

Atmospheric mineral particles collected at Qira in the Taklamakan Desert, China

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Abstract

Aerosol particles were collected in the situation of the widespread dust suspension on 21 February 1991 at Qira in the southern edge of the Taklamakan Desert, western China. The collected particles were examined by a transmission electron microscope equipped with an energy-dispersive X-ray (EDX) analyzer in order to obtain the size and elemental composition of individual mineral particles.

On the basis of EDX analyses for 386 particles, mineral particles were present in high number fractions (>99%) of particles in the radius range of 0.1–4 μm. Particles mainly composed of silicates comprised 76% of mineral particles. “Ca-rich” particles were detected in 7% of all the particles. Ca in the particles would be present not only as CaCO₃ but also as an internal mixture of CaCO₃ and CaSO₄. Particles containing halite (NaCl) were detected in number proportions of about 10% and were mainly present in the radius range of ≥0.5 μm. Some halite particles would be modified by chemical reactions with sulfuric acid.

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1. Introduction

Asian mineral particles play an important role in atmospheric radiation (e.g., Arai and Ishizaka, 1986) and cloud physical processes (e.g., Isono et al., 1959). Moreover, transport of these particles from land is also important to evaluate the sediments in the ocean floor (e.g., Duce et al., 1980).

Although studies on mineral particles in the desert and loess atmosphere of China have been carried out

(Zhang et al., 1993; Okada and Kai, 1995; Yabuki et al., 1998; Zhang et al., 1998; Okada et al., 2001; Yabuki et al., 2002), knowledge on atmospheric mineral particles in the atmosphere over the desert is still very meager. In particular, physico-chemical properties of the individual particles should be clarified in order to evaluate the effect on atmospheric processes. Moreover, the modification of mineral particles during long-range transport is an important issue to be clarified. Previous researches (e.g., Okada et al., 1987; Iwasaka et al., 1988; Okada et al., 1990; Zhou et al., 1996) suggested the modification of Asian dust-storm particles during transport from China to Japan. However, knowledge on the physical and chemical properties of individual dust particles over the source regions is very limited by now. The results of atmospheric mineral particles over

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desert areas by single-particle analysis have also become basic information for evaluating the modification of mineral particles during long-range transport.

The aim of this paper is to obtain the composition of individual atmospheric mineral particles collected at Qira in the Taklamakan Desert during the period of the widespread dust suspension in February 1991.

2. Method

The Taklamakan Desert is located in the central part of Asia and surrounded by mountains with altitudes of 4000–5000 m except for the eastern side. Collection of aerosol particles was carried out on 21 February 1991 at Qira (37°01'N, 80°44'E; 1380 m altitude), which is located in the southern edge of the desert.

Individual aerosol particles were collected on carbon-coated nitrocellulose (collodion) films with an impactor of 1-mm-diameter jet. A volumetric air flow-rate of 1.3 l min⁻¹ with a collection time of 10 min was adopted. The evaluation of the collection efficiency was carried out by using experimental results of Ranz and Wong (1952) on the assumption of mineral particle density of 2.6 g cm⁻³. This impactor has a collection efficiency of 50% at 0.37 μm radius and 100% at ≥0.55 μm radius.

Particles collected on a carbon film were coated with Pt/Pd alloy (7 Å in thickness) at a shadowing angle of arctan 0.5 and were examined with a transmission electron microscope (Hitachi, H-600) in order to assess the size and morphology. Elemental composition of particles on carbon film was investigated with an energy-dispersive X-ray (EDX) analyzer (KeveX, delta 5) equipped with the microscope. The electron beam was irradiated at the center of a particle at an accelerating voltage of 50 kV. The X-ray spectrum was obtained through a KeveX UTW (ultra thin window) detector with a counting time of 50–100 s. For the quantitative

analysis of the individual particles with the EDX, a thin film method (Cliff and Lorimer, 1975) was applied using KeveX QuantexTM software for energy-dispersive microanalysis. The weight percentages of elements Na, Mg, Al, Si, S, Cl, K, Ca, Ti and Fe were calculated on the basis of characteristic X-ray fitted with a Gaussian distribution, not the peak intensity. Quantitative analysis was applied to the particle with thickness less than 2 μm in order to reduce the absorption of generated characteristic X-ray in the particle. Radius of a particle (r) was evaluated by averaging the long width (a) and the orthogonal width (b) as follows: $r = (a + b)/4$.

