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Chemical-Physical Transformation of KOSA (Asian Dust) Particles: The Effects on Atmospheric Environment in East Asia

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Abstract - Measurements of morphology and chemical elements of aerosol were made in August of 2002 and March of 2003 at Dunhuang ($40^{\circ}00'N$, $94^{\circ}30'E$), China, on the basis of direct sampling of free tropospheric aerosols with a balloon-borne particle impactor, to understand mixing state of dust particles in spring over the desert areas in the Asian continent. Electron microscopic experiments of the particles directly showed that dust particles were major constituents of coarse mode particles in spring time in the free troposphere over the Taklamakan desert. Si-rich particles and Ca-rich particles were typical types of those dust particles, and the values of [number concentration of Ca-rich particles]/[number concentration of Si-rich particles] showed large difference between spring and summer: about 0.3 in spring, 2003 and about 1.0 in summer, 2002 in height of 3-5km above the sea level. It is likely that situation of the ground surface and strength of vertical mixing are potential factors causing the difference. It is, comparing chemical compositions of the particles with those of particles collected over the Japan, strongly suggested that chemical modifications of particles occurred during long-range transport in the free troposphere. Analysis of wind systems showed that combination of predominating westerly in the free troposphere and surface wind strongly controlled by geographical structure of the Tarim basin was important process characterizing long-range transport KOSA particles originating in the Taklamakan desert.

I. Introduction

Asian dust particles (KOSA particles, KOSA literally means yellow sand in Japanese), according to recent many investigations, make important contribution to changes in regional and/or global climate and environment since particles can scatter and/or absorb solar radiation [1, 2], act as chemical reaction site [3, 4, 5] and play as cloud condensation nuclei or ice nuclei [6, 7, 8] in the atmosphere. Long-range transport of KOSA particles and chemical-physical transform of KOSA particles during transport, therefore, becomes a matter of great concern and many campaigns such as ACE-Asia including various type of field observations have been conducted [9].

Many lidar measurements made in Japan showed that long-range transport of KOSA particles was extremely active in the free troposphere over East Asia and west Pacific regions in spring [10, 11, 12, 13], and Taklamakan desert has been suggested as possible one of strong sources of KOSA particles.

Some investigators, on the basis of lidar measurements, suggested that weak KOSA events which were so small that we could not detect near the ground, in addition to severe KOSA events, made possible contribution of geochemical cycle of minerals [10, 11]. Aircraft-borne measurements also confirmed effect of weak KOSA events on chemical composition of particulate matter in the free troposphere over Japan [14]. More recently weak KOSA events were suggested even in summer when few effects of Kosa on atmospheric aerosols over Japan and Pacific ocean have been believed since Pacific high covered west Pacific region and westerly wind largely [15, 16, 17].

Analysis of trajectories of air masses indicated the possibility that westerly becomes extremely weak in the lower troposphere in summer over the eastern-north Asian regions but not in the upper

troposphere [16]. Iwasaka et al. [18] showed, on the basis of field observations including lidar and balloon-borne measurements at Dunhuang ($40^{\circ}00'N$, $94^{\circ}30'E$), China, wind system and geographical feature of Taklamakan desert areas as important factor controlling weak KOSA events. However, very few observations have been made concerning with free tropospheric KOSA particles over particle source areas and integration of observations is desired.

We made electron-microscopic experiments of the particles collected in the free troposphere over Dunhuang ($40^{\circ}00'N$, $94^{\circ}30'E$), China in March, 2003 (spring) following the balloon-borne measurements made in August, 2002 (summer). Here particle mixing states, chemical compositions, wind systems controlling particle transport were discussed on the basis of comparison with those field measurements which were made in different season; summer and spring.

II. Method

Aerosol particles were directly collected with balloon-borne aerosol sampler in the free troposphere over Dunhuang, China ($40^{\circ}00'N$, $94^{\circ}30'E$), on March 24, 2003 and August 29, 2002. The Dunhuang city, about 1200 m above the sea level, is located in the east side of the Taklamakan desert which is one of important sources of Kosa particles. Many investigators believe that air-masses including mineral dust derived from Taklamakan desert are frequently passed the free troposphere over Dunhuang. However, the states of aerosol in the free troposphere are little information. Therefore, this observational site is considered to be effective place to investigate the feature of individual Kosa particles on the initial state of long-range transport.

TABLE I
Sampling and weather condition

a) 24 MAR, 2003

Time (hh:mm, GMT)	Altitude (km) (ASL)	Ascending speed (m/min)	Sampling Time	Wind		Weather
				Direction	Speed (m/s)	
01:43-01:52	< 3km	237	—	WSW	6.5	
01:52-02:00	3~5km	286	7.75min	W	11.6	
02:00-02:07	5~7km	286	7.75min	WNW-W	16.8	Fine
02:07-02:13	7~9km	318	6.5min	WNW	18.6	

b) 29 AUG, 2002

Time (hh:mm, GMT)	Altitude (km) (ASL)	Ascending speed (m/min)	Sampling Time	Wind		Weather
				Direction	Speed (m/s)	
03:31-03:31	< 3.2km	290	—	ENE	6.6	
03:31-03:37	3.2~5.2km	332	6min	NNE-WNW	5	
03:37-03:42	5.2~6.9km	320	5.25min	NW	9.2	Fine
03:42-03:45	6.9~8.0km	374	3min	NNW-WNW	14.3	

Table 1 shows data of balloon-borne sampling and weather conditions during the measurement. We made it possible to collect particles in the three different layers of 3-5km, 5-7km, and 7-9km above sea level, using balloon-borne sampler containing three low-volume impactors (LVI) each with two stages. Aerosol particles were collected, to make single particle observation with an electron microscope, on carbon-coated nitrocellulose (collodion) films supported by Ni grids set on each stages of two-stage LVI. The jet diameters of the first and second stages of the impactor are 1.3 mm and 0.4 mm, respectively. The efficiency of 50% aerodynamical cut-off diameter under the flow rate of 1.8l min⁻¹ and standard atmospheric conditions (1013 hPa, 20°C) are evaluated to be 1.36 μm and 0.10 μm, respectively.

We individually examined the shape, size, and elemental composition of the particles with coarse mode ($d>1\mu\text{m}$) which is recognized as typically size of Kosa particles, collected in the first stage of impactor considering that most of Kosa particles were in the coarse mode range. The scanning electron microscope (SEM; Hitachi, S-3000N) and energy dispersive X-ray analyzer (EDX; Horiba, EMAX-500) were used here. The X-ray spectrum of a particle was obtained through a ultra thin window (UTW) detector with a counting time of 50 seconds and was generated from a square covering the particle which was irradiated the electron beam at a 20 kV accelerating voltage. It should be noted, although the EDX is able to quantitatively detect the relative weight and atom ratios of elements ($Z \geq 5$) in a single particle, that the current method is incapable of calculating relative weight ratios of carbon, nitrogen, oxygen and nickel owing to the inclusion of film or grid. The diameter of a particle was evaluated by the arithmetical mean of long width and the orthogonal width.

The balloon-train is schematically shown in Fig. 2. The balloon-borne sampler used here was already specified by Iwasaka et al. [18], and only the sampling operation system was changed to the system which is automatically controlled by signals from the Global Positioning System (GPS) data. On-off of valves of air flow system was operated from the command of pressure sensor on the basis of GPS signals. Distance between balloon launching and landing position of the sampler was about 48km for the measurement of March 24, 2003 (Fig. 3 (a)). Sampler landed on the desert area was able to be immediately recovered about two hours after sampling. Therefore, it made possible to avoid effect of contamination from surrounding air. After samples were recovered, each grid was kept in a plastic capsule, which was then sealed in a plastic bag together with paper-packaged silica gel. Electron microscopic experiment of those particulate materials was performed in Japan (Nagoya University).

III. Results

As shown in Table 1 and Fig. 2, the wind, during the measurement of spring (March 24, 2003), blown mainly from west, and was stronger, especially in the free troposphere, as compared with summer case (August 29, 2002). The visibility was very long and the weather clear. and its condition was similar to the case of summer observation. The ascending rate of balloon was about 300 m min⁻¹.

