INDIVIDUAL AEROSOL PARTICLES IN THE ANTARCTIC UPPER TROPOSPHERE

Yasunobu IWASAKA, Kikuo OKADA and Akira ONO

Water Research Institute, Nagoya University, Furo-cho, Chikusa-ku, Nagoya 464

Abstract: Individual aerosol particles were collected in the Antarctic upper troposphere (24000 ft=7.2 km, 15000 ft=4.5 km and 12000 ft=3.6 km) and morphological features of the particles were investigated through electron microscopy. The results suggested that sulfate-containing particles were dominant in the submicron size range throughout three measurements made on 25 January 1983, 29 October 1983 and 13 January 1984. At the end of January 1983 particles with irregular shape coated with sulfuric acid solution were present at an altitude of 24000 ft=7.2 km. The presence of these particles would be due to the transportation of particles originated from the eruption of El Chichón (April 1982, Mexico).

1. Introduction

Many investigators' interest have been directed to the dispersion processes of stratospheric aerosol particles from a viewpoint of the transport of minor constituents in the stratosphere, the effect of stratospheric particles on the transfer of solar radiation and atmospheric radiation, and so on. Recently, the volcanic eruption of El Chichón has severely influenced the stratosphere.

Volcanic explosions are very important sources of stratospheric aerosols because of the injection of sulfur-containing gases into the stratosphere. The volcanic materials brought into the stratosphere have been considered to be dispersed on a global scale by the large-scale motion of the air.

Information on the volcanic materials transported into the polar stratosphere and troposphere is important in various fields, such as meteorology, geochemistry and glaciology. Good and typical example showing the global transport of volcanic materials is found in the chemical analysis of the ice core sampled in the Antarctic and Arctic regions which reveals the presence of "Anomolous Enriched Elements (AEE)", Pb, Cd, Cu, Zn, Ag and Hg due to a possible global dispersion of volcanic materials into the stratosphere by severe volcanic eruptions.

According to MITCHELL (1975) the observed variations of the enrichment factors of AEE's, Pb, Cd, Zn and Cu at Dome C during the period from 1914 to 1974 coincided fairly well with the variations of the past volcanic activity. BOUTRON (1980) also reported that the period of the 1880's to 1977 was characterized by several exceptionally strong volcanic eruptions which produced world-wide effect.

The eruption (April 1982, Mexico) of El Chichón (17.33°N, 93.2°W) certainly produced a large amount of stratospheric aerosols. The ground-based lidar data showed the enhancement of stratospheric aerosol layer by the eruption of El Chichón to be the

largest event experienced in the last 10 years. MCCORMICK *et al.* (1984) undertook complehensive studies on the dispersion of El Chichón clouds using airborne lidar and satellite. According to their airborne lidar measurements in October–November 1982, the volcanic clouds had dispersed from the northern hemisphere to the southern hemisphere by that time and the most massive and optically thick regions having mixing ratio of particles from 10 to 30 were found between 35°N and 10°S, and the region of mixing ratio from 1 to 3 between ~40°N and ~35°S. From their results it may be possible to speculate that the volcanic clouds had spread into the southern polar stratosphere by the end of 1982.

Fortunately we succeeded in sampling the airborne particles over Syowa Station (69°00′S, 39°35′E) in January 1983, October 1983 and January 1984. In this paper we should like to present the results of the electron microscopic examinations on the individual aerosol particles collected by an impactor in the Antarctic atmosphere (sampling heights are 24000 ft=7.2 km, 15000 ft=4.5 km, and 12000 ft=3.6 km), and discuss the effect of the volcanic eruption of El Chichón on the polar atmosphere.

2. Methods

The aircraft of Japanese Antarctic Research Expedition (JARE), a PC-6 Turbo Porter, was used to collect aerosol particles in the Antarctic troposphere. Air was introduced into a cabin of the aircraft through the isokinetic decelerator (Fig. 1). And

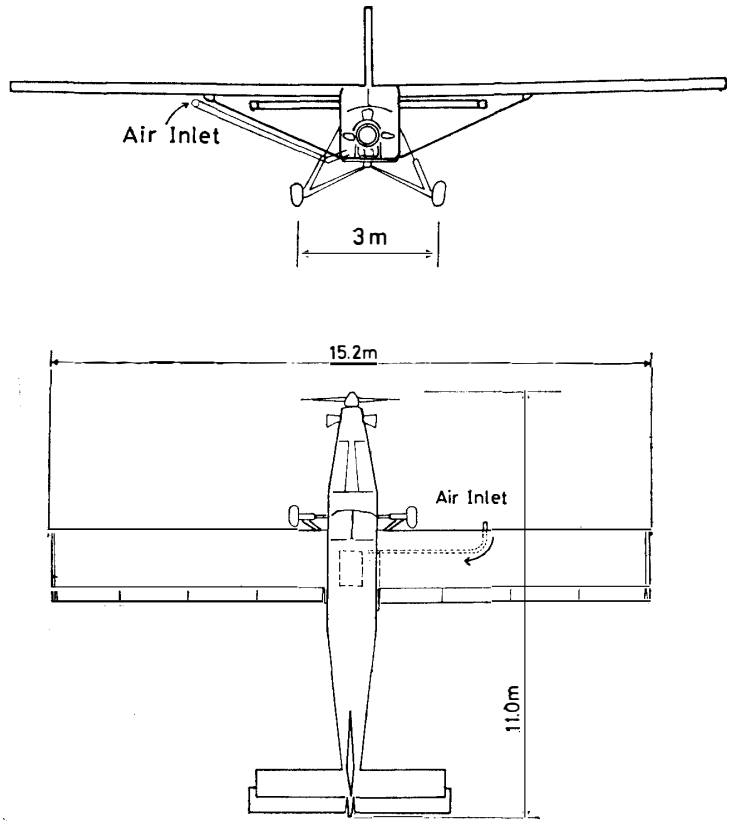

Fig. 1. Air inlet for the aerosol sampling. The location of the air inlet is chosen to eliminate the contamination due to the outgassing from the aircraft, PC-6 Turbo Porter, and to minimize the line losses of particles.