3. Results

3.1. Meteorological situation during aerosol collections

The collection periods and meteorological situation are indicated in Table 1. Figs. 1a and b show the time changes in visibility and air temperature during the period from 15 to 23 February 1991 at Qira Meteorological Observatory, respectively. Visibility before 18 February is more than 40 km. A rapid decrease in visibility is found after 18 February in the association with the invasion of cold air. Snowfall was observed during the period from 02 to 04 GMT on 18 February. Minimum visibility of 1.5 km was observed at 06 and 12 GMT on 19 February. Although visibility after the rapid decrease tends to increase gradually, the values are limited to be less than 10 km in the period. Widespread dust suspension in air (not raised by wind at the time of meteorological observation) was observed continuously after 09 GMT on 18 February. The collections were carried out in the situation of the widespread dust suspension with a visibility of 6 km.

Table 1
Sampling period and meteorological condition at Qira on 21 February 1991

Sample ID	Collecting film	Sampling period ^a (GMT)	Meteorological condition ^b					Remarks
			T (°C)	Td (°C)	WD	WS (ms ⁻¹)	Visibility (km)	
TAKA	Carbon film	04:18–04:28	-3.0	-6.0	carm		6	Widespread dust in suspension WW ^c =06 (06:00 GMT)
TAKB	Carbon film	09:20–09:30	-2.0	-5.4	NNE	2	6	Widespread dust in suspension WW ^c =06 (12:00 GMT)

^aTime differences between GMT and BST, and between GMT and LST are 8 h and 5.5 h, respectively.

^bThe data were obtained at Qira Meteorological Observatory.

^cInternational meteorological code of present weather.

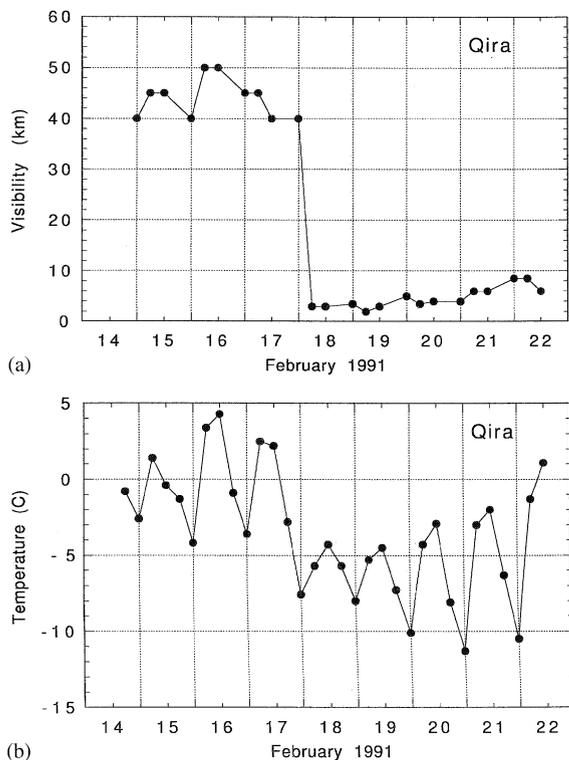


Fig. 1. Time changes in (a) visibility and (b) air temperature during the period from 15 to 23 February 1991 at Qira Meteorological Observatory.

3.2. Features of particles

Fig. 2 shows an electron micrograph of aerosol particles collected during the period from 09:20 to 09:30 GMT on 21 February 1991 (sample TAKB), along with some EDX spectra. As found in Fig. 2, particles made up of electron-opaque material with irregular shape are dominant in the radius range of 0.1–2 μm . The statistics of aspect ratio, height-to-width ratio and circularity factor of mineral particles collected at Qira were shown elsewhere (Okada et al., 2001). The X-ray spectrum from particle B shows the presence of mineral elements of Mg, Al, Si, K, Ti and Fe. The X-ray spectrum from particle B shows the same elements as particle A, with the exception of the dominant peak of Fe. Particle D indicates the dominant peaks of Ca, along with small peaks of Si and Fe. Particle C shows dominant peaks of Na and Cl. Though not shown here, particles which show the presence of Na and Cl were determined to be NaCl by electron diffraction. Particles with elements Na and Cl were not detected abundantly in Zhangye, northwestern China (Okada and Kai, 1995).

3.3. Types of individual particles

On the basis of the quantitative results by EDX analyses, all the particles (386 particles) in the radius range of 0.1–4 μm were classified into eight types. Table 2 shows the results of particle classification for each sample.