The graph in Fig. 4 illustrates the relative weight ratios on elements detected from individual particles. The horizontal axis corresponds to each particle: total number of coarse mode ($d>1\mu\text{m}$) particles collected in 3-5km was 298 and that in 5-7km was 48. As described below, the mixing state of particles collected in both 3-5km and 5-7km heights were very similar. Main particles in both layers were clay mineral such as aluminosilicate which mostly contain Si and Al, Mg, K or Fe in a single particle. Appearance ratio of particles which Si detected was 92.5% in 3-5km and 93% in 5-7km of all analyzed particles. Si-dominant particles, containing Si more than 80% in weight ratio of all detected element from analyzed single particle, were account for 13.7% in 3-5km and 12.5% in 5-7km of total Si-containing particles. Moreover, abundance ratio of particles with Si of between 30% and 80% by relative weight of detected elements indicates 65.8% in 3-5km and 70% in 5-7km.

Combinations of chemical elements of the particles collected during the balloon-borne measurements appear in Table 2. Combinations of particles are given on the basis of the quantitative results of EDX analysis by following relation according to Okada and Kai [19];

Weight ratio of element X, P(X)

$$= X/(Na + Mg + Al + Si + S + Cl + K + Ca + Ti + Mn + Fe)$$

The “X-rich” particle in Table 2 is defined as threshold values of $P(X) > 65\%$. Major component of mineral particles was Si-rich particles in the spring. Si-rich and Ca-rich particles were, respectively, about 70% and 20% of total coarse particles collected in both 3–5 km and 5–7 km heights (Table 2). It was suggested that the type of particles existed in free troposphere over source area of Kosa was mostly mineral particles. On the other hand, the type of particles in summer in 3–5km heights was also mainly mineral particles. However, it should be emphasized that the compositions of individual mineral particles considerably vary.

Figure 5 shows typical examples of SEM images and X-ray spectra of Si-rich and Ca-rich particles collected in 3–7km in spring (March 24, 2003). Those had irregular shapes which were frequently observed for mineral particles. X-ray spectrum of Fig. 5(a) shows the peak of Si, Al and K, suggested to be mica, and that of Fig. 5(b) shows the peak of Ca and the weak peak of Si and Al, which is suggested to be calcite ($CaCO_3$) with just a small amount of aluminosilicate. In the case of the spring, the abundance ratios of Ca-rich and Si-rich particles were mostly the same, which was 36% and 38%, respectively. It is worth to note that particles containing Na were found in the layer of 3–5km in summer observation [18], although such particles were not generally found on the ground air [20]. Here, Ca-rich and Na-rich particles which were often found in the summer are also shown in Fig. 6. Figure 6(c) shows a Ca-rich particle, which is suggested to be calcite ($CaCO_3$), but weak peak of sulfur was also detected from particle. It is not clear whether its peak was derived from mineral origin such as gypsum ($CaSO_4 \cdot H_2O$) or from anthropogenic origin. Na-rich particle shown in Fig. 6(d) seems to be thenardite (Na_2SO_4). The existence of these particles, i.e. Ca-rich particle and Na-rich particle, were supported that saline soils in saline lands around the desert areas in Xinjian, China contain significant amount of sulfate salt [21]. The shape of Ca-rich particles was different from the spring. In summer case, Ca-rich particles with round shape were frequently found, and it is thought that changes of number of those Ca-rich particles contribute to the abundance ratio of Ca-rich particles when few dust storms occur. However the sources of Ca-rich particles with round shape were still unknown.

Figure 7 shows the ratios of number concentration of Ca-rich particles to that of Si-rich particles. The value of [number concentration of Ca-rich particles]/[number concentration of Si-rich particles] in the range of 3–5km and 5–7km altitude in spring is considerably similar (Fig. 7(a)). The ratio is much higher, concerning with the distribution in 3–5km altitude in summer, than that in spring (Fig. 7(b)). Comparison values in 3–5km and those in 5–7km showed also similar feature in spring.

Although the number of coarse particles collected in the 1st stage was clearly decreased with increasing of altitude, mineral particles were dominant in coarse particles at any height. Figure 8 shows vertical distributions of particle number concentration which is sized at diameter $> 0.3\mu m$, $0.5\mu m$, $0.8\mu m$, $1.2\mu m$, and $3.6\mu m$, on the basis of measurements with a balloon-borne Optical Particle Counter (OPC; SIGMA TEC) made at Dunhuang corresponding to the particle collection with this balloon-borne impactor, in spring and summer seasons. It is also clear from the OPC observation that the particle number concentration decreases with increasing of altitude. The peak of the particle number concentration around 8km altitude in 17 August, 2001 was found. It is suggested that the high concentration of particle is due to thin cloud layer. By comparison between the OPC data in spring and winter above 3km altitude, the particle number concentration of larger than $1.2\mu m$ in spring were much higher than that in summer.

TABLE II
Collected particles larger than $1.0\mu\text{m}$ in diameter

Type of Particles	2003/3/24		2003/3/24		2002/8/29	
	3-5km number	%	5-7km number	%	3-5km number	%
Al-rich	0		0		0	
Si-rich	187	73.6	32	74.4	16	35.6
Si-dominant	58		9		10	
Si+Al	81		15		3	
Si+Mg	4		2		0	
Si+Fe(+Al)	25		3		3	
others	19		3		0	
Ca-rich	51	20.1	9	20.9	17	37.8
Ca-dominant	31		6		14	
Ca+Si	3		1		0	
Ca+Mg	7		1		1	
Ca+S	6		0		0	
others	4		1		1	
Fe-rich	5	2.0	0		0	
Fe-dominant	3		0		0	
others	2		0		0	
Ti-rich	2	0.8	0	0.0	0	
Ti-dominant	2		0		0	
Na-rich	6	2.4	1	2.3	5	11.1
Na-dominant	0		0		2	
Na+Cl	3		1		0	
Na+S	2		0		1	
others	1		0		2	
Cl-rich	1	0.4	0		0	
Cl+Na	1		0		0	
S-rich	1	0.4	1	2.3	5	11.1
S-dominant	1		1		5	
Others	1	0.4	0	0.0	2	4.4
Total	254	100	43	100	45	100
Ca-rich/Si-rich	0.27		0.28		1.06	
Si-rich+Ca-rich, %	93.7		95.3		73.3	

IV. Discussion

A. Atmospheric condition and mixing state of particles

Figure 9 shows potential temperature distributions in spring (April 30, 2002 and March 24, 2003) and in summer (August 17, 2001 and August 27, 2002) in Dunhuang, which were obtained with radio sonde (VAISALA, RS80-18E) during the measurements with the balloon-borne OPC. It is, from the gradient of potential temperature, suggested that the height of top of mixing layers in spring and summer were about 2.5-3km and 4km altitude, respectively (Fig. 9).

Vertical distributions of particle number concentration with $0.3\text{-}0.8\mu\text{m}$ (fine particle) and those of particles with $D > 1.2\mu\text{m}$ (coarse particle) are compared in Fig. 10, in which curves are averaged by running mean of about 300m interval. It was suggested that the cloud like structure with high relative humidity and high aerosol concentration layer existed at around 7.7km altitude in August 17, 2001 and at around 2.3km altitude in April 30, 2002 (Figs. 8 and 10). Concentrations of coarse particles in the boundary mixing layer in spring were several times higher than that in summer. Similarly, concentrations of coarse particles in the free troposphere in spring were one order of magnitude higher than that in summer (on August 17, 2001). Although the concentration of coarse particles between 4km and 6km altitudes in August 27, 2002 was as high as that in spring cases, this high concentration was suggested to be the dust layer by lidar measurement [18]. The free troposphere in spring over the Taklamakan Desert is dustier than that in summer owing to high concentration of dust particles. The high concentrations of dust in the free troposphere in spring is also supported from the fact that frequencies of dust outbreaks in the Taklamakan desert in spring, on the basis of routine surface meteorological reports, are higher than that in summer [22].