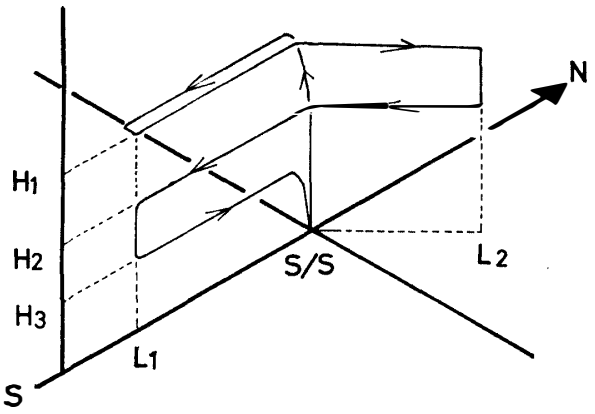

Fig. 2. A schematic map of typical aircraft flight. S/S is Syowa Station (69°00'S, 39°35'E). H_1, H_2 and H_3 are the heights of flight level of 24000, 15000 and 12000 ft, respectively. Distances from Syowa Station L_1 and L_2 are 150 and 100 km, respectively.

Table 1. Sampling periods and altitudes.

25 January 1983	1139–1209 LST	24000 ft
	1225–1255	12000
	1306–1336	4000
29 October 1983	1241–1341	24000
	1351–1451	15000
13 January 1984	1650–1720	15000
	1752–1808	9000

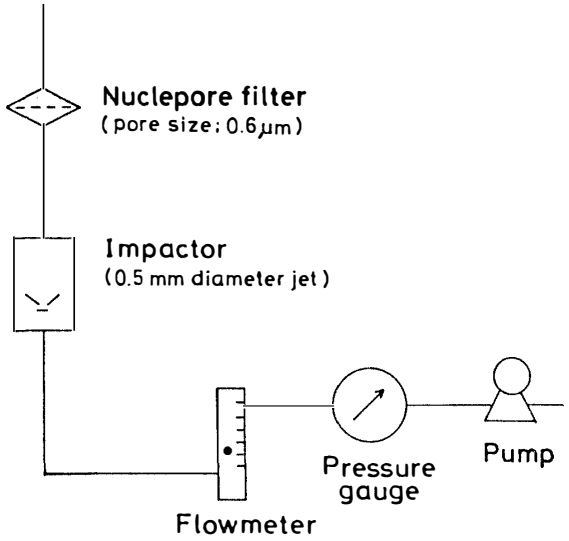

Fig. 3. A schematic diagram of a low-pressure impactor (OKADA, 1983).

a tube from the intake to a sampler was kept as short as possible. Flight paths are shown in Fig. 2, where "S/S" shows the location of Syowa Station. The collection of particles was carried out at altitudes of H_1, H_2 and H_3. The horizontal distances from Syowa Station, L_1 and L_2, were 150 and 100 km, respectively. In Table 1 the sampling heights and the periods are summarized.

Individual aerosol particles were collected on a carbon-covered nitrocellulose film with a low pressure impactor. Figure 3 shows a schematic diagram of the low-pressure impactor used in the present study. Aerosol particles passing through a Nuclepore filter (pore size: 0.6 μm) were collected with the impactor of 0.5-mm diameter jet. This impactor was designed to collect particles of 0.03–0.35 μm radius. As shown later the particles with radii larger than 0.3 μm were often collected on the impactor

stage owing to the presence of pore size larger than 0.6 μm. More detailed explanation of this impactor was given by OKADA (1983). Morphology of individual particles was examined by a transmission electron microscope (HITACHI HS-9). For most of samples morphological features of particles were investigated.