“Si-rich” particles (which have the maximum weight ratio of Si in analyzed elements with atomic numbers ≥ 11) are present in 72–80% of all the particles in two samples. “Si-dominant” particles, which were determined by Si weight ratio, $P(\text{Si})$, of ≥ 0.65 , would be mainly composed of quartz (SiO_2). The weight ratio of element X, $P(X)$, is as follows:

$$P(X) = X / (\text{Na} + \text{Mg} + \text{Al} + \text{Si} + \text{S} + \text{Cl} + \text{K} + \text{Ca} + \text{Ti} + \text{Fe}).$$

Most of the “Si-rich” particles are those containing Si and Al (the second most abundant element) (see the spectrum of particle B in Fig. 2). “Si-rich” particles are considered to be mainly composed of silicates.

“Ca-rich” particles are detected in 6–9% of all the particles. “Ca-dominant” particles (see the spectrum of particle D in Fig. 2) are classified if the particles have $P(\text{Ca}) \geq 0.65$. “Ca-dominant” particles are considered to be mainly composed of CaCO_3 . This is consistent with the findings of Hseung and Jackson (1952) and Yabuki et al. (2002). “Ca+Si” particles were also collected, suggesting that CaCO_3 was mixed internally with silicates.

In desert soil, the weight percentage of CaSO_4 is in large values up to 14% (Hseung and Jackson, 1952). As shown in Table 2, “Ca+S” particles are present in approximately 20% of “Ca-rich” particles. One of the possible materials in the particles is considered to be gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$). Ca was detected as the second most abundant element in one “S-rich” particle.

“Cl-rich” and “Na-rich” particles were detected in approximately 10% of total particles. Though not shown here, the backward trajectory from 500 m height above Qira at the time of sampling showed that the air parcel did not arrive over oceanic areas within five days. The presence of atmospheric halite particles in the Taklamakan Desert was reported by Molnár et al. (1993) and Yabuki et al. (1998, 2002). Hence, “Cl-rich” and “Na-rich” particles were suggested to be mainly composed of halite. “Na-rich” particles contained S as the second most abundant element, together without Cl. One “S-dominant” particle was detected from the two samples. This particle was evaporated upon the irradiation of electron beam. Hence, it would be composed of sulfate, not originating from mineral sources. As found from Table 2, there are other types of mineral particles such as “Mg-rich”, “Ti-rich”, “Fe-rich”. On the basis of the particle classification, mineral particles were present