Figure 11 shows isentropic backward trajectories of air-masses for three days. The calculation was performed with the software provided by NOAA (HYSPLIT transport and dispersion model) to see the history of air-masses on March 24, 2003 and August 29, 2002. The air-masses of 3-5km and 5-7km in March 24, 2003 came through the free troposphere over the Taklamakan desert, and reached over Dunhuang. On the other hand, the air-mass of 3-5km in August 29, 2002 were transported through north part of the Tianshang Mountains. This difference certainly causes interesting difference in mixing state of particles, as described in detail later.

Figure 12 shows the wind vectors at 850 hPa at 1200 (GMT) on March 23 and at 0000 (GMT) on March 24, 2003. It is indicated that the wind of the north side of the Mts. Tianshan and the south side of the Mts. Altai just entered the Tarim Basin as inhaled. The winds entered the Tarim Basin converged around the south-west part of the Tarim Basin. This agreed with the wind system in the Tarim Basin which was reported on the basis of the moving directions of sand dunes in the Taklamakan Desert by Sun et al. [23]. In contrast, the Gobi Desert were mainly affected by westerly or north-westerly winds, and it is suggested, from the wind vectors at 850 hPa, that the air-masses originated from Gobi Desert were directly transported to easterly direction through lower altitude. The place of Dunhuang was located on just turning point of the wind direction in boundary layer. The wind mostly corresponded with the pressure system over the Tibet Plateau except around the Mts. Kunlun and the Mts. Qilian. In this region the wind is prevented by the high mountains, so that the wind direction was changed into westerly.

Figure 13 shows the upper-level chart of 500 hPa at 1200 (GMT) on March 23, 2003. It is suggested, from this chart, that the prevailing winds were westerlies at 500 hPa over the Tarim Basin and Dunhuang area, although the wind over these regions were mainly easterly or north-east winds at surface of 850 hPa. Owing to the geographical condition that the north, west, and south of the Taklamakan Desert are surrounded by high mountains (average elevation $> 5000\text{m}$), dust at an elevation lower than 5000m cannot easily move out of this desert [23]. The balloon-borne observation on March 24, 2003 indicated also effect of westerlies in the free troposphere, although the easterly wind prevailed in the lower altitude. It is likely that dust particles lifted to higher than 5000m were transported the long-range distance, and we can recognize the dust collected in the free troposphere over Dunhuang as the particles without chemical-physical transformations caused on dust particle surface during the long-range transport in the atmosphere.

As mentioned in results, the similarity of chemical compositions of dust particles in the layers of 3-5km and 5-7km in spring, which were mainly composed of Si-rich particles as shown in Fig. 4, was found. It is suggested that the dust lifted to the free troposphere in the source area were well mixed, although some very stable layers were sometimes found in the free troposphere over observational site, for example around 4km, 5.5km, 6.8km altitude in March 24, 2003. In other words, once the dust lifted to the free troposphere, even if stable layers appear after that the effect of the appearance of stable layer can be hardly detected in the dust particle distribution, the mixed state remains.

The mixing state of dust particles in the free troposphere in summer was significantly different from that in spring, and the appearance ratio of Ca-rich particles in summer was considerably high (Fig. 7 and Table 2). One possible explanation is the difference in the history of the air-masses; the air-mass was transported through the north side of the Tianshang Mountains in summer but not in spring (Fig. 11). Yabuki et al. [24] has reported that air borne particles collected in the Tianshan Mountain contain considerable amount of evaporate. Another possibility is that concentration of dust particles in summer is relatively low, comparing with the spring cases (Figs. 8 and 10). Figure 14 shows the diagram to compare values of [number concentration of Ca-rich particles]/[number concentration of Si-rich particles] of particles collected in the surface atmosphere in spring (on April 29, 2002; March 25, 2003) and summer (on August 17; August 18, 2001; August 26, 2002) in Dunhuang (for details on April 29, 2002; August 17, 2001; August 18, 2002, Trochkin et al., 2003). Weather conditions were fine and calm during observations except for April 29, 2002 (dust storm). Interestingly, the ratio of Ca-rich/Si-rich particles in summer was also much higher (0.72) than that in spring (0.35). Ca-rich particles had round shape as shown in Fig. 6 (c) when the relative appearance ratio was high. It is possible to suggest, although the origin of Ca-rich particles with round shape is hardly possible to be identified only from, that the ratio of Ca-rich particles relatively increase when the concentration of coarse particles, i.e. dust particle, is low such as summer cases. It is likely that situation of the ground surface and strength of vertical mixing are potential factors causing the difference of chemical mixing state.

B. Diffusion of dust existed in surface atmosphere

Figure 15 shows appearance frequencies on Si-, Ca-, Na- and Fe-rich particles of three cases; i) the results of particles collected in 3-5km altitude over Dunhuang (on March 24, 2003), ii) those near ground surface during dust-storm periods in Dunhuang (on April 29, 2002) [20], and iii) those near ground surface during strong winds or dust-storm in Zhangye (39°52'N, 100°23'E) (on March 13, 1990; April 27, 1991; May 7, 1991) [19]. It should be note here that the value of Zhangye measurements is based on the analysis of the particles with size of 0.1 μm -6 μm [19].

Interestingly, the appearance frequencies of these rich particles in the free troposphere and near surface atmosphere during dust-storm period in Dunhuang were mostly the same with particles collected during dust-storm or strong winds, although the Zhangye station is located in about 500km east from observational station in Dunhuang. The appearance ratios of Si-rich particles and Ca-rich particles collected in the free troposphere in spring or near ground under stormy condition were significantly different from the ratios in very calm and fine weather in summer. The types of detected particles in the free troposphere in spring were significantly similar to the values obtained in Dunhuang and Zhangye during dust-storm period. This is highly suggestive because particles collected over Dunhuang can be considered to show typical mixing state of mineral particles during dust-storm period in source regions.

Relative weight ratios of Al, S, and Ca for the mineral particles are shown in Fig. 16. The dashed line shows the weight ratio of S/Ca for gypsum (=0.8) in scattering diagram. If there are many plots on right of the dashed line, it can be assumed that little uptake of SO_x on mineral particles is occurred. Most of dust particles over Dunhuang in spring and summer, from relative weight ratios of S to Ca in Fig. 16, indicated the value of lower than 0.8. The ratios of S to Ca for dust particles collected in the surface atmosphere in Dunhuang also indicated the value of lower than 0.8 [20]. Moreover, dust particles collected in the surface atmosphere in Zhangye indicated the similar result [19].

The fact that uptake of SO_x on the dust particles were hardly occurred over the source area is of considerable effective to discuss the modification during long-range transportation. Trochkine et al. [20] has pointed out, from comparison with dust particles collected over Japan and near the ground surface, that approximately 40-45% of mineral particles mixed internally with sulfate during their transport in the troposphere. The results obtained here can confirm that dust particles are considerably modified during transportation in the free troposphere over urban areas in China, Korea, and Japan.

V. Conclusions

Dust particles collected in the spring free troposphere over the Taklamakan Desert with balloon-borne impactor suggest followings:

(1) Electron microscopic experiments directly showed that mineral dust particles were major constituents of coarse mode particles in spring and summer in the free troposphere over the Taklamakan Desert. This suggests that in the free troposphere over the Taklamakan Desert dust particles constantly existed, although the number concentrations of dust particles was higher in spring than that in summer.

(2) Analysis of wind systems showed that combination of predominating westerly in the free troposphere and surface wind strongly controlled by geographical structure of the Tarim basin was important process characterizing transport processes of KOSA particles originating in the Taklamakan desert. Dunhuang was located at just turning point of the wind direction in relatively low altitude such as in the boundary layer.

(3) The values of [number concentration of Ca-rich particles]/[number concentration of Si-rich particles] in the layers of 3-5km and 5-7km altitude in spring is considerably similar. It is likely that once the dust lifted to the free troposphere, even if stable layers appear after that the effect of the appearance of stable layer can be hardly detected in the dust particle distribution, the mixed state remains.

