The composition of individual aerosol particle in the troposphere and stratosphere over Xianghe (39.45°N, 117.0°E), China

Li Xu[a],*, Kikuo Okada[b], Yasunobu Iwasaka[c], Keiichiro Hara[c], Yasuhiko Okuhara[c], Yukitomo Tsutsumi[b], Guangyu Shi[d]

[a]National Climate Center, No. 46, Baishiqiao Road, Beijing 100081, People's Republic of China
[b]Meteorological Research Institute, Tsukuba, Ibaraki 305-0035, Japan
[c]Solar-Terrestrial Environment Laboratory, Nagoya University, Nagoya 464-8601, Japan
[d]Institute of Atmospheric Physics, Chinese Academy of Science, Beijing 100029, People's Republic of China

Abstract

A balloon observation was carried out on 22 August in 1993 from Xianghe Scientific Balloon Base (39.75°N, 117.0°E) near Beijing in China. Individual aerosol particles in the five samples collected in the troposphere and lower stratosphere were analyzed by using a transmission electron microscope equipped with an energy-dispersive X-ray (EDX) analyzer. Types of particles were classified by the quantitative EDX analysis and particle morphology. Following results were obtained by the analyses of aerosol particles in the radius range of 0.1–0.5 μm: (1) Sulfate particles were dominant (80%) in aerosol particles collected between 4 and 6 km altitude. (2) Sulfuric acid particles were present in 74% of particles at ~ 8 km altitude, 91% at 11 km, 95% at 17 km and 88% at 21.2 km. (3) "S-rich" particles with K were collected both in the troposphere and lower stratosphere. It was considered that the particles containing K found at ~ 5, ~ 8 km altitude could originate from burning processes in the continent including the Tibetan plateau and be transported to the middle troposphere. (4) Sulfuric acid particles with Fe were present in 20–30% of sulfuric acid particles in the lower stratosphere. (5) Particles mainly composed of minerals were present in 6, 11% of particles at ~ 5, ~ 8 km, indicating the vertical transport to the upper troposphere. (6) Mineral particles which contain sulfuric acid and sulfate suggest the formation of sulfuric acid and sulfate on mineral particles by heterogeneous processes in the troposphere. (7) Sea-salt particles with and without minerals were collected in the troposphere and lower stratosphere, suggesting the vertical transport by convective clouds. © 2001 Elsevier Science Ltd. All rights reserved.

Keywords: Aerosol particle composition; Troposphere and stratosphere; Electron microscopy; EDX analysis

1. Introduction

Although aerosol particles occupy a tiny fraction in the atmosphere, they influence many atmospheric processes. The impact of aerosol particles on the radiation balance of the Earth's atmosphere is directly related through the scattering and absorption of solar and terrestrial radi-ations. Indirectly, aerosol particles participate in the formation of clouds which affects the radiation transmission and water cycle in the atmosphere (Charlson et al., 1987). Meanwhile, aerosol particles act as the surface for heterogeneous reactions in the atmosphere. As for the destruction of stratospheric ozone, heterogeneous reactions are much more effective than gas-phase reactions (WMO/UNEP, 1993). The properties of aerosol particles also serve as indicators of aircraft emission, volcanic eruption, air exchange and diffusion between the troposphere and stratosphere. To evaluate the effect of aerosol

particles on these processes, the physicochemical properties of aerosol particles need to be studied.

Balloon-borne observations of aerosol particles have also been carried out using an optical counter (e.g., Rosen, 1964; Hofmann et al., 1975). However, detailed information on the composition of stratospheric aerosol particles cannot be obtained by the observations mentioned above. Direct collection of particles by a balloon-borne sampler, which is useful and essential to study the composition of aerosol particles, has been carried out using a balloon (Junge et al., 1961; Bigg et al., 1970; Gras and Laby, 1978; Iwasaka et al., 1992; Sheridan et al., 1994; Wu et al., 1994; Okada et al., 1997; Xu et al., 1998). Since most of these studies were mainly concerned with stratospheric particles, information on the vertical difference in the composition of aerosol particles from the troposphere through the stratosphere is very meager.