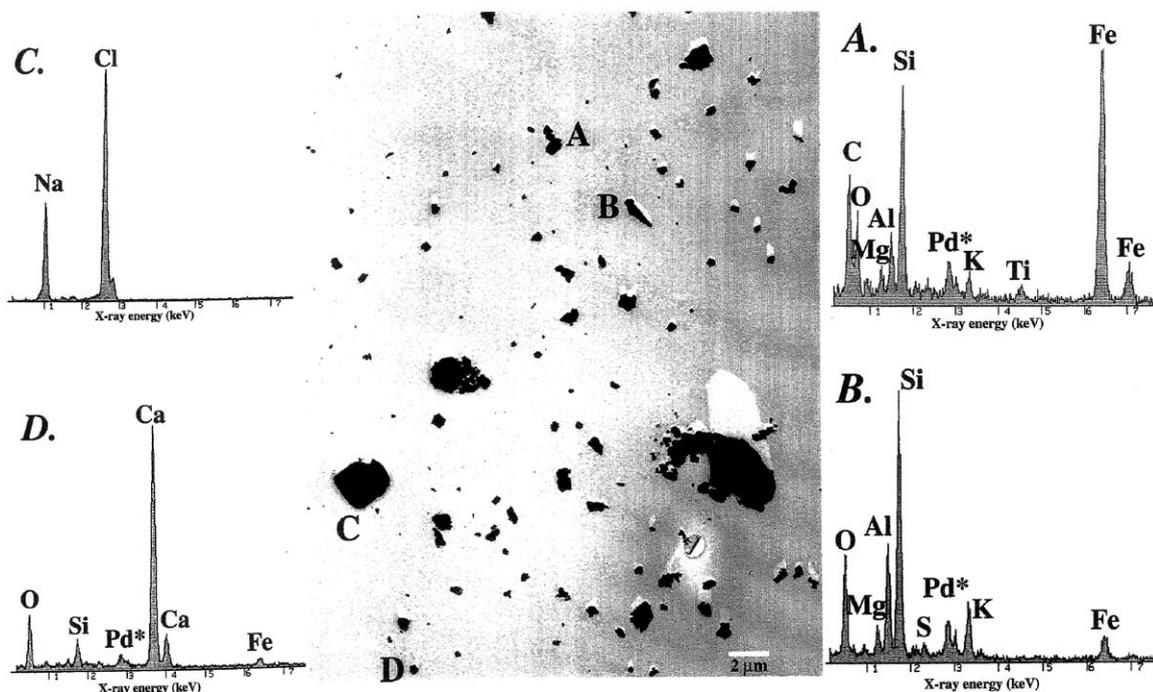


Fig. 2. Electron micrograph of aerosol particles collected during the period from 09:20 to 09:30 GMT on 21 February 1991 (sample TAKB), along with some EDX spectra.

in high number fractions (>99%) of particles in the radius range of 0.1–4 μm .

The abundance of types of mineral particles at Qira is compared with that obtained in spring and summer at Zhangye (39–40°N, 100°E) (Okada and Kai, 1995). Particle collections at Zhangye were carried out near/in the southern border of the Badain-Jaran Desert. The results at Zhangye (487 particles analyzed) were obtained from the analyses of individual particles in the radius range similar to that at Qira. Mineral particles at Zhangye were present in about 98% of aerosol particles of 0.1–6 μm radius. For particles other than those with halite, the abundance of particle types showed no large differences between the two locations. The collected particles will represent mineral particles of the desert atmosphere.

3.4. Calcium in particles

The presence of Ca in the desert and loess soils in China was reported by Hseung and Jackson (1952), Liu et al. (1985), Tanaka et al. (1986), Suzuki and Tsunogai (1988), Yabuki et al. (1998) and Mori et al. (1999). Besides, atmospheric aerosol particles collected from the desert and loess areas in China contained high weight proportions of Ca (Molnár et al., 1993; Zhang et al., 1993; Okada and Kai, 1995; Ren et al., 1995; Yabuki et al., 2002). However, the state of mixture

of Ca in atmospheric mineral particles is not fully clarified.

Fig. 3 shows Ca weight ratio, $P(\text{Ca})$, in individual particles as a function of radius. In the whole radius range, particles with $P(\text{Ca})$ greater than 0.1 and 0.02 are present in 12% and 19%, respectively. And 3% of particles are “Ca-dominant” particles with $P(\text{Ca}) \geq 0.65$. The results shown in Fig. 3 imply that most of the Ca is mixed internally with other minerals. The number fraction of particles with $P(\text{Ca}) \geq 0.65$ tends to decrease with increasing radius, that is, the fraction is 4% (0.1–0.5 μm radius), 2% (0.5–1.0 μm radius) and 1% (1.0–4.0 μm radius). For particles with $P(\text{Ca}) \geq 0.1$, the number fraction is 10% (0.1–1.0 μm radius) and 23% (1.0–4.0 μm radius). Features of the size dependence of $P(\text{Ca})$ are similar to those collected in spring at Zhangye (Okada and Kai, 1995).

In order to estimate the mixture degree of Ca in individual mineral particles, relative weight ratios of Al, Si and Ca are shown in Fig. 4. The data for halite-containing particles (shown later) and one “S-dominant” particle were not used for the figures. Fig. 4a shows the relative weight ratios for 301 particles of 0.1–1.0 μm radius. Most of the data are located on the line between Al and Si with the relative weight ratio of Al less than 0.5. Particles with the relative weight ratios of Ca ≥ 0.2 are present in number percentage of 13% in the particles used. Particles with the ratios of Ca ≥ 0.8

Table 2
Types of aerosol particles in the radius range of 0.1–4.0 μm evaluated by the EDX analysis