(4) The mixing state of dust particles, especially the appearance ratio of Ca-rich particles, in the free troposphere were of significantly difference between spring and summer, although Si-rich particles and Ca-rich particles were typical types of those dust particles. On the other hand, the ratio of Ca-rich /Si-rich particles of dust collected near the ground surface in Dunhuang and Zhangye during dusty period were very similar to free tropospheric dust. It is likely that the ratio of Ca-rich particles relatively increase when the concentration of dust is low, and situation of the ground surface and strength of vertical mixing are potential factors causing the difference.

The results obtained in this study provide very useful information to discuss the modification of Asian dust-storm particles during long-range transportation, the transport system of KOSA particle, and the radiative effects. For further understandings of the spatial and seasonal distribution of dust particles and chemical composition on individual those dust particles originated from the Taklamakan Desert, field measurements on individual particle composition over the desert and wind systems of the free troposphere in the north-west China are desired.

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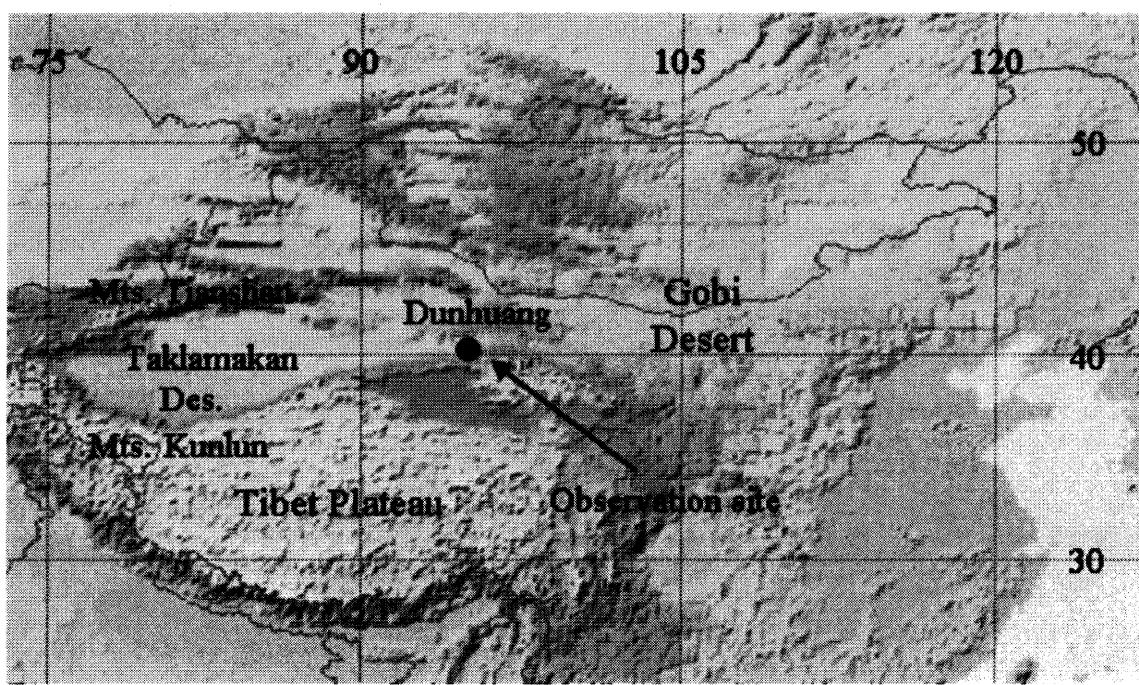


Figure 1. Observation site, Dunhuang, China (45°00'N, 94°30'E).

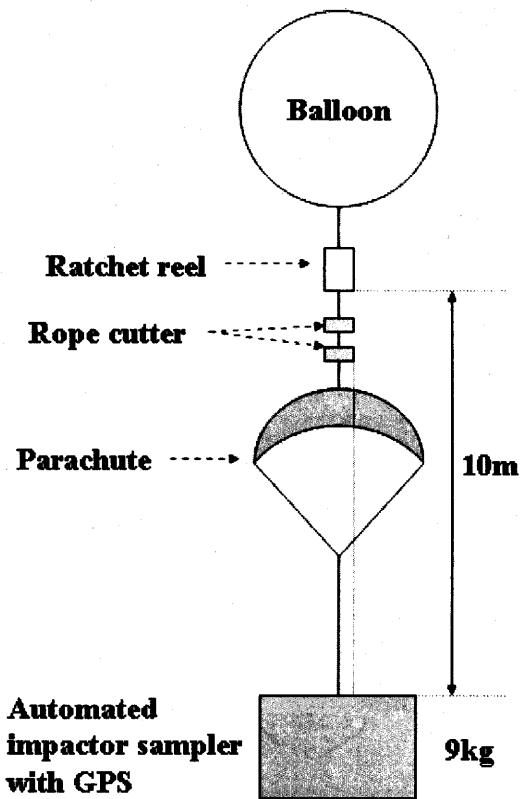


Figure 2. Balloon train used for aerosol collection; March 24, 2003 and August 29, 2002, Dunhuang China.

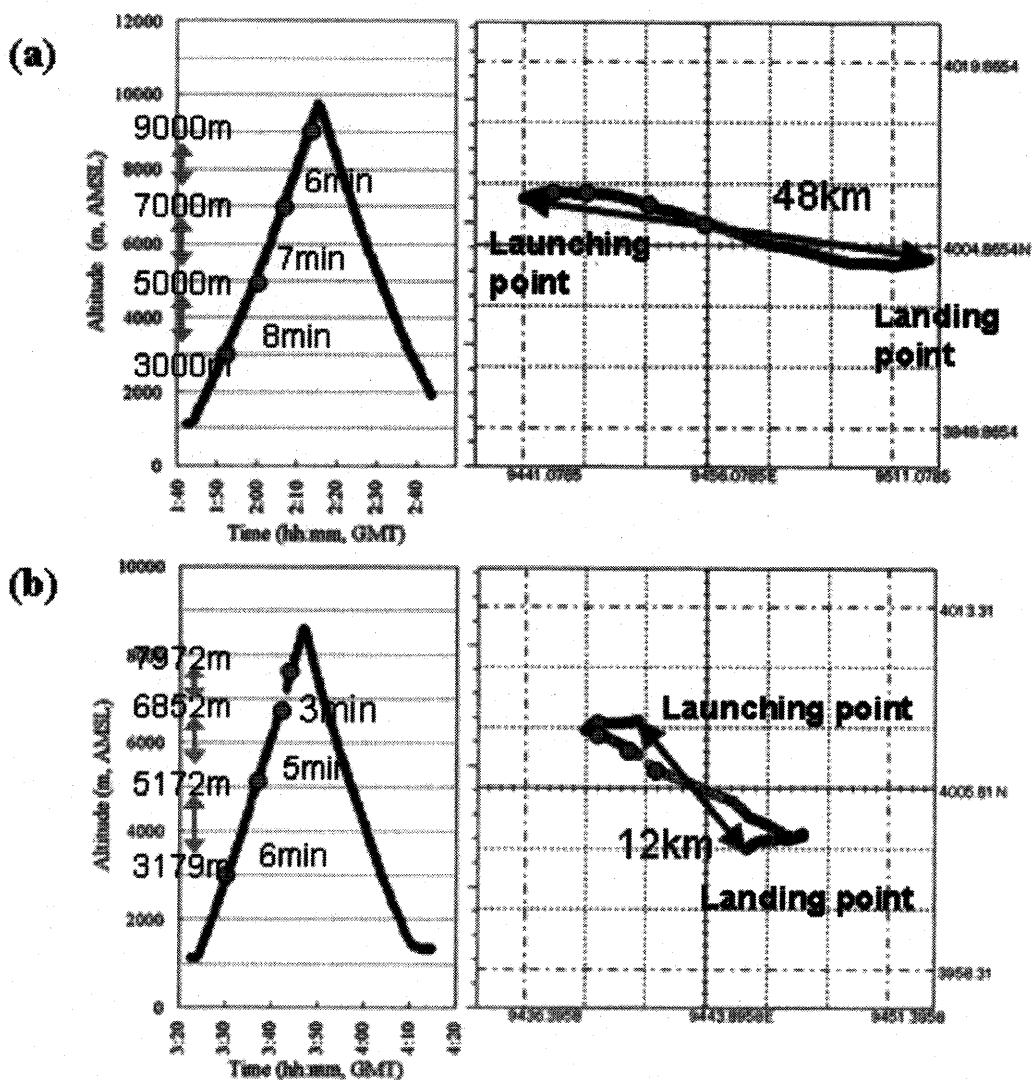


Figure 3. Tracks of the balloon-borne sampler deduced by Global Positioning System (GPS) data. (a) March 24, 2003, and (b) August 29, 2002.