The aim of this paper is to show the morphology, size and elemental composition of individual aerosol particles in the troposphere and stratosphere over China by an electron microscopy. A vertical profile of the number concentration of aerosols with radii larger than $0.2\,\mu m$ is also indicated in this paper.

2. Experimental and analytical descriptions

A balloon-borne impactor was used for the collection of individual aerosol particles in the troposphere and stratosphere. The individual particles were sampled on a carbon-coated nitrocellulose film supported on an electron microscope grid. The jet diameter of the impactor is 2 mm and the flow rate of air is $22\,l\,min^{-1}$. The efficiency of the pump used in the impactor was $100 \pm 4.6\%$ under the condition of atmospheric pressure from 1000 to 5 hPa (Tsuchiya et al., 1996). The collection efficiency of particles for particle density of $2.0\,g\,cm^{-3}$ is calculated using the results of Ranz and Wong (1952) by taking the effect of air pressure on the efficiency into consideration. The 50% cutoff radius of the impactor is evaluated to be $0.27\,\mu m$ on the ground, $0.19\,\mu m$ at 10 km altitude and $0.05\,\mu m$ at 20 km. A balloon-borne optical particle counter (OPC) was used to measure the vertical profile of number concentration of aerosols with radii larger than $0.2\,\mu m$ at altitudes from 2 to 34 km.

The balloon-borne impactor, OPC and other instruments in a gondola were launched from Xianghe Scientific Balloon Base ($39.45°N$, $117.0°E$), which is located at about 60 km southeastward from Beijing, on 22 August in 1993. It took about 90 min to arrive at 34 km height. A sample was collected from an air layer of about 1.5 km thickness in the balloon ascent. To avoid the contamination from particles near the ground, the air pumps for the impactor and the OPC began to work from 1.3 km altitude and 2 km, respectively.

The particles collected on the films were analyzed by using a transmission electron microscope (TEM) (Hitachi, H-600) equipped with an energy-dispersive X-ray (EDX) analyzer (Kevex, Sigma 2) to examine particle morphology, size and elemental composition. Before the TEM + EDX analysis, the individual particles on the film were coated with Pt/Pd alloy at an angle of arctan 0.5 to obtain a shadow of single particle and the micrographs were taken by the TEM. The size of the particle was calculated by the measured apparent diameter d and shadow length l on the micrographs. The particles were basically divided into two types for size evaluation, a spherical cap and a disk. The particle with l/d ratio less than 0.62 is assumed to be present as a spherical cap on the collection surface, and, the particle with l/d larger than 0.62 is regarded as a disk. The size of the particle with satellite droplet rings on the collection surface was calculated by the method proposed by Gras and Ayers (1979). In addition to the TEM image, the images of some particles were also taken by a scanning electron microscope (SEM) (Hitachi, S-2150) after the TEM + EDX analysis.

EDX spectrum was obtained by the irradiation of electron beams at the central part of a particle. An accelerating voltage was 50 kV and a counting time of 100 s for the examination. Quantitative analysis of elemental composition of individual particle was processed by using thin-foil method (Cliff and Lorimer, 1975). The quantitative analysis was carried out using Kevex QuantexTM software for energy-dispersive microanalysis. The weight percent of elements was calculated on the basis of characteristic X-ray fitted with a Gaussian distribution, not the peak intensity.

To evaluate the transport path of aerosol particles, the isentropic backward-trajectory calculations of air parcels were carried out by using the JMA (Japan Meteorological Agency) global analysis data (GANAL) on every day 00 and 12 GMT.