Type of particles	Averaged major elements ^a (wt%)	Number of particles	
		Sample TAKA	Sample TAKB
Mg-rich	Mg (26), Cl (18), Si (17)	1 (1)	0
Si-rich			
Si-dominant ^b	Si (84 \pm 11), Al (8 \pm 7)	6	13
Si + Al	Si (50 \pm 6), Al (27 \pm 5), K (8 \pm 4)	98 (11) ^c	106 (8)
Si + Mg	Si (49 \pm 6), Mg (26 \pm 6), Al (10 \pm 5)	5	8
Si + Ca	Si (34 \pm 7), Ca (27 \pm 6), Al (11 \pm 6)	7 (2)	3
Si + Fe + Al	Si (43 \pm 10), Fe (24 \pm 5), Al (17 \pm 4)	17 (2)	25 (2)
others		3 (1)	3
Ca-rich			
Ca-dominant	Ca (81 \pm 12)	6 (1)	5
Ca + Si	Ca (46 \pm 14), Si (24 \pm 5)	7 (5)	3
Ca + Mg	Ca (56 \pm 6), Mg (31 \pm 2)	1	1
Ca + S	Ca (50 \pm 4), S (41 \pm 7)	3 (1)	2 (1)
Ti-rich			
Ti-dominant	Ti (73)	1	0
Fe-rich			
Fe-dominant	Fe (80 \pm 7)	2	2
Fe + Si	Fe (43 \pm 12), Si (30 \pm 4), Al (15 \pm 6)	6	6
Na-rich			
Na + S	Na (40 \pm 24), S (30 \pm 15)	1 (1)	1
Cl-rich			
Cl + Na	Cl (58 \pm 4), Na (37 \pm 3)	20 (6)	20 (13)
Cl + Si	Cl (30), Si (26)	1 (1)	0
S-rich			
S-dominant	S (98)	1	0
S + Mg	S (48), Mg (33)	1 (1)	0
S + Ca	S (46), Ca (45)	1 (1)	0
Total number		188 (34)	198 (24)

^aAveraged weight percent and standard deviation.

^bX-dominant: the weight percent (wt%) of element X larger than 65%.

^cThe number of particles larger than 1 μm is indicated in parenthesis.

occupy 35% of particles with those of $\text{Ca} \geq 0.2$. For 34 coarse particles (Fig. 4b), particles with the relative weight ratios of $\text{Ca} \geq 0.2$ constitute 32% of the particles plotted. And particles with the ratios of $\text{Ca} \geq 0.8$ occupy 26% of particles with those of $\text{Ca} \geq 0.2$. These results suggest that the degree of internal mixture of Ca is larger in coarse mineral particles than in submicrometer mineral particles.

3.5. Halite particles

Fig. 5 shows Na weight ratio, $P(\text{Na})$, in individual particles as a function of radius. The data exhibit the presence of $P(\text{Na})$ around 0.4 in the radius range of

0.5–3 μm . For particles with radii less than 0.5 μm , there are few particles with ratios larger than 0.2. These results are different from those for Ca (Fig. 3).

Yabuki et al. (2002) reported that feldspar was detected from their samples collected at Qira. One of the kinds of feldspar is albite ($\text{NaAlSi}_3\text{O}_8$) which contains Na. In order to remove Na-containing minerals other than halite, particles with weight ratios of Na/Si larger than 0.25 (Niimura et al., 1994) and $P(\text{Na}) \geq 0.02$ were determined to be halite-containing particles. Fig. 6 shows the weight ratio of Cl/Na in halite-containing particles as a function of radius. The total number of particles is 49. The data concentrate in the radius range of $>0.5 \mu\text{m}$. The weight ratios of Cl/Na are located

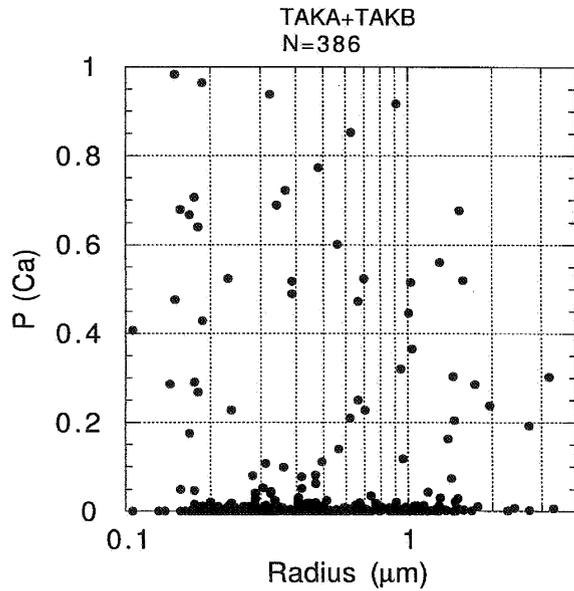


Fig. 3. Ca weight ratio, $P(\text{Ca})$, in individual particles as a function of radius. Total number of particles is 386.