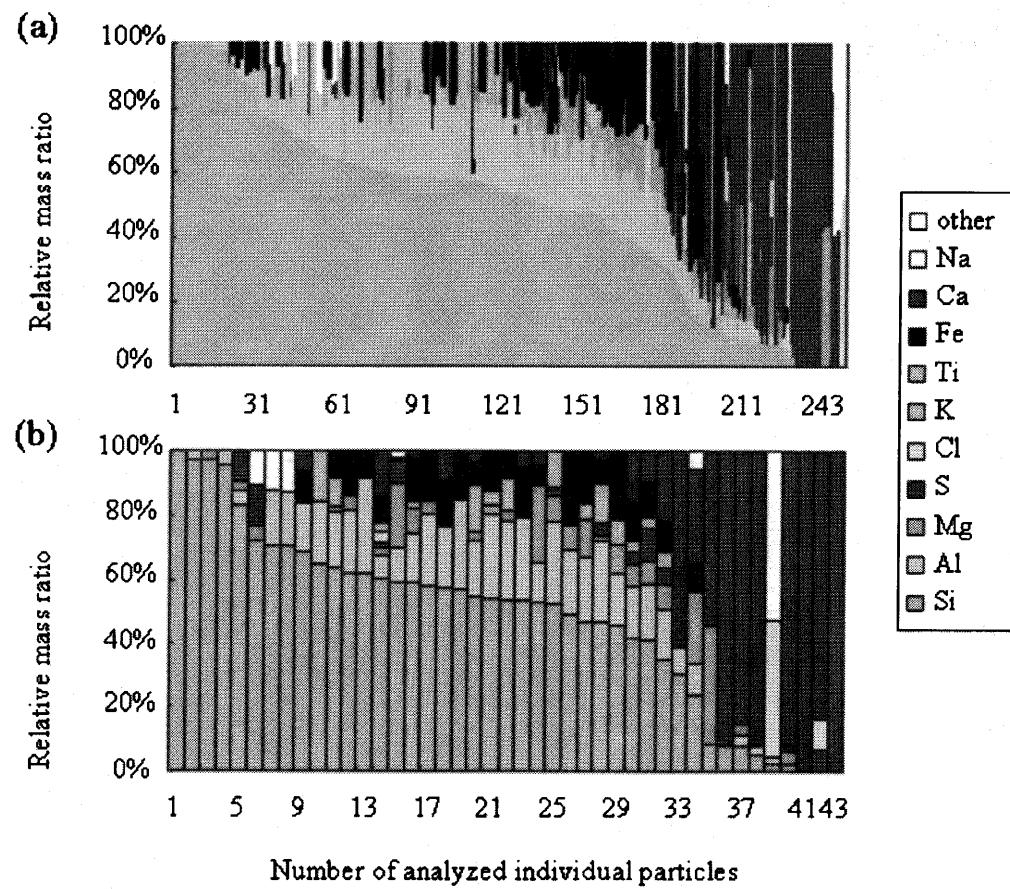


Figure 4. Relative mass ratio on the basis of single particle analysis. (a) March 24, 2003, and (b) August 29, 2002.

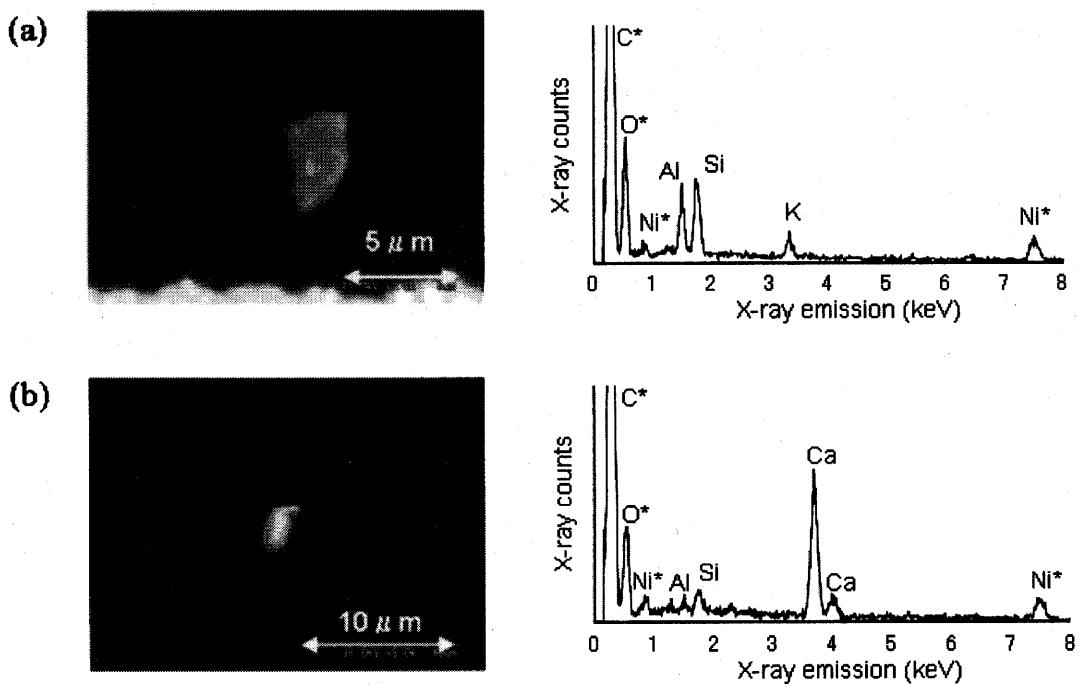


Figure 5. Typical example of electron micrographs and X-ray spectra of particles collected in 3-5km in spring (March 24, 2003). Particle (a) and (b) are Si-rich particle and Ca-rich particle, respectively. The Ni* peak is due to the Ni contained in grid used for the collection surface.

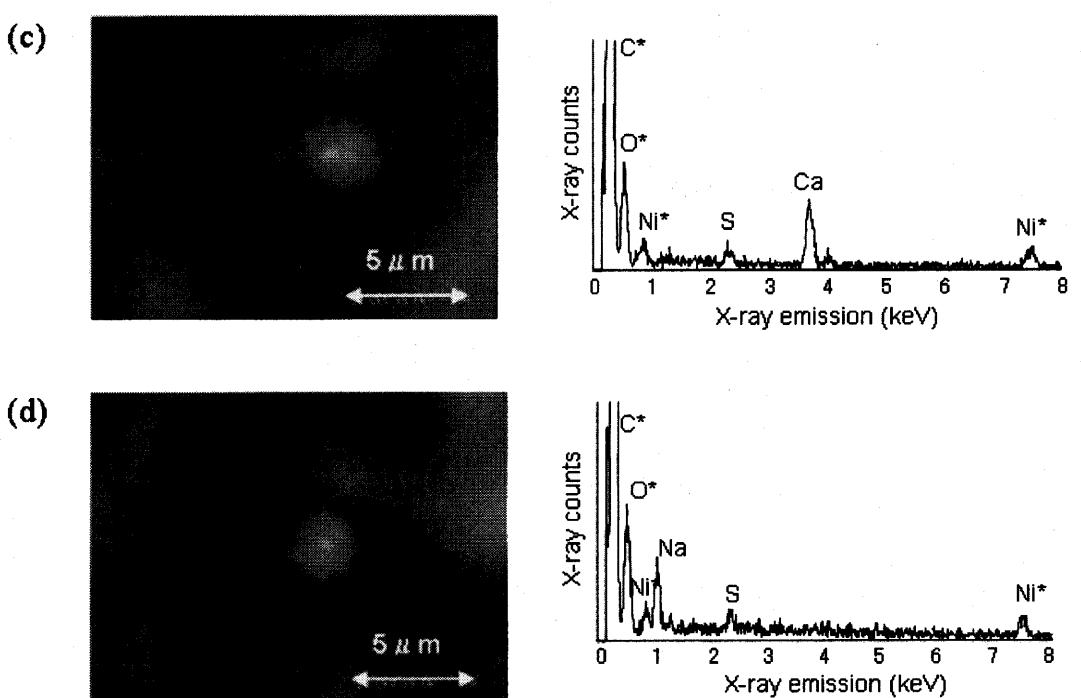


Figure 6. Typical example of electron micrographs and X-ray spectra of particles collected in 3-5km in summer (August 29, 2002). Particles (c) and (d) are Ca-rich particle and Na-rich particle, respectively. The Ni* peak is due to the Ni contained in grid used for the collection surface.