3. Results and discussion

3.1. Vertical profile of aerosol number concentration

The vertical distributions of aerosol number concentration (radius $(r) \geqslant 0.2\,\mu m$) and atmospheric temperature from 2 to 34 km altitude are shown in Fig. 1. The stratospheric aerosol layer is obvious and the peak number concentration is $2.2\,cm^{-3}$, which is larger than that in calm background period. According to Hofmann et al., 1975, Hofmann and Rosen, 1982, Deshler et al., 1992, measurements, the number density of the particles with $r \geqslant 0.15\,\mu m$ in the stratosphere during the calm period was in the range of 0.5–$1.5\,cm^{-3}$. The presence of high concentrations in the stratospheric aerosol layer suggests that the effect of aerosols formed by the oxidation of SO_2
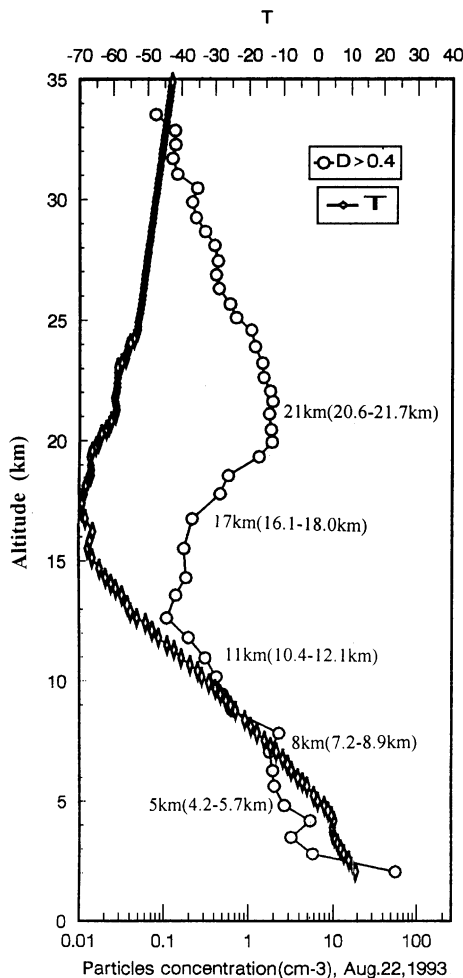
Fig. 1. The vertical distributions of aerosol number concentration ($r \geqslant 0.2\,\mu m$) and air temperature.

emitted from Mt. Pinatubo volcanic eruption in June 1991 still remained in the stratosphere in 1993 (e.g., Wu et al., 1994; Shi et al., 1997). The symbols of 5 km (collected at 4.2–5.7 km altitude), 8 km (7.2–8.9 km), 11 km (10.4–12.1 km), 17 km (16.1–18.0 km) and 21.2 km (20.6–21.7 km) in Fig. 1 indicate the samples used for the analyses in this paper. From the temperature profile, the tropopause was determined to be located at 15.6 km altitude.

3.2. Features of individual particles

The morphology and X-ray spectrum of typical particles collected in this balloon observation are displayed in Figs. 2a–e. The micrographs of Figs. 2b–e were taken by a SEM. There were many particles showing satellite droplet rings on the collection surface which are characteristic morphology of sulfuric acid (Frank and Lodge,

1967). Fig. 2a presented a typical particle with a large diameter of satellite droplet rings, which was collected at 7.2–8.9 km altitude. From the X-ray spectrum and the large diameter of satellite droplets $14.5\,\mu m$, it is determined to be mainly composed of sulfuric acid. Fig. 2b shows a mineral particle surrounded by liquid droplets collected at 10.4–12.1 km. The radius of mineral found as a central part was about $0.68\,\mu m$. Quantitative analysis of elements from the central part of the particle showed that the weight percent of elements was 95% Si and 5% S. Sulfur-containing particle without satellite droplet rings collected at 4.2–5.7 km altitude is displayed in Fig. 2c. From the morphology and X-ray spectrum, the particle is considered to be mainly composed of ammonium sulfate (Xu et al., 1998). Fig. 2d shows a mineral particle collected at 4.2–5.7 km altitude. The particle was mainly composed of the elements Si, Al, K and Fe, etc. These elements are the indicators of mineral particles. An interesting finding was the presence of a sea-salt particle containing crustal elements at 16.1–18.0 km altitude (Fig. 2e). From the information on the morphology and elemental composition of this particle, we can decide that this is a sea-salt particle mixed with minerals. Note that the morphology of particles (mainly $r > 1.0\,\mu m$) collected in the same balloon observation was examined by a scanning microscope and the results were shown in another paper (Iwasaka et al., 1997).