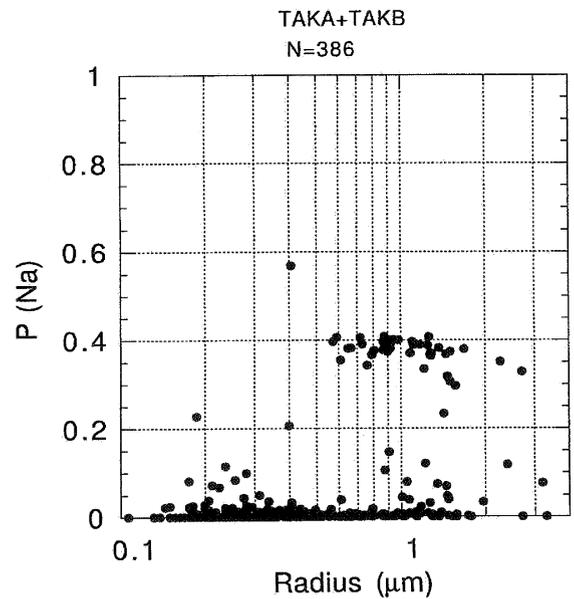


Fig. 5. Na weight ratio, $P(\text{Na})$, in individual particles as a function of radius. Total number of particles is 386.

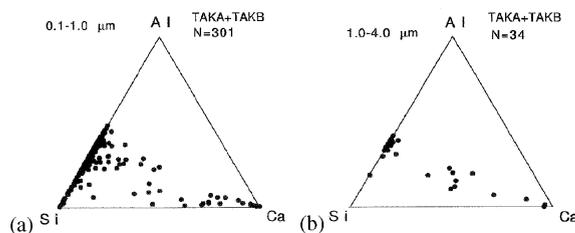


Fig. 4. Relative weight ratios of Al, Si and Ca for (a) 301 particles of 0.1–1.0 μm and (b) 34 particles of 1.0–4.0 μm . The data for the particles with halite and “S-dominant” particle were not used.

around the ratio for halite (1.54). However, there are particles with ratios less than 1 and larger than 2. Note that particles without Cl (complete Cl loss) are detected in 6% of halite-containing particles.

3.6. Sulfur in mineral particles

Fig. 7 shows the S weight ratio, $P(\text{S})$, in individual particles as a function of radius. The data for halite-containing particles are shown by open circles. One “S-dominant” particle of 0.64 μm radius shows the ratio of 0.99. Other data are present in $P(\text{S})$ lower than 0.5. In the whole radius range, particles with $P(\text{S}) \geq 0.1$ and ≥ 0.02 are present in 4% and 6% of mineral particles, respectively. We classified mineral particles to those with and without halite and evaluate the $P(\text{S})$ values for each group. Fig. 8 shows the histograms of $P(\text{S})$ for mineral particles with and without halite. The numbers of

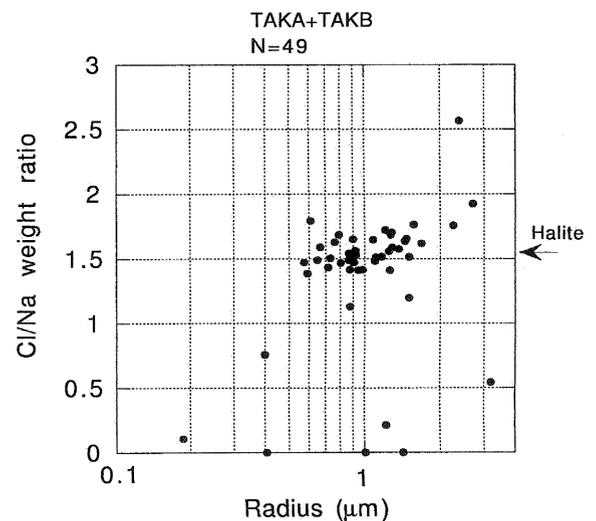


Fig. 6. Weight ratio of Cl/Na in halite-containing particles as a function of radius. Particles with weight ratios of Na/Si larger than 0.25 and $P(\text{Na}) \geq 0.02$ were determined to be halite-containing particles. Total number of particles is 49.

particles with and without halite are 336 and 49, respectively. For mineral particles without halite, the data showing $P(\text{S}) \geq 0.1$ were present in 3% of the particles. On the other hand, 12% of halite-containing mineral particles exhibited $P(\text{S}) \geq 0.1$. The difference in $P(\text{S})$ between two types of mineral particles implies that halite-containing particles tend to contain S content larger than those without halite.