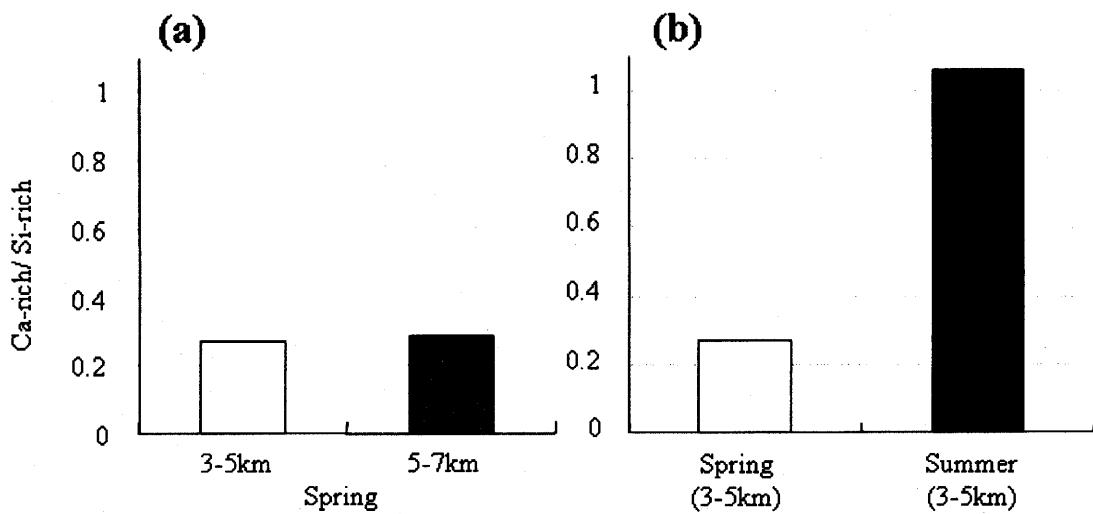


Figure 7. Comparisons of the value of [number concentration of Ca-rich particles]/[number concentration of Si-rich particles] between 3-5km and 5-7km in spring (a), and between spring time and summer time of the same layer of 3-5km (b).

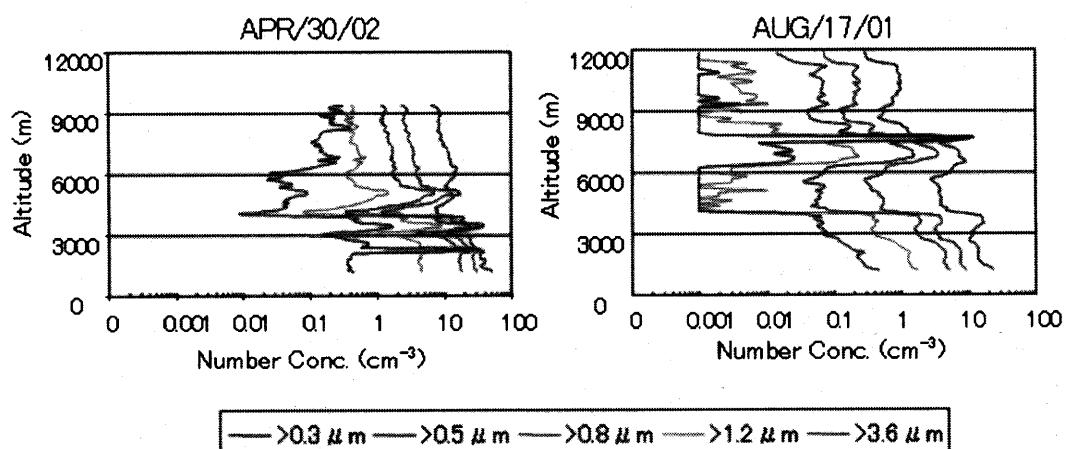


Figure 8. Aerosol number concentration and diameter observed with a balloon-borne optical particle counter in spring (April 30, 2002) and summer (August 17, 2001) at Dunhuang, China. Particle sizing was made at particle diameter = 0.3, 0.5, 0.8, 1.2, and $3.6 \mu\text{m}$.

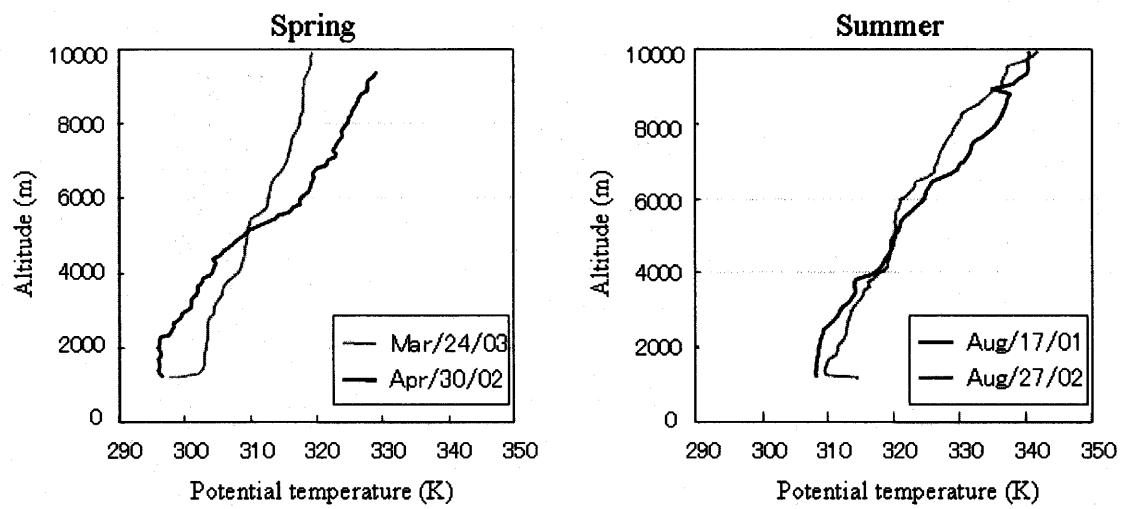


Figure 9. Potential temperature distributions in spring (April 30, 2002; March 24, 2003) and in summer (August 17, 2001; August 27, 2002).