3.3. Types of particles

Quantitative analysis was made for the elements of Na, Mg, Al, Si, S, Cl, K, Ca, Ti, Cr, Mn, Fe, Ni and Pb in individual particles. Weight percent $P(X)$ of element X was obtained as follows:

$$P(X) = 100\% \times X/(Na + Mg + Al + Si + S + Cl + K$$
$$+ Ca + Ti + Cr + Mn + Fe + Ni + Pb).$$

In addition to the quantitative results obtained by the EDX from the central part of particle, particle morphology was also examined. Types of particles were evaluated by this combined information.

Firstly, the types of particles are classified by the quantitative EDX analysis. The particle which has the maximum weight percent of element X in analyzed elements with atomic numbers $\geqslant 11$ is termed "X-rich" particle. Secondly, the "X-rich" particles are classified further into various types using morphological appearance and elemental composition. As for "S-rich" particles, the particles with and without satellite droplet rings on the collecting surface were determined to be "sulfuric acid" and "sulfate", respectively (Gras and Ayers, 1979). As shown in Fig. 2b, mineral particles which showed satellite droplet rings are termed mineral with sulfuric acid. Sea-salt particles are determined by weight percent of Na > 10%, not by weight percent of Cl.

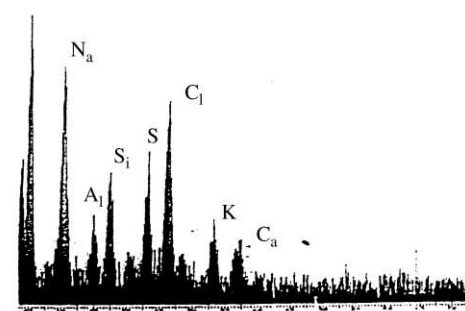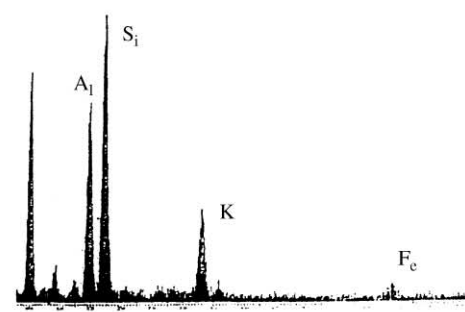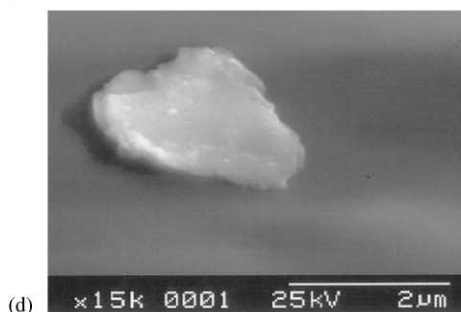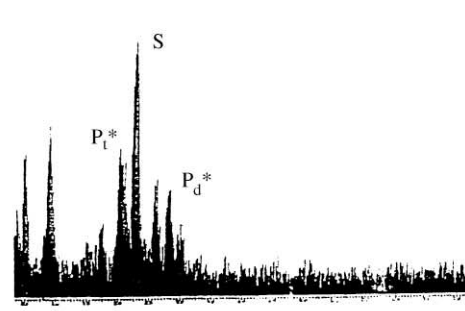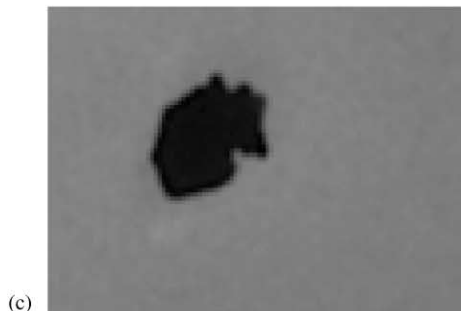Table 1
Types of aerosol particles in the radius range of 0.07–2 μm evaluated by morphology and EDX analysis[a]

