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Balloon observation of aerosols in the antarctic troposphere and stratosphere

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ABSTRACT

Balloon soundings of aerosols up to 15 km in height were carried out in 1983 at Syowa Station (69°00′S, 39°35′E), Antarctica. The vertical distribution of the concentration and size distribution of Mie particles (diameter larger than 0.3 μm) was obtained on 3 June and on 16 October. The vertical distribution of the concentration of Aitken particles (diameter larger than 0.004 μm) was obtained on 17 October. This paper gives a review of the findings obtained by these soundings and discusses the significance of these findings in aerosol processes in the antarctic atmosphere.

1. Introduction

Increasing attention has been directed to the problem of global background aerosol pollution because of its possible effect on climate. In order to clarify the mechanism of global background aerosol pollution, an essential requirement is to obtain a better understanding of aerosols in a "clean" atmosphere over remote regions. Antarctica is an important region in the study of the background aerosol.

From extensive surface observations of aerosols carried out at Syowa Station (69°00′S, 39°35′E), Antarctica, it has been revealed that various phenomena relating to the long-range transport of trace constituents and/or photochemical processes resulting in the production or growth of aerosol particles are clearly observed in Antarctica (Ito and Iwai, 1981; Ito, 1983, 1985). Furthermore, the importance of the polar stratosphere in aerosol study has been recognized in the recent work of Rosen and Hofmann (1983) and McCormick (1985). However, the direct observation of aerosols over Antarctica using an airplane or a balloon has been quite rare (Hofmann et al., 1973, 1976, 1977; Hogan, 1979). Most of these observations were made during summer. Considering the noticeable seasonal change of the stratospheric aerosol layer, it is necessary to gain information on aerosols in seasons other than summer in order to obtain a comprehensive understanding of polar aerosols.

As a part of the Middle Atmosphere Program by the Japanese Antarctic Research Expedition, a program of observation of gases and aerosols over Antarctica was started in 1983 at Syowa Station, using lidar, a Fourier Transform Infrared Spectrometer, an instrumented airplane, and aerosol sondes. The preliminary reports on some of these observations are given elsewhere (Ito et al., 1985; Iwasaka et al., 1985a; Morita et al., 1985). This paper gives a review of our recent findings obtained by balloon observations of antarctic aerosols carried out in 1983 at Syowa Station and discusses the significance of these findings in aerosol processes in the antarctic atmosphere.

2. Instrument

The vertical distribution of number concentration and size distribution of aerosol particles with diameter greater than 0.3 μm (hereafter referred as Mie particles) was observed by a sonde which comprises a light scattering aerosol counter. The sonde is similar to that used by Hofmann and Rosen (1981) in principle, and the details are given by Morita and Takagi (1983). The light scattering aerosol counter has two pulse height discriminators which enable separate counting of particles larger than 0.3 μm and those larger than 0.5 μm in diameter, assuming spherical particles with a refractive index of 1.4. Thus, a rough indication of the size distribution is obtained, although aerosol particles do not always have a refractive index of 1.4 and a spherical shape. The time taken to complete one set of measurement of all parameters (temperatures of the air and the pump, pressure, concentration of particles larger than 0.3 μm and that larger than 0.5 μm) is 75 s. In order to calibrate the rate of air flow and the light source intensity, the lobe pump rotation rate and the light intensity are checked at intervals of about 10 min during flights. The total weight of the sonde is about 12 kg.

The vertical distribution of number concentration of aerosol particles with diameter larger than 0.004 μm (hereafter referred as Aitken particles) was observed by a sonde which is comprised of an adiabatic expansion fog chamber with photoelectric detection. The sonde is completely different from that used by Rosen and Hofmann (1983) which consists of a thermal gradient growth chamber and a light scattering aerosol counter. The details of the present sonde are given by Ito et al. (1983). The concentration of Aitken particles is measured in terms of the maximum intensity of the light scattered laterally from a light pencil traversing the fog which is produced by adiabatic expansion in the chamber. The temperature of the fog chamber is maintained at about 20°C by waste heat from a water activated battery which is used as a power source for the sonde. The working fluid of the fog chamber is water. A fixed expansion ratio of 1.20, irrespective of ambient pressure, is applied, which produces a supersaturation of about 230%. The signal of measured concentration is inserted into the rawinsonde signal and transmitted to a ground station, every min for 10 s. The total weight of this sonde is 8 kg, comprising a 4 kg fog chamber, a 3.5 kg battery and a 0.5 kg rawinsonde.