3. Results

Vertical distributions of air temperature, dew point temperature and wind during

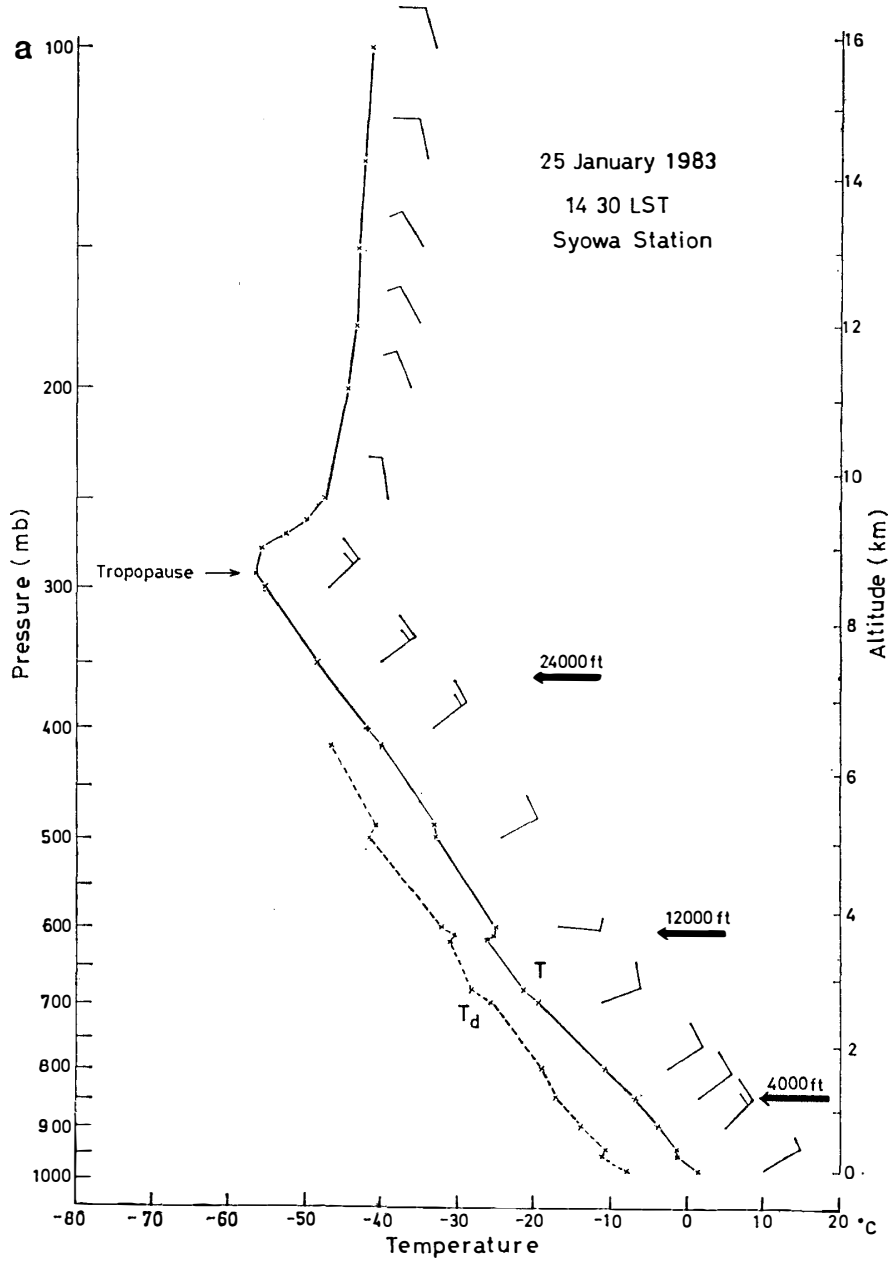

Fig. 4a.

Figs. 4a–4c. Vertical distributions of air temperature, dew point tempoerature and wind on the days when the collections of aerosol particles were carried out. The altitudes of the collections are also shown.

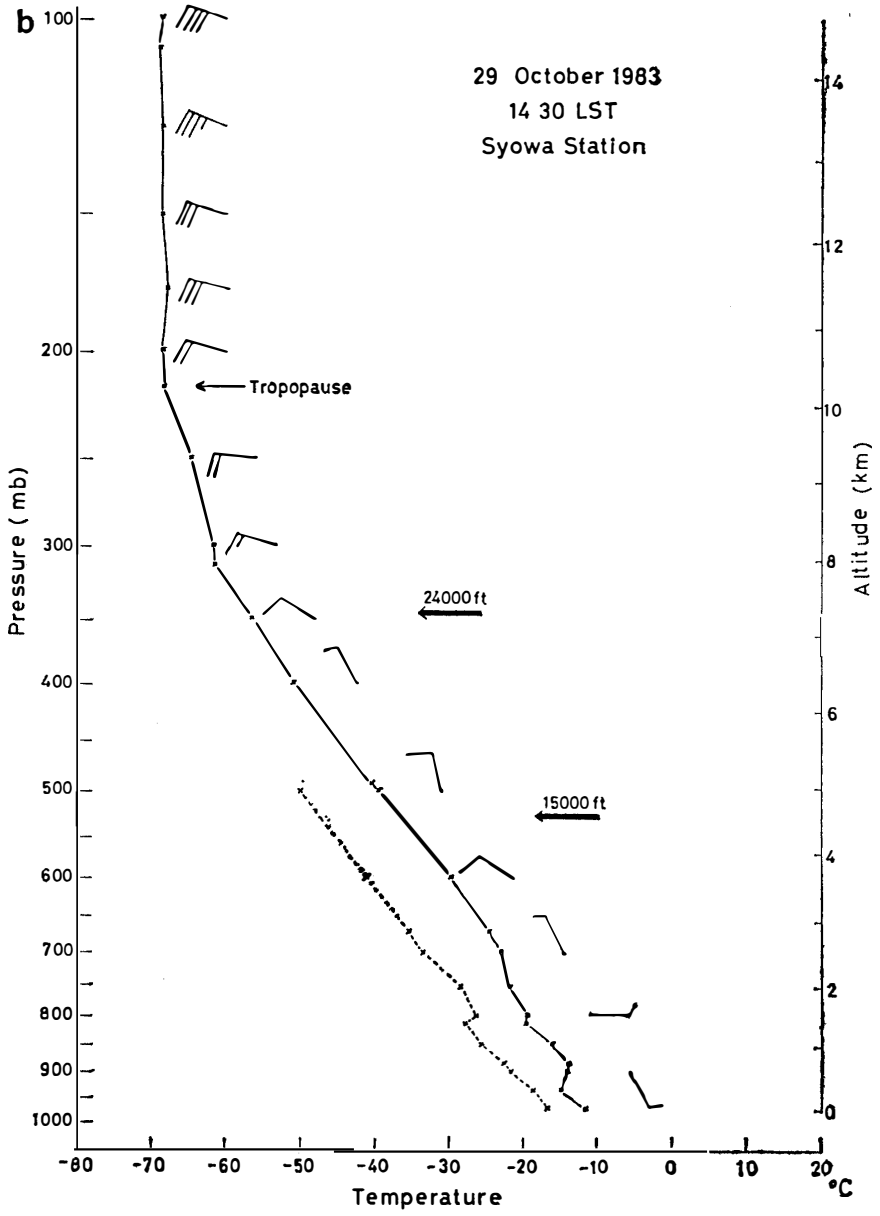

Fig. 4b.

the observational periods are shown in Figs. 4a–4c, together with the altitudes where particles were collected.

3.1. Particles sampled on 25 January 1983

Figure 5 shows the electron micrograph of aerosol particles collected at an altitude of 24000 ft=7.2 km during the period from 1139 to 1209 LST on 25 January 1983. As shown in the figure most particles are those with satellite droplet rings which are peculiar sulfuric acid particles as pointed out by FRANK and LODGE (1967). The particle marked by A in Fig. 5 is found to be composed of the material with satellite droplet rings on the collecting surface and contain no inclusion of electron-dense material. The volume fraction of sulfuric acids would be high in the particle. The particle designated by B is an example of particles with electron-dense inclusions surrounded by satellite droplet rings. The electron-dense inclusion is irregular in shape