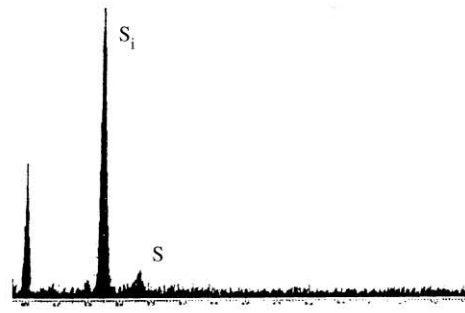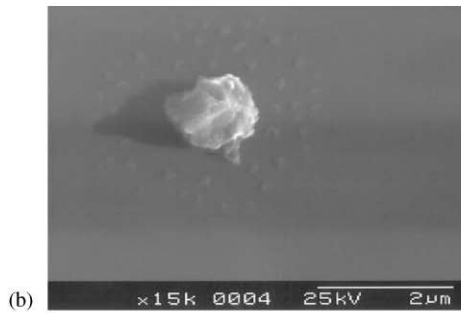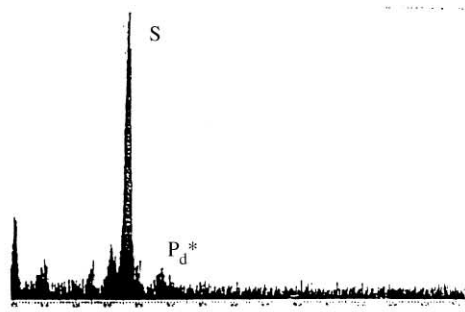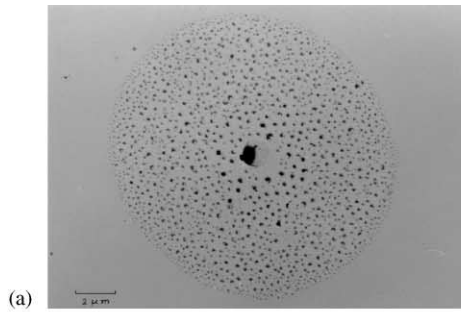| Type of particles S-rich	Sample 4.2–5.7 km			Sample 7.2–8.9 km			Sample 10.4–12.1 km		Sample 16.1–18.0 km			Sample 20.6–21.7 km		Sum of line
	S	M	L	S	M	L	M	L	S	M	L	S	M	
Sulfuric acid														
S-dominant (S > 90)		2	4	1	35	3	45	10		49		1	29	179
S + K (K > 10)				1						1				2
With minerals														
S + Al + Si (Al + Si > 10)			1										1	2
S + Fe (Fe > 10)							3			13			14	30
S + Mn (Mn > 10)													1	1
Sulfate														
S-dominant (S > 90)	8	37		5	5		2			1			3	61
S + K (K > 10)	1	2			2		1		1	1				8
With minerals														
S + Al + Si (Al + Si > 10)		4												4
S + Fe (Fe > 10)		1												1
K-rich														
K + S													1	1
K + Si					1								1	2
Mineral														
Fe-dominant (Fe > 90)							2							2
Al + Si + Fe (Al + Si > 50)		6	1											7
With sulfuric acid														
Al + Si + S (S > 20)			2		1			1						4
With Sulface														
Al + Si + S (S > 20)					2									2
Seasalt														
Seasalt (Na > 20)		1								1				2
With minerals														
Seasalt (Si, Fe > 10)		1									1			2
Soot		1												1
Cr-rich						1								1
Ni-rich													1	1
Pb-rich					1									1
Sum of column	9	55	8	7	47	4	S = 0, 53	11	1	66	1	1	51, L = 0	
Σ = S + M + L	72			58			64		68			52		314

[a]The total number of S is 18, M is 272, L is 24. S: Radius range of 0.07–0.1 μm. M: Radius range of 0.1–0.5 μm. L: Radius range of 0.5–2 μm. Weight percent of element (for example, S > 90: weight percent of S is more than 90%).

Particles made up of chain aggregations of electron-opaque spherules are considered to be soot (e.g., Ogren and Charlson, 1983).

◄

Fig. 2. Electron micrograph and X-ray spectrum from the central part of particle. (a) Sulfuric acid particle collected at 7.2–8.9 km altitude. (b) Particle mainly composed of minerals surrounded by satellite droplet rings (sulfuric acid) collected at 10.4–12.1 km altitude. (c) Sulfur-containing particle without satellite droplet rings collected at 4.2–5.7 km. (d) Mineral particle collected at 4.2–5.7 km altitude. (e) Sea-salt-particle-containing crustal elements collected at 16.1–18.0 km altitude.

