. Harris, A. Kattenberg, K. Marshall, Cambridge University Press. Cambridge, UK and New York, USA.

Ishizaka, Y., Ono, A., 1982. Mass size distribution of the principal minerals of yellow sand dust in the air over Japan. Idojaras 86, 249–253.

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 40 B, 494–503.

Kanayama, S., Yabuki, S., Fumitaka, Y., Abe, O., 2002. Geochemical features and source characterization from Sr isotopes of “Kosa” particles in Red Snow that fell on Yamagata Prefecture, NE Japan in January and March 2001. Journal of Arid Land Studies 11 (4), 291–300.

KOSA, 1991. In: Water Research Institute of Nagoya University (Ed.), KOSA. KOKON Press, Tokyo (in Japanese).

Levin, Z., Price, C., Ganor, E., 1990. The contribution of sulfate and desert aerosols to the acidification of clouds and rain in Israel. Atmospheric Environment 24A (5), 1143–1151.

Liu, T., Gu, X., An, Z., Fan, Y., 1981. The dust fall in Beijing, China on April 18, 1980. Geological Society of America Special Paper 186, 149–157.

Liu, C.-Q., Masuda, A., Okada, A., Yabuki, S., Fan, Z.L., 1994. Isotope geochemistry of Quaternary deposits from the arid lands in northern China. Earth and Planetary Science Letters 127, 25–38.

Matsuo, S., 1989. Geochemistry. Kodansha-Scientific, Japan (in Japanese).

Mori, I., Iwasaka, Y., Masataka, N., Quan, H., 1996. Non sea salt (Sr^{2+}/Ca^{2+}) in particulate matter: effective indicator for transportation of Kosa aerosols. Journal of Environmental Chemistry 6 (4), 567–573 (in Japanese).

Nakano, T., Tanaka, T., 1997. Strontium isotope constraints on the seasonal variation of the provenance of base cations in rain water at Kawakami, central Japan. Atmospheric Environment 31, 4243–4245.

Nishikawa, M., Kanamori, S., Kanamori, N., Mizoguchi, T., 1991. Ion equivalent balance in water soluble constituents of Kosa aerosol. Journal of Aerosol Research, Japan 6 (2), 157–164 (in Japanese).

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

Svensson, A., Biscaye, P., Grousset, F., 2000. Characterization of late glacial continental dust in the Greenland Ice Core Project ice core. Journal of Geophysical Research 105 (D4), 4637–4656.

Tanaka, M., 1998. Isotopic composition of strontium in aerosol collected from Tsuruoka and Yamagata. Masters Thesis, Yamagata University (in Japanese).

Tanaka, M., Yanagisawa, F., Kotani, T., 1998. Seasonal variation of the concentration of water-soluble components in dry deposition in Tsuruoka and Yamagata Prefecture, Japan. Journal of Arid Land Studies 8 (2), 209–213 (in Japanese).

Tegen, I., Fung, I., 1995. Contribution to the atmospheric mineral aerosol load from land surface modification. Journal of Geophysical Research 100, 18707–18726.

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

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.

Zhang, D., Shi, G.-Y., Iwasaka, Y., Hu, M., 2000. Mixture of sulfate and nitrate in coastal atmospheric aerosols: individual particle studies in Qindao (36°04′N, 120°21′E), China. Atmospheric Environment 34, 2669–2679.