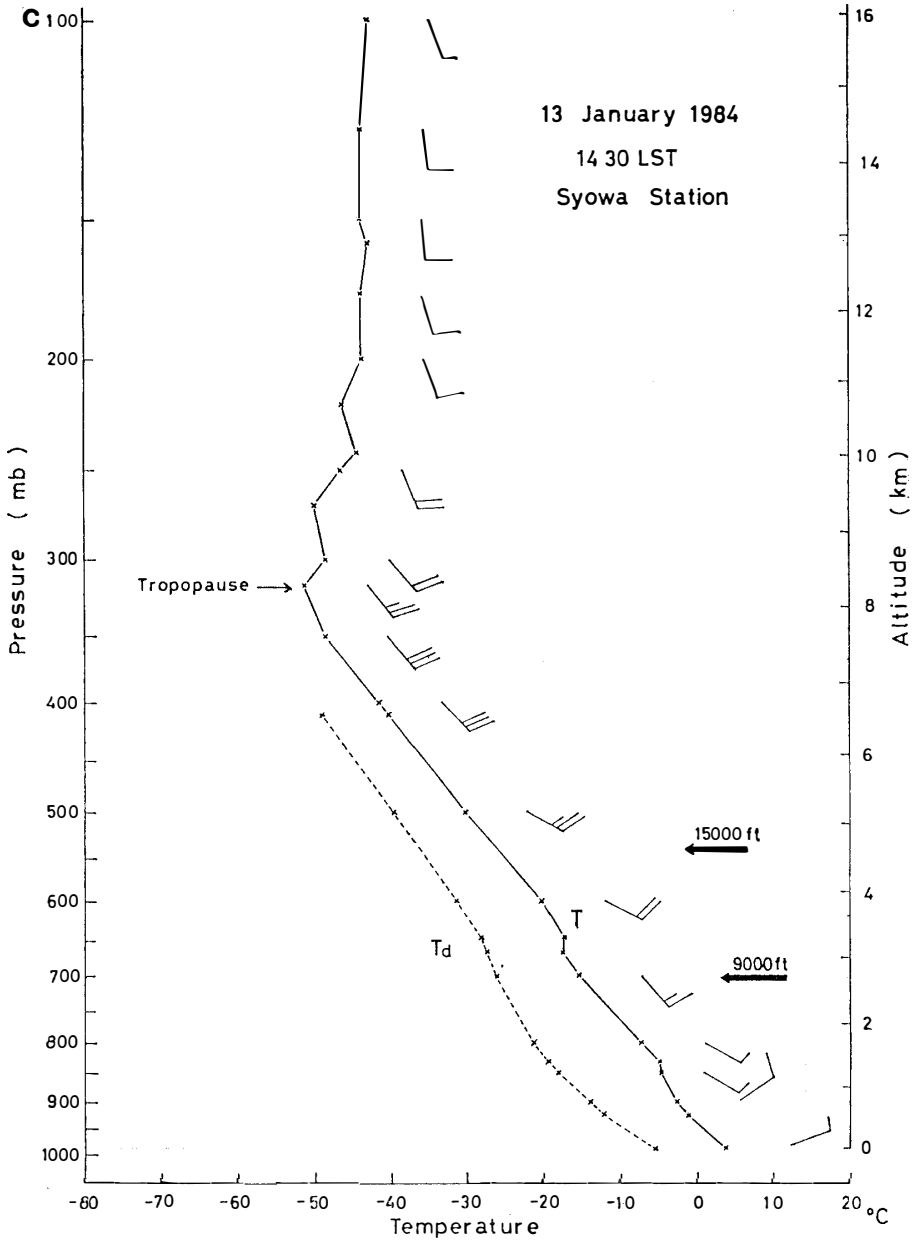

Fig. 4c.

suggesting water-insoluble material. The particle marked by C is one without satellite droplet rings and contains volatile material under the electron beam of weak intensity. On the basis of the morphological investigation, the particles collected at 24000 ft= 7.2 km are characterized by a high number fraction of sulfuric acid-containing particles and by the presence of irregular shaped inclusion which seems to be water-insoluble material.

Figure 6 shows the electron micrograph of aerosol particles collected at an altitude of 12000 ft=3.6 km during the period from 1225 to 1255 LST on 25 January 1983. Features of the particles are different from those at 24000 ft=7.2 km. Although the particles with satellite droplet rings are also dominant, central particles surrounded by droplet rings are usually present and they show the morphological features similar

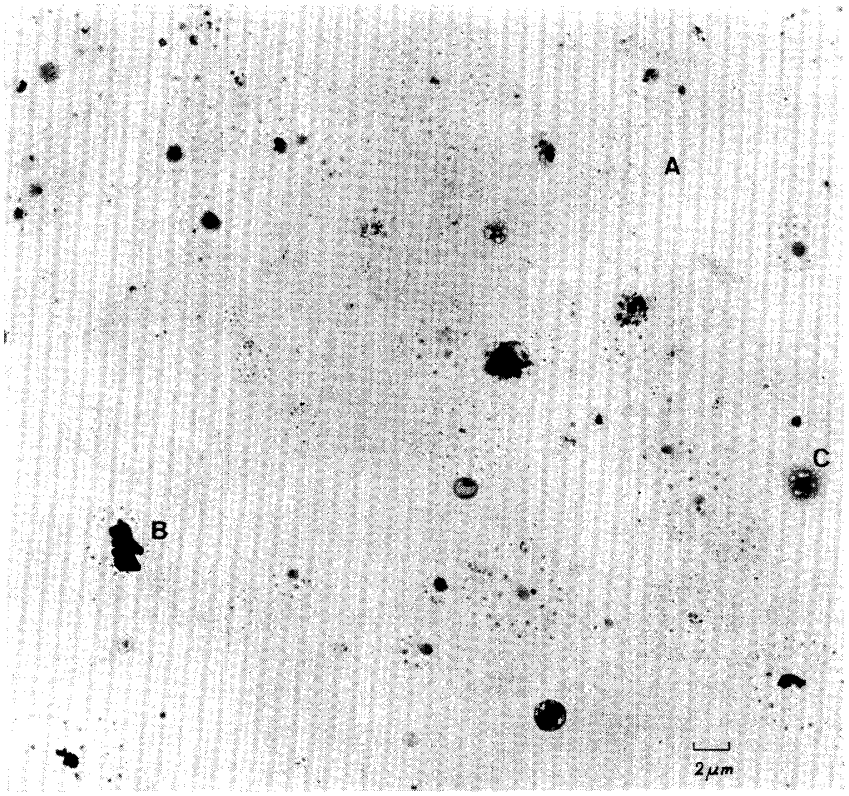

Fig. 5. Electron micrograph of aerosol particles collected at 24000 ft on 25 January 1983. The collection was carried out during the period from 1139 to 1209 LST.