The type of particles are listed in Table 1. Number of particles in each type was summarized in each sample with three radius ranges (S: 0.07–0.1 μm, M: 0.1–0.5 μm, L: 0.5–2 μm). Total number of particles in each sample is 72 (sample 5 km), 58 (8 km), 64 (11 km), 68 (17 km) and 52 (21.2 km), respectively. Since the number of particles examined is small in the radius ranges of 0.07–0.1 μm (size class S), no small particles at 11 km, and 0.5–2 μm (size class L), no large particles at 21.2 km, difference in particle types in the radius range of 0.1–0.5 μm (size class M) is mentioned here.

As evaluated from Table 1, "S-rich" particles were present in 84% (46/55 particles), 89% (42/47) in the

samples 5 and 8 km. In the samples 11, 17 and 21.2 km, the proportion of "S-rich" particles are found up to 94–98%. Then it can be said that most particles both in the troposphere (4–16 km) and lower stratosphere (16–22 km) were "S-rich" particles. Secondly, particles mainly composed of mineral ("Mineral" in Table 1) were collected in the troposphere (4–12 km) and the proportions are 11% in sample 5 km, 6% in sample 8 km and 3% in sample 11 km, respectively. Consequently, "S-rich" and mineral particles occupied approximately 97% of collected particles (M = 272). Other particles are "K-rich", sea-salt, soot, "Cr-rich", "Ni-rich" and "Pb-rich" particles. The number of soot, "Cr-rich", "Ni-rich" and "Pb-rich" particles collected is only one for each type.

3.4. Abundance of sulfate and sulfuric acid particles in "S-rich" particles

"S-rich" particles were divided into sulfate and sulfuric acid particles by judging the presence of satellite droplet rings on the collecting surface (Gras and Ayers, 1979). In the particles of 0.1–0.5 μm radius (Table 1), sulfate particles were present in 96% (44/46 particles) of "S-rich" particles and 80% (44/55) of total particles in the sample 5 km, although sulfuric acid particles ($r > 0.5$ μm) were found in that layer. Above 7 km altitude, the fraction of sulfate particles tended to decrease with increasing altitude. On the other hand, sulfuric acid particles were dominant in "S-rich" particles of 0.1–0.5 μm radius: that is, sulfuric acid particles were present in 83% of "S-rich" particles in sample 8 km, 94% at 11 km, 97% at 17 km and 94% at 21.2 km, respectively. The number faction of sulfuric acid particles in all particles of 0.1–0.5 μm radius was 74–91% in the middle and upper troposphere (8 and 11 km) and 88–95% in the lower stratosphere (17 and 21.2 km). The high number fraction of sulfuric acid particles in the stratosphere is nearly consistent with the results obtained over the United States (Sheridan et al., 1994).

Sulfate particles appeared mainly at 4–6 km altitude in this observation. This is consistent with the previous results, which showed the presence of ammonium sulfate particles in the lower and middle troposphere (Yamato and Ono, 1989; Xu et al., 1998). However, sulfate particles were collected even in the stratosphere although the number of the particles was very small.

3.5. Potassium in particles

Some "S-rich" particles contained potassium (K). In this analysis, "S + K" particles without minerals were used to reduce the effect of minerals on K content. "S-rich" particles with K ($P(K) > 10\%$) were indicated in "S + K" particles in Table 1. They consisted of 10 particles out of 314. In the samples collected at ~5 and

8 km, 5 out of 6 of the "S + K" particles were present as sulfate particles (see Table 1).