4. Discussion

We will discuss S content in two types of mineral particles. At first, S in mineral particles without halite is examined. Figs. 9a and b show the relative weight ratios of Al, S and Ca for halite-free mineral particles of 0.1–1.0 μm radius and 1.0–4.0 μm radius, respectively. The data between Al and CaSO_4 shown by a dotted line

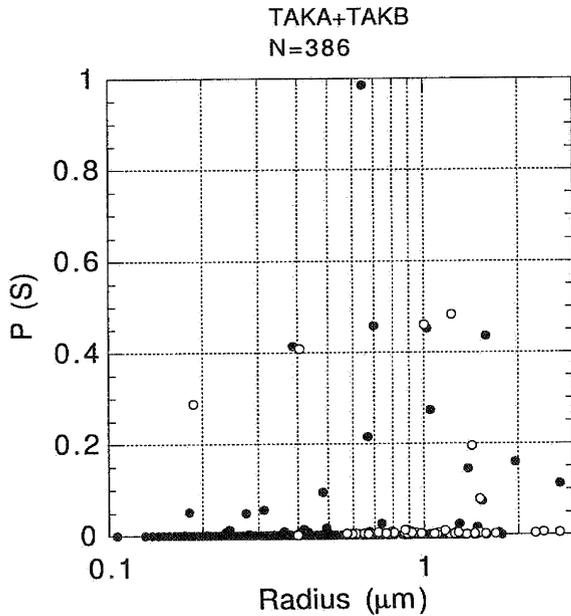
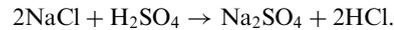


Fig. 7. S weight ratio, $P(S)$, in individual particles as a function of radius. The data for halite-containing particles are shown by open circles. Total number of particles is 386.

indicate the constant weight ratios of S/Ca (0.8) for gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$). Although there are several particles with S/Ca ratios similar to gypsum in the two size ranges, most of the data are located close to a line between Al and Ca, indicating low relative weight ratios of S. Particles with the relative weight ratios of $S \geq 0.1$ constitute 3.3% of particles with radii 0.1–1.0 μm and 18% of particles with radii 1.0–4.0 μm . This indicates the higher abundance of S in coarse mineral particles. These results imply that Ca in mineral particles is present not only as CaCO_3 but also as an internal mixture of CaCO_3 and CaSO_4 . Since desert soil particles contain relatively large amounts of S in the form of CaSO_4 (Hseung and Jackson, 1952; Tanaka et al., 1986; Yabuki et al., 1998, 2002), in the present study we could not judge the modification of CaCO_3 by chemical reactions with sulfuric acid. However, the information will be useful for studying the modification of mineral particles during long-range transport.

Next, the relative weight ratios of Na, S and Cl for halite-containing particles are shown in Fig. 10. As found in Fig. 10, most of the data are located around the ratio for halite. However, six particles (12% of halite-containing particles) show low relative weight ratios of Cl. There are three particles with complete Cl losses. These particles contain the ratios between S and Na_2SO_4 , indicating that the halite particles have been entirely modified and then have accumulated excess S with respect to Na_2SO_4 . As is known, NaCl could be modified by the following chemical reaction:



Hence, the results in Fig. 10 imply the occurrence of chemical reactions between halite and sulfuric acid in the

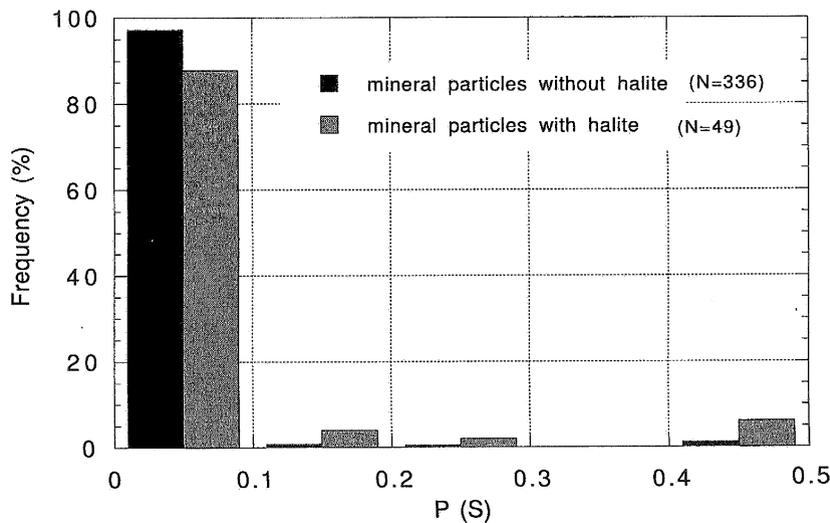


Fig. 8. Histograms of $P(S)$ for mineral particles with and without halite. The numbers of particles with and without halite are 336 and 49, respectively. The data was plotted in the $P(S)$ ranges of 0–0.1, 0.1–0.2, 0.2–0.3, 0.3–0.4 and 0.4–0.5.