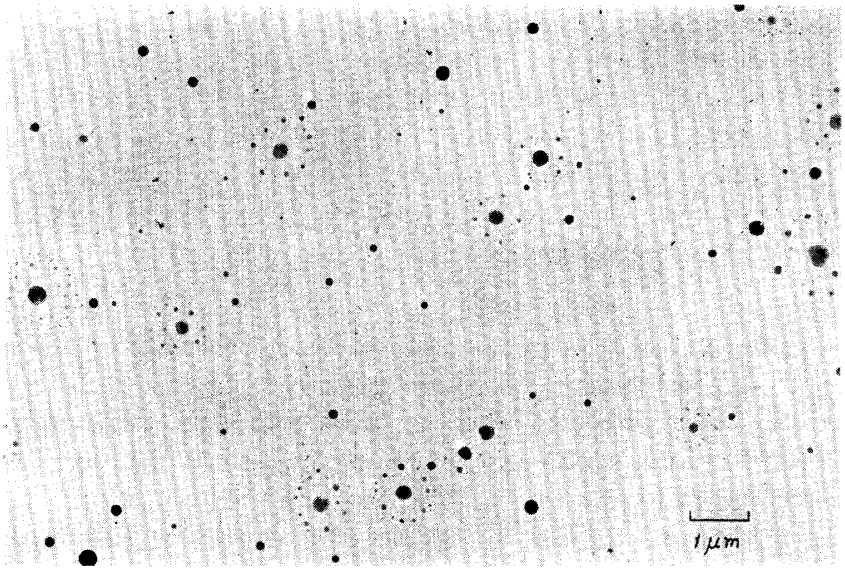

Fig. 6. Electron micrograph of aerosol particles collected at 12000 ft on 25 January 1983. The collection was carried out during the period from 1225 to 1255 LST.

to ammonium sulfate particles reported by HEARD and WIFFEN (1969). Thus, these particles collected 12000 ft=3.6 km are probably an admixture of sulfuric acid and ammonium sulfate. It should be noted that electron-dense material with irregular shape as shown in Fig. 5 was not detected in the sample.

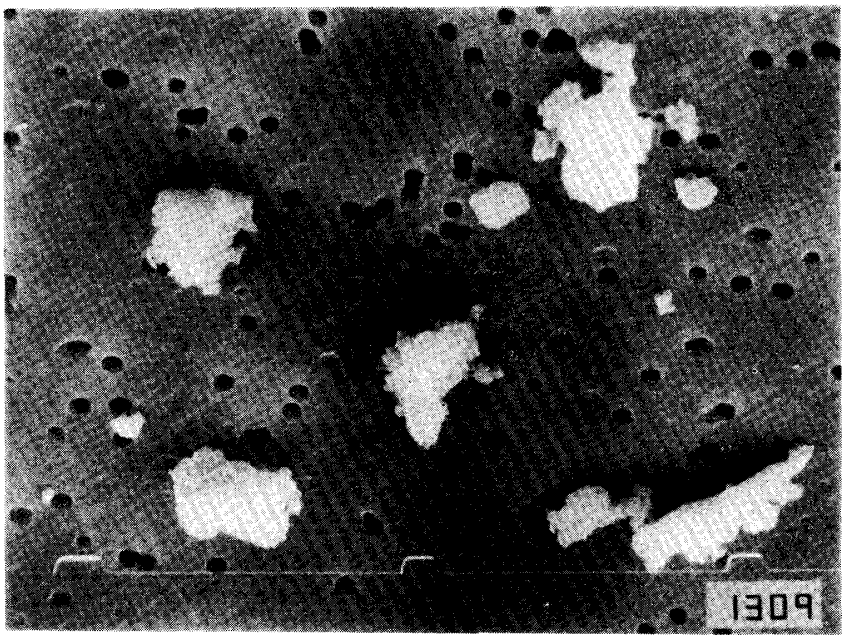

Fig. 7. Scanning electron micrograph of particles collected on a Nuclepore filter (pore size: 0.6 μm) used as the pre-filter of the low-pressure impactor shown in Fig. 3. The surface structure of the filter is also illustrated here.

Figure 7 shows the scanning electron micrograph of particles collected on a Nuclepore filter (pore size, 0.6 μm) used as the pre-filter of the low-pressure impactor during the collections on 25 January. It is found that particles with radius of about 0.2 μm are present and they are irregular in shape. Although it is difficult to determine the collection altitude, there is a possibility of the presence of these particles at an altitude of 24000 ft=7.2 km, because of the absence of irregular-shaped particles at 12000 ft=3.6 km altitude. Together with this, information on the surface structure of the Nuclepore filter is also given in Fig. 7. The pore size is not always 0.6 μm and the value tends to scatter. The filter of this type may lead to the presence of particles of about 1 μm radius on the collecting surface shown in Fig. 5.