Backward trajectories were calculated to assess the paths of air mass for the sampling altitudes. Figs. 3a and b show the horizontal- and longitudinal-height projections of the isentropic backward trajectories of air parcels from 8 and 11 km altitudes over Beijing (12 GMT on 22 August 1993), respectively. The negative numbers indicated on the trajectories in the figure denote days prior to the sampling over Beijing. The falling speed for particles of 1 μm radius is evaluated to be approximately 0.55 m h^{-1} at 5 km altitude (Kasten, 1968). Since the falling speed is much smaller than the synoptic-scale vertical velocity (on the order of 100 m h^{-1}), particles with $r < 1$ μm could move with air parcels. Within 2 days before the sampling, the air parcel calculated from 8 km altitude over Beijing (solid line) was located at 7–8 km altitude over the regions to the east of the Tibetan
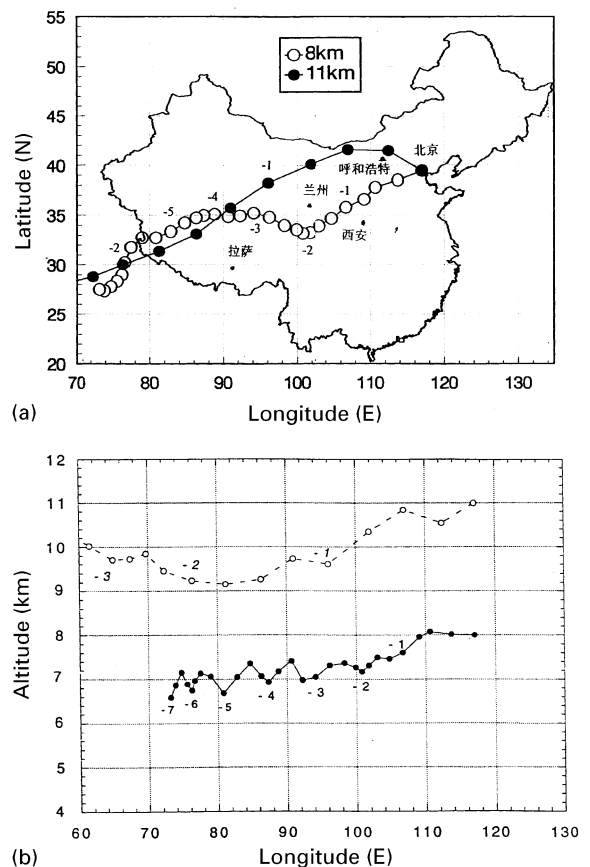

Fig. 3. Horizontal (a) and longitudinal height projections (b) of the isentropic backward trajectories of air parcels from 8 and 11 km altitudes over Beijing (12 GMT on 22 August 1993). Negative numbers indicated on the trajectories in the figure denote days prior to the sampling over Beijing.

plateau. Two to five days before, the air parcel was located just over the Tibetan plateau. On the other hand, the trajectory calculated from 11 km altitude over Beijing (broken line) shows that air parcel is located at 9–11 km within 2 days before sampling over the similar areas in the continent as the trajectory from 8 km. It means that air mass at 8, 11 km samples came from a height lower than the sampling height.

It is known that particles with K form originally by biomass burning (e.g., Andreae, 1983; Christensen et al., 1998.). On the observation day, summer season, it is popular to burn the wastes of crop after harvest in the countryside fields of China. Moreover, Okada et al. (1999) showed the weight ratios of S/(S + K) in individual particles collected at Tanggula, which is located at 5150–5600 m altitude and Lhasa (3600 m) on the Tibetan plateau and found low ratios of S/(S + K) in smaller particles. They stated that the presence of sulfur particles with K collected in the atmosphere of Tibetan plateau is likely to be formed by biomass burning. Tang et al. (1999) also showed that major ionic components in aerosols in Lhasa are SO_4^{2-}, NO_3^-, K^+ and Ca^{2+} and high concentrations of K^+ may be attributed to biomass burning and religious activities.

It is considered that the "S + K" particles found in samples 5, 8 km would be from burning processes in the continent including the religious activities in the Tibetan plateau and be transported to the middle troposphere. As found in Table 1, there were "S-rich" particles with K and "K-rich" particles in the lower stratosphere. The results may suggest the occurrence of the transport of particles originated from burning emission into the lower stratosphere.