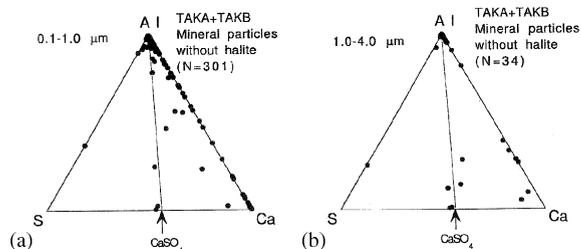


Fig. 9. Relative weight ratios of Al, S and Ca for halite-free mineral particles of (a) 0.1–1.0 μm radius and (b) 1.0–4.0 μm radius. The number of particles is 301 for particles of 0.1–1.0 μm radius and 34 for particles of 1.0–4.0 μm radius. The data between Al and CaSO_4 shown by a dotted line show the constant weight ratios of S/Ca (0.80) for gypsum ($\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$).

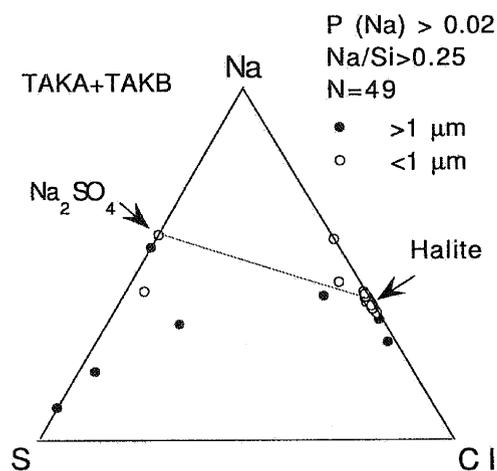


Fig. 10. Relative weight ratios of Na, S and Cl for halite-containing particles. The data for particles of $< 1 \mu\text{m}$ radius and particles of $> 1 \mu\text{m}$ radius were indicated by open and closed circles, respectively. Chemical reaction between halite and sulfuric acid produces the data on a dotted line between halite and Na_2SO_4 . Total number of particles is 49.

atmosphere which produced the Cl losses, and of further heterogeneous processes which produce the excess S with respect to Na_2SO_4 . Although the origin of sulfur is not clarified in this study, one of the possible origins is considered to be anthropogenic sources in the cities including Qira around the Taklamakan Desert.

5. Concluding remarks

Aerosol particles were collected in the situation of the widespread dust suspension on 21 February 1991 in the surface atmosphere of Qira in the southern edge of the Taklamakan Desert, western China. The collected

particles were examined by a transmission electron microscope equipped with an EDX analyzer in order to obtain the size and elemental composition of individual mineral particles. The weight percentages of elements Na, Mg, Al, Si, S, Cl, K, Ca, Ti and Fe in individual particles were obtained using the EDX spectra.

On the basis of EDX analyses for 386 particles, mineral particles were present in high number fractions ($> 99\%$) of particles in the radius range of 0.1–4 μm , that is, only one “S-dominant” particle was detected as the particle composed of sulfate other than minerals. Particles mainly composed of silicates comprised 76% of mineral particles. “Ca-rich” particles were detected in 7% of all the particles. Particles containing halite (NaCl) were detected in number proportions of about 10% and they were mainly present in the radius range larger than 0.5 μm . Determination of the cause for the presence of halite in the limited radius range remains a task for the future. Some halite particles would be modified by chemical reactions with sulfuric acid.

Elemental composition of mineral particles was examined with special interest in the presence of S for halite-free mineral particles. It was considered that Ca in the particles was present not only as CaCO_3 but also as an internal mixture of CaCO_3 and CaSO_4 . In the present study, we could not judge the modification of CaCO_3 by chemical reactions with sulfuric acid. However, the information will be useful for studying the modification of mineral particles during long-range transport.

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