3.2. 29 October 1983

Figure 8 shows the electron micrograph of aerosol particles collected at an altitude of 24000 ft=7.2 km during the period from 1241 to 1341 LST on 29 October 1983. Figure 8a is the electron micrograph taken before the evaporation of barium chloride. Although particles with satellite droplet rings are detected, most particles have the morphological features similar to ammonium sulfate particles. The morphological features of the particles with the droplet rings are different from those collected on 25 January 1983 at 24000 ft=7.2 km, that is, the content of sulfuric acid in particles would be small as compared with that obtained on 25 January. Figure 8b shows the electron micrograph of the same particles as those in Fig. 8a, coated with a vapor-deposited thin film of barium chloride (20 Å in thickness). The evaporation of barium chloride was made in high vacuum of about 4×10^{-6} torr with a shadowing angle of arctan 0.5 (26.57°). The shadow length of particles in Fig. 8b is quite short, which indicates that the shape of sulfate particles on the substratum is flat. Figure

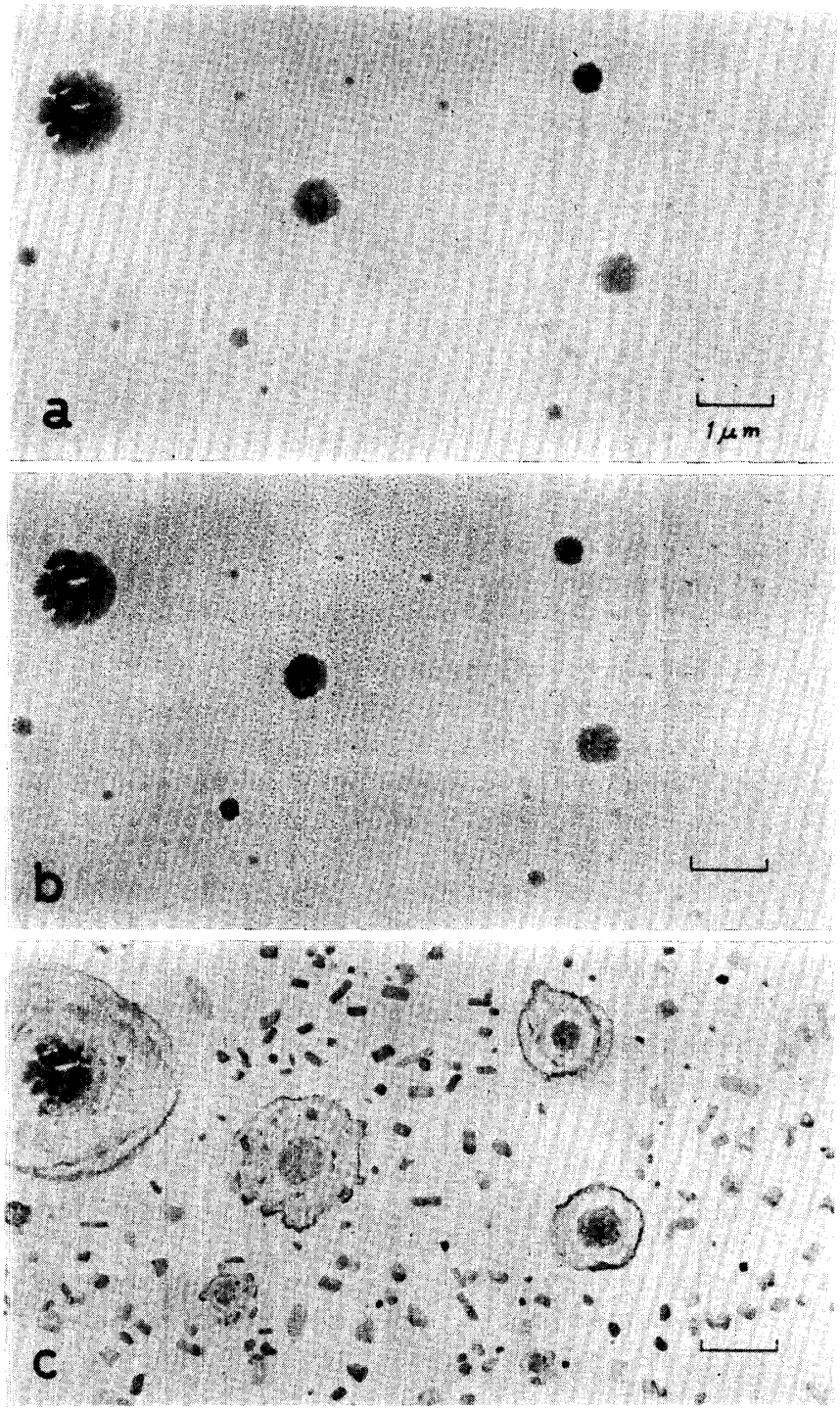

Fig. 8. a. Electron micrograph of aerosol particles collected at 24000 ft on 29 October 1983. The collection was carried out during the period from 1241 to 1341 LST. This photograph was taken before the evaporation of barium chloride. b. Electron micrograph of aerosol particles shown in Fig. 8a coated with a vapor-deposited thin film of barium chloride (20 Å thickness). The evaporation of barium chloride was carried out in high vacuum of about 4×10^{-6} torr with a shadowing angle of arctan 0.5 (26.57°). c. Electron micrograph of the reaction product of barium chloride with particles shown in Fig. 8a after exposure to octanol vapor for 24 h.

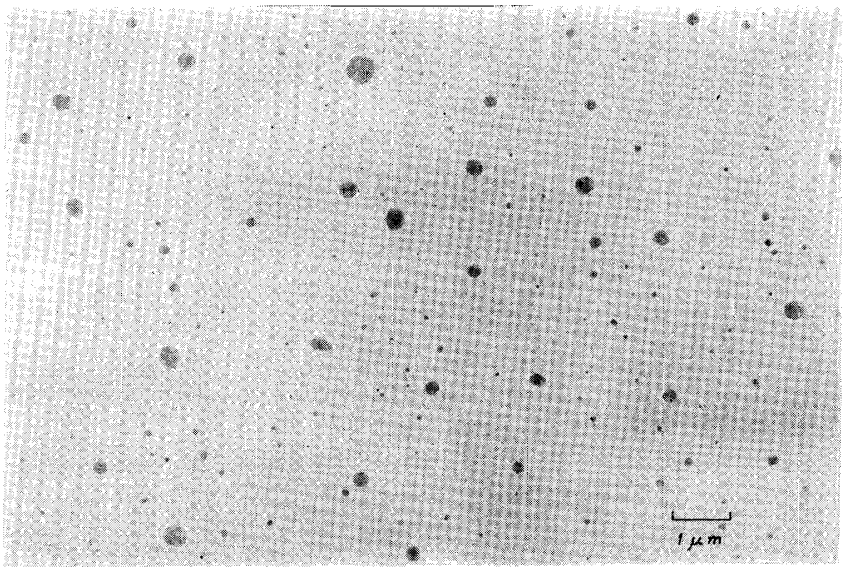

Fig. 9. Electron micrograph of aerosol particles collected at 15000 ft on 29 October 1983. The collection was carried out during the period from 1351 to 1451 LST.