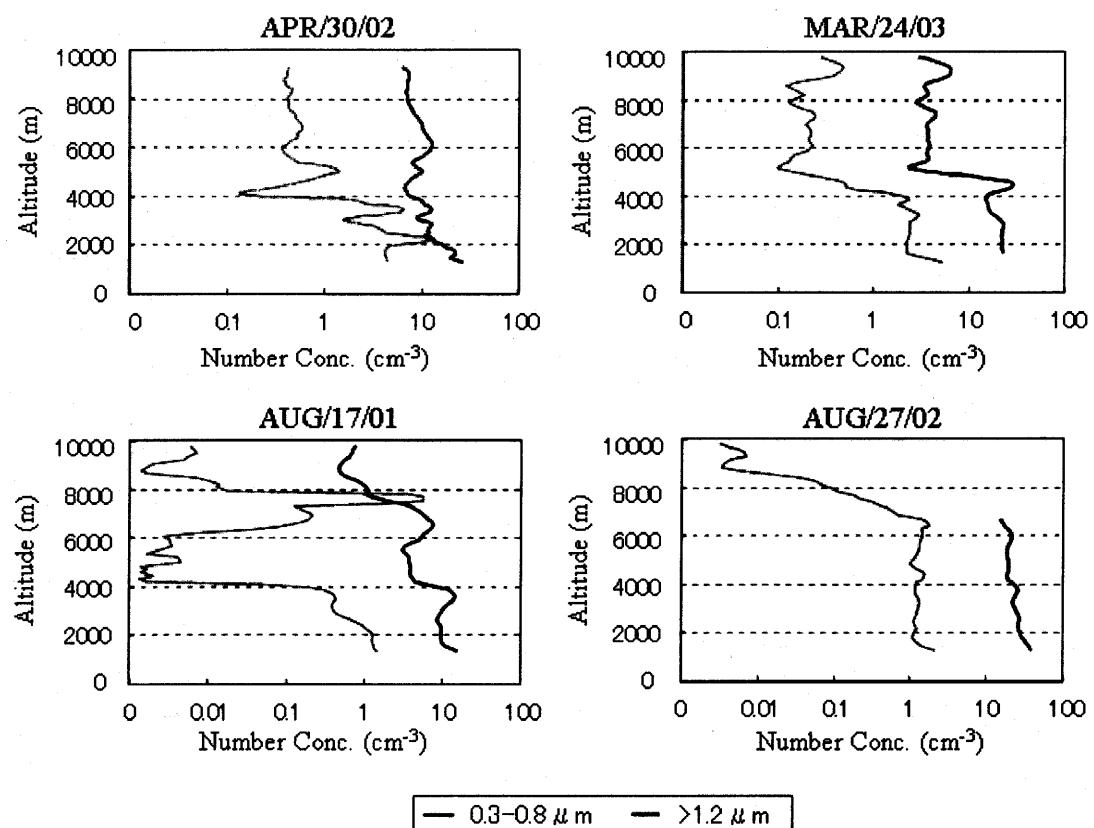


Figure 10. Vertical distributions of aerosol number concentration of coarse particle ($d>1\mu\text{m}$) and fine particle ($0.3<\text{d}<0.8\mu\text{m}$). The profiles are due to the about 300 m interval running mean.

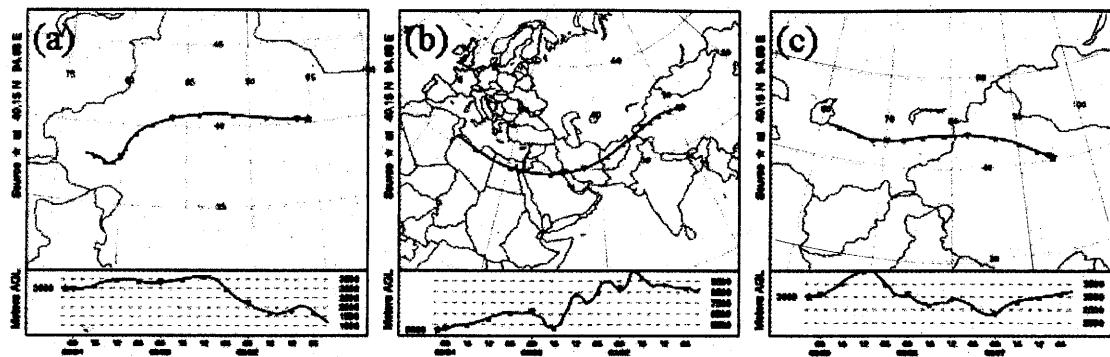


Figure 11. Three-day backward trajectories of air-masses which were observed at 3000m above the ground level (a) and 5000m above the ground level (b) on April 24, 2003, and at 3000m above the ground level on August 29, 2003 (c) at Dunhuang, China (on isentropic surface). These trajectories were analyzed with the HYSPLIT model provided from NOAA Air Resources Laboratory's web server.

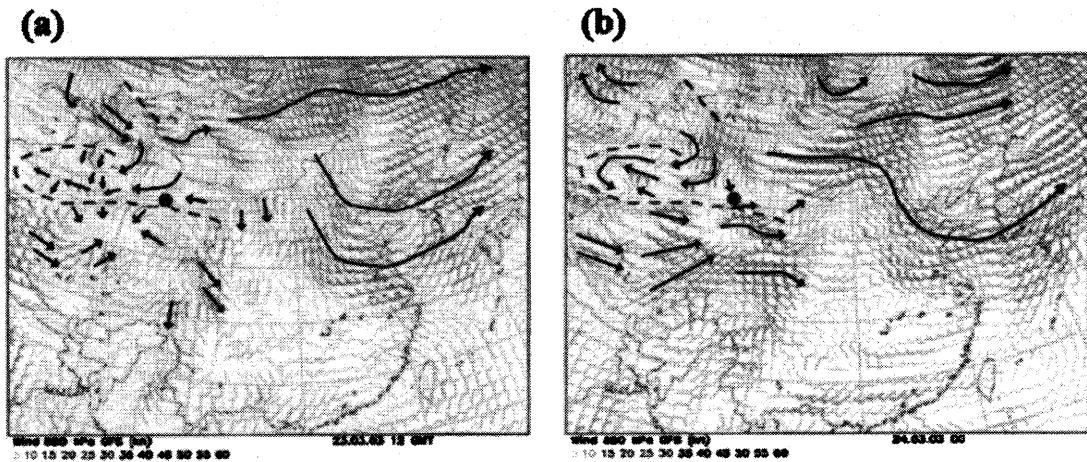


Figure 12. Wind vectors at 850 hPa at 1200 (GMT) on March 23 and at 0000 (GMT) on March 24, 2003. Arrows (\uparrow) show predominant wind, and the filled circle indicates the observational site, Dunhuang ($45^{\circ}00'N$, $94^{\circ}30'E$). Broken lines show mountain ranges around the Taklamakan Desert.

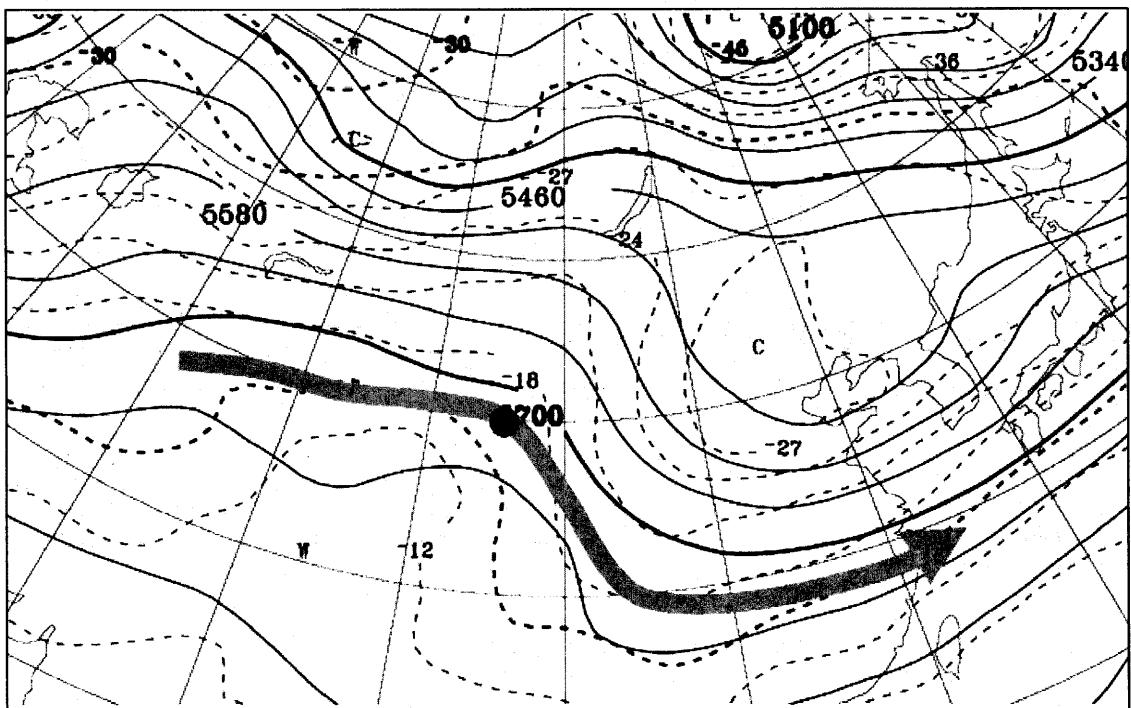


Figure 13. Wind on 500 hPa surface at 1200 (GMT) on March 23, 2003. An arrow (\uparrow) shows direction of predominant wind, and filled circle indicates the observational site, Dunhuang ($45^{\circ}00'N$, $94^{\circ}30'E$).