3.6. Mineral particles

Fifteen particles mainly composed of mineral were found at altitudes of ∼ 5, ∼ 8 and ∼ 11 km, Table 1, indicating that mineral particles could be transported to the upper troposphere. Mineral particles with sulfuric acid (4 particles), with sulfate (2) in the troposphere, were also collected, see Table 1 and Fig. 2b. These results suggest the formation of sulfuric acid and sulfate on mineral particles by heterogeneous processes, similar to the findings of Sheridan et al. (1992) and Ikegami et al. (1993). Twenty-seven sulfuric acid particles with Fe were found in the lower stratosphere (P(Fe) > 10%). In the troposphere 3 such particles were found at ∼ 11 km. Mossop (1965) suggested that small Fe-containing particles may be of extra-terrestrial origin.

3.7. Sea-salt particles

Four sea-salt particles were found, 2 at ∼ 5 km and 2 at ∼ 17 km. One sea-salt particle mixed internally with

minerals (Fig. 3e) revealed the formation by cloud processes on the way of the transport to the lower stratosphere (Andreae et al., 1986; Niimura et al., 1998). Sea-salt particles in the stratosphere over continent suggest transport through severe convective processes and air circulation.

4. Summary and conclusions

Aerosol particle samples from the lower troposphere to the lower stratosphere were obtained by using a balloon-borne impactor, together with the measurement of aerosol number concentration using a balloon-borne optical particle counter (OPC) at Xianghe (39.45°N, 117.0°E), China. Individual aerosol particles in the 5 samples (3 samples in the troposphere and 2 samples in the lower stratosphere) were analyzed by using a transmission electron microscope equipped with an energy-dispersive X-ray (EDX) analyzer to examine particle morphology, size and elemental composition. Some summary and conclusions are given as follows.

(1) The vertical distribution of aerosol number concentration ($r \geqslant 0.2 \mu m$) showed a maximum value of the stratospheric aerosol layer with concentration of 2.2 cm^{-3} which was much more than that in the background concentration. It suggests that the effect of Mt. Pinatubo eruption still remained in the stratosphere after the volcanic eruption more than 2 years ago.

(2) The particle types in the radius range of 0.1–0.5 μm (Table 1, column M) are summarized as follows: Sulfate particles were dominant 96% (44/46 particles) in the "S-rich" particles collected between 4–6 km altitude. Above 7 km altitude, the fraction of sulfate particles tended to decrease with increasing altitude. And, sulfuric acid particles were dominant in "S-rich" particles, that is, sulfuric acid particles were present in 83% (35/42) of "S-rich" particles at ∼ 8 km altitude, 94% (48/51) at ∼ 11 km, 97% (63/67), at ∼ 17 km, and 94% (45/48) at ∼ 21.2 km. On the other hand, mineral particles were collected in the troposphere (4–9 km) and the proportions were 6–11%. In the summary, the particles of "S-rich" (252) and mineral (11) appeared about 97% (263/272) in collected samples of 0.1–0.5 μm radius range.

(3) "S + K" particles ("S-rich" particles with K) were collected both in the troposphere and lower stratosphere. The presence of "S + K" particles in the troposphere was also studied using backward trajectory of air parcel. It was considered that the "S + K" particles between 4 and 9 km altitude would be originated from biomass burning processes and the religious activities in the Tibetan plateau and be transported to the middle troposphere. The presence of "S + K" particles and "K-rich" particles in the lower stratosphere may suggest the occurrence of the transport of particles originated from burning emission

into the lower stratosphere as expected by sea-salt particles found in the lower stratosphere in this research.

(4) Sulfuric acid particles with Fe were present in the lower stratosphere. In the radius range of 0.1–0.5 μm, 20–30% of sulfuric acid particles contained Fe (P(Fe) > 10%). The source of Fe detected in sulfuric acid particles may be extra-terrestrial particles.

(5) Particles mainly composed of minerals were collected in the troposphere, 11% (6/55 particles) at 5 km, 6% (3/47) at 8 km, 3% (2/64) at 11 km, indicating the vertical transport particles from near surface to the upper troposphere.

(6) Sulfuric acid and sulfate detected in the mineral particles suggest the formation of sulfuric acid and sulfate on mineral particles by heterogeneous processes in the troposphere.

(7) The presence of sea-salt particles in the lower stratosphere is due to the vertical transport to the upper troposphere by convective clouds and then the invasion to the lower stratosphere by air exchange between the troposphere and stratosphere or tropopause folding.

Acknowledgements

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