8c shows the electron micrograph of reaction product of barium chloride with particles shown in Fig. 8a after exposure to octanol vapor for 24 hours. Comparison of these electron micrographs reveals that particles which formed the Liesegang's rings are those containing sulfate ions. The validity of this technique was presented by ONO *et al.* (1981). On the basis of the examination, it can be said that most particles are sulfate-containing particles which are probably composed of ammonium sulfate with a high volume fraction, though some particles have satellite droplet rings indicating the presence of sulfuric acid in small fractions.

Figure 9 shows the particles collected at an altitude of 15000 ft=4.5 km during the period from 1351 to 1451 LST on 29 October 1983. Particles with satellite droplet rings are hardly detected in the sample. Substantially all of the particles show the same morphological features as ammonium sulfate particles reported by HEARD and WIFFEN (1969).

3.3. 13 January 1984

Figures 10a and 10b show the electron micrographs of aerosol particles collected at an altitude of 15000 ft=4.5 km during the period from 1650 to 1720 LST on 13 January 1984. Morphology of particles in Fig. 10a are similar to those in Fig. 9. However, it is remarkable that only particles larger than about 1 μm radius show the satellite structure in Fig. 10a, which implies the presence of sulfuric acid.

4. Discussion

On the basis of electron microscopic examinations on the aerosol particles collected in the upper Antarctic troposphere on 25 January 1983, 29 October 1983, and 13 January 1984, the important features are summalized as follows:

1) Concerning the shape of particles under the electron microscope there is a noticeable difference between the results on 25 January 1983 and those on the other

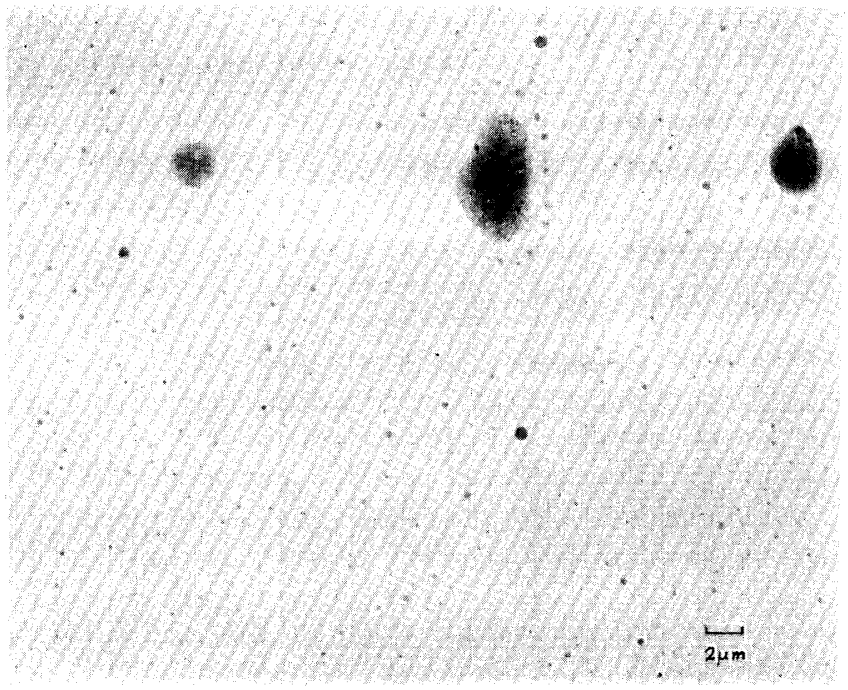

Fig. 10a.

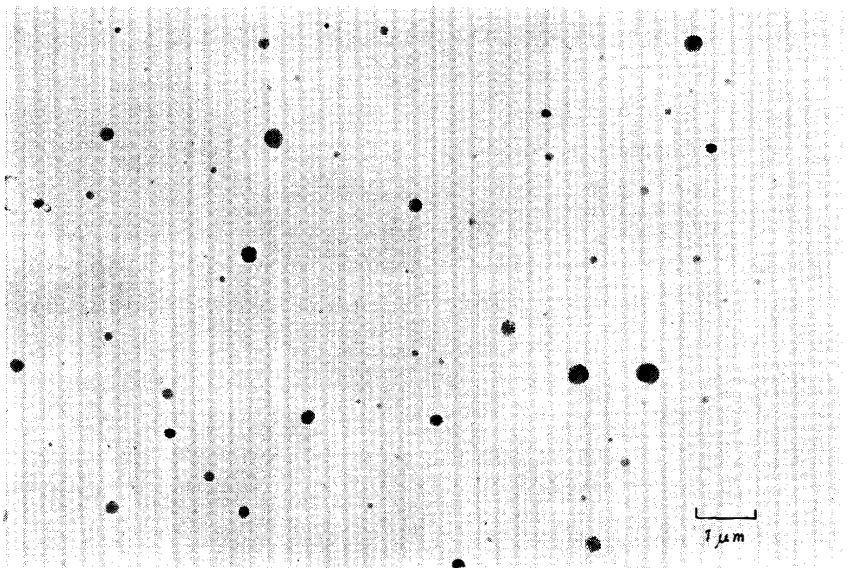

Fig. 10b.

Fig. 10. Electron micrographs of aerosol particles collected at 15000 ft on 13 January 1984. The collection was carried out during the period from 1650 to 1720 LST. In Fig. 10a only larger particles than 1 μm radius indicate satellite structure.

days. The particles with irregular-shaped material, possibly water-insoluble material, were collected just below the tropopause on 25 January 1983. Such particles were not detected in the samples collected on 29 October 1983 and 13 January 1984.

2) Most of the particles collected just below the tropopause on 25 January 1983 showed a satellite structure on the collecting surface which has been frequently observed in the stratospheric aerosol particles composed of sulfuric acid solution (*e.g.*, GRAS and LABY, 1979).