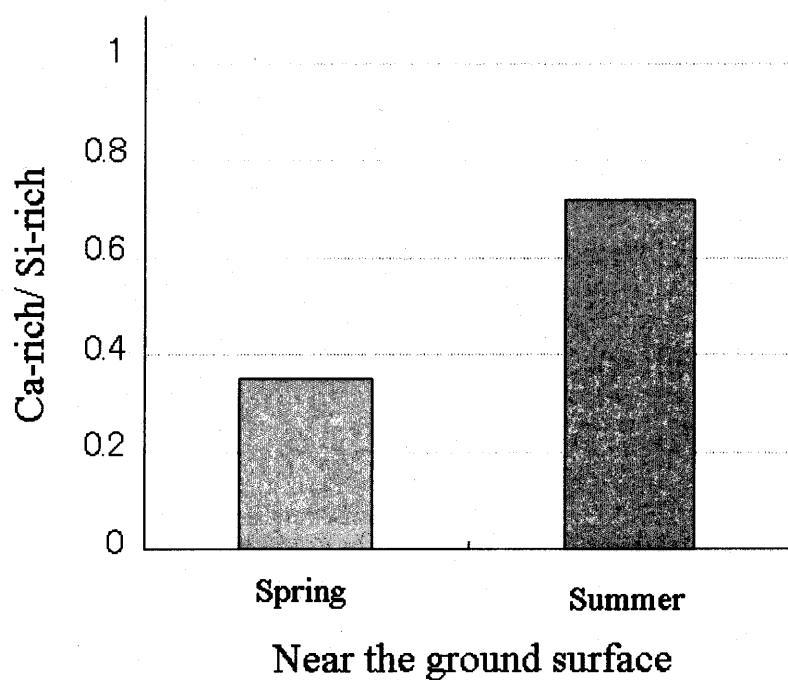


Figure 14. Comparison of the values of [number concentration of Ca-rich particles]/[number concentrations of Si-rich particles] collected in the surface atmosphere in spring and summer in Dunhuang. The data used in this graph are the results on the basis of the sampling in spring (on April 29, 2002; March 25, 2003) and summer (on August 17, 2001; August 18, 2001; August 26, 2002).

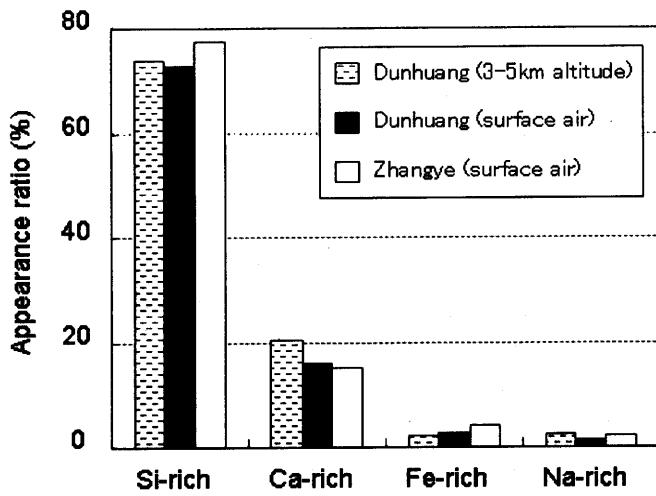


Figure 15. Appearance frequencies on Si-rich, Ca-rich, Na-rich, and Fe-rich particles of the three case; i) the results of particles collected in 3-5km altitude over Dunhuang (on March 24, 2003), ii) those near ground surface during dust-storm periods in Dunhuang (on April 29, 2002), and iii) those near ground surface strong wind or dust-storm in Zhangye (on March 13, 1990; April 27, 1991; May 7, 1991). The data of chemical composition of dust is from samples taken at Zhangye (Okada and Kai, 1995).

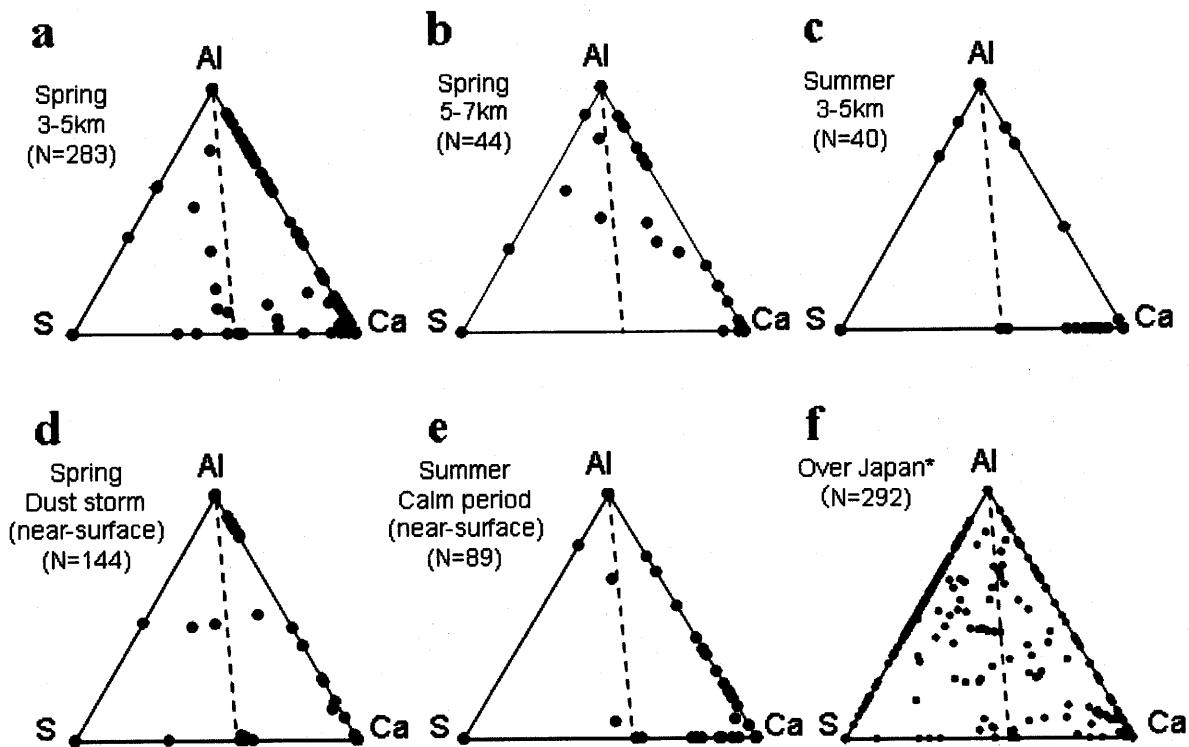


Figure 16. Relative weight ratios of Al, S and Ca for mineral particles. Those particles were collected in 3-5 km altitude over Dunhuang on March 24, 2003 (a), those in 5-7 km altitude over Dunhuang on March 24, 2003 (b), those in 3-5 km altitude over Dunhuang on August 29 (c), those in the surface atmosphere at Dunhuang on April 29, 2002 (d), those in the surface atmosphere at Dunhuang (e), and those in the free troposphere over Japan (f). The graph (f) is from Trochkin et al. (2003).