3) The so-called "satellite structure" was not distinct in the samples collected on 29 October 1983 and 13 January 1984. However, thin sulfuric acid solution, as shown in Fig. 8a, possibly coated the surface of particles.

4) Most particles collected one year after the first observation did not show a satellite structure, except for the larger size particles.

The volcanic eruption of El Chichón disturbed the stratosphere on the global scale. The aerosol particles that had been brought into the stratosphere dispersed not only to the northern hemisphere but also to the southern hemisphere. According to the airborne lidar measurements (MCCORMICK *et al.*, 1984) the volcanic clouds reached the mid-latitudes region (about 50°S) by October 1982 and appeared to spread well throughout the latitudes of 56°S to 72°N by May 1983. Considering the rapid dispersion rate (about 10°/month) of volcanic material it is reasonable to consider that the volcanic clouds influenced the polar stratosphere by the end of 1982. Therefore, it is necessary to take this effect into consideration when we interpret the electron micrographs of the particles collected over Syowa Station (69°00′S, 39°35′E), Antarctica.

As far as we know, the large water-insoluble particles such as the those collected on 25 January 1983 have not been found near the tropopause during the undisturbed atmospheric periods. Moreover, it is interesting that such large water-insoluble particles were not found in the measurements on 29 October 1983 and 13 January 1984.

The cold winter polar stratosphere, as suggested by IWASAKA (1985) and MCCORMICK *et al.* (1985), possibly cleaned out the particulate matter from the strtaosphere during the winter. If so, the volcanic effect can be hardly detected in the polar stratosphere immediately after the 1983 winter. Then, the active air mass exchange associated with sudden warming events certainly transports various materials from low- and mid-latitudes. The lidar measurements at Syowa Station (IWASAKA, 1985) and the balloon observation (MORITA *et al.*, 1984; ITO *et al.*, 1984) showed that the Antarctic stratosphere was still under the influence of the volcanic eruption of El Chichón. Especially, the results on the Aitken particles (ITO *et al.*, 1984) showed that very active particle formation advanced in the Antarctic stratosphere in middle October 1983. Considering the situation, Fig. 10a shows very interesting features that only large size particles had the so-called satellite structure suggesting the sulfuric acid solution and most of the smaller size particles seemed to be ammonium sulfate particles. One possible interpretation of this state is that the large size particles were originated in the stratosphere where the partial pressure of sulfuric acid vapor enabled the formation of the thin layer of sulfuric acid solution coating the aerosol particles.

5. Conclusion

Summarizing the results, the aircraft measurements on the upper tropospheric particles suggest that the volcanic materials of El Chichón were transported into the Antarctic stratosphere and upper troposphere by the end of January 1983. Unfortunately it is impossible to make the trajectory analysis of the air mass due to the lack of meteorological data. However, it is hoped that a more clear interpretation will

be given in the near future by combining the related results gained at Syowa Station, such as lidar data, balloon data and so on.

Acknowledgments

We should like to thank the wintering members of the 24th Japanese Antarctic Research Expedition (leader Prof. S. MAE, Hokkaido University) for their kind assistance during the aerosol sampling. One of us (Y.I.) extends his thanks to Prof. A. TAKAHASHI, Kobe University, who gave technical support and advice for the observation of collected aerosols by a scanning electron microscope at Syowa Station.

References

BOUTRON, C. (1980): Respective influence of global pollution and volcanic eruptions on the past variations of the trace metals content of Antarctic snows since 1880's. J. Geophys. Res., **85**, 7426–7432.

FRANK, E. R. and LODGE, J. P., Jr. (1967): Morphological identification of airborne particles with the electron microscopy. J. Microsc., **6**, 449–456.

GRAS, J. L. and LABY, J. E. (1979): Southern hemisphere stratospheric aerosol measurements; 2. Time variations and the 1974–1975 aerosol events. J. Geophys. Res., **84**, 303–307.

HEARD, M. J. and WIFFEN, R. P. (1969): Electron microscopy of natural aerosols and the identification of particulate ammonium sulfate. Atmos. Environ., **3**, 337–340.

ITO, T., IKEGAMI, M., KANAZAWA, I. and IWASAKA, Y. (1984): Balloon-borne observation of Aitken nuclei in the Antarctic troposphere and stratosphere. Program and Abstracts; International MAP Symposium, Kyoto, Nov. 26–30, 143.

IWASAKA, Y. (1985): Lidar measurement of the stratospheric aerosol layer at Syowa Station (69.00°S, 39.35°E), Antarctica. J. Meteorol. Soc. Jpn., **63**, 283–287.

MCCORMICK, M. P., SWISSLER, T. J., FULLER, W. H., HUNT, W. H. and OSBORN, M. T. (1984): Airborne and ground-based lidar measurements of the El Chichón stratospheric aerosol from 90°N to 56°S. Geofis. Int., **23-2**, 187–221.

MCCORMICK, M. P., HAMILL, P. and FARRUKH, U. O. (1985): Characteristics of polar stratospheric clouds as observed by SAM II, SAGE, and lidar. J. Meteorol. Soc. Jpn., **63**, 267-276.

MITCHELL, M. J. (1975): A reassessment of atmospheric pollution as a case of long term changes of global temperature. The Changing Global Environment, ed. by S. F. SINGER. Dordrecht, D. Reidel, 149–173.

MORITA, Y., TAKAGI, M., IWASAKA, Y. and ONO, A. (1984): Balloon measurements of aerosols in the Antarctic stratosphere. Program and Abstracts; International MAP Symposium, Kyoto, Nov. 26–30, 142.

OKADA, K. (1983): Nature of individual hydroscopic particles in the urban atmosphere. J. Meteorol. Soc. Jpn., **61**, 727–736.

ONO, A., OKADA, K. and AKAEDA, K. (1981): On the validity of the vapor-deposited thin film of barium chloride for the detection of sulfate in individual atmospheric particles. J. Meteorol. Soc. Jpn., **59**, 419–424.

(Received May 22, 1985; Revised manuscript received September 25, 1985)