3. Observations

The location of Syowa Station is indicated on the map in Fig. 1. The first Mie particle sounding in 1983 was flown at 10:30 LT on 3 June using two 3 kg-rubber balloons. On 2 June, the day before the sounding, the troposphere was warm and moist and a northerly wind prevailed, indicating that the air coming from lower latitudinal maritime regions prevailed in the entire troposphere over Syowa Station. On 3 June, the prevalence of maritime air was also recognized from the rawinsonde observation, although the sky was cloudless. In the stratosphere, we observed extremely cold air with a temperature of −82.3°C centered at 21 km and temperatures lower than −70°C above 13.5 km in height.

The second Mie particle sounding was flown at 14:30 LT on 16 October using three 3 kg-rubber-balloons. It was followed by the sounding of Aitken particles which was flown at 16:30 LT on 17 October using two 3 kg-rubber-balloons. These two soundings were planned to obtain direct information on the large enhancement of stratospheric aerosol which was detected on 15

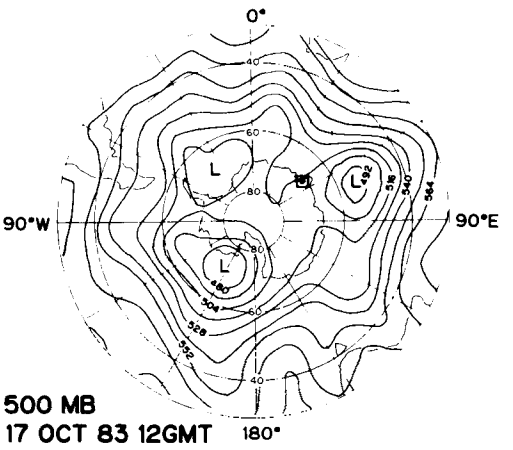

Fig. 1. Location of Syowa Station marked on the 500 mb map of 12:00 GMT on 17 October 1983.

October 1983 by lidar at Syowa Station (Iwasaka et al., 1985a). At the time of the soundings, this enhancement was imagined to have a relation to the sudden warming in the stratosphere because a slight temperature increase was observed before the enhancement. The air temperature at 14 km, which remained at around −75°C during the previous week, increased 6°C in three days before the aerosol enhancement. However, the temperature increase did not proceed further and the strongest sudden warming event was delayed to 26 October after the lowest temperature of −81°C at 12 km on 18 October. Thus, these two soundings documented the state of the stratospheric aerosol during a period in which stratospheric temperature field was experiencing a dramatic change from that of a cold season to that of a warm season, although their original aim of documenting the state of the stratospheric aerosol during a stratospheric sudden warming event failed to be accomplished.

As for the troposphere, no cloud was seen over Syowa Station from 15 to 17 October. From 11 to 14 October, the entire troposphere over Syowa Station was in a northern wind regime, whereas during the period of the two soundings it was in a southern wind regime and the air in the troposphere was rather dry. The synoptic weather map at 500 mb on 17 October shown in Fig. 1 suggests that the meridional exchange of air was rather large in the troposphere over East Antarctica during these aerosol soundings in October. It can be also noted, however, that the air over Syowa Station seems to come from the interior of Antarctica and the maritime component in the air, if any, seems to have long time history before arriving over Syowa Station.

4. Results

4.1. Mie particle sounding on 3 June in 1983

Fig. 2 shows the vertical profile of Mie particle concentration and aerosol count ratio of the two size ranges ($D \geqslant 0.3$ μm and $\geqslant 0.5$ μm) obtained on 3 June 1983. This is the first successful aerosol balloon sounding in the winter antarctic stratosphere. In the sounding, one of two balloons carrying the sonde burst at about 14 km height, but during the slow descent of the sonde after the balloon burst the concentration measurements were continued successfully. The result obtained in the descending measurement is shown with a dashed line in Fig. 2.

Both the ascent and descent measurement in the stratosphere show similar vertical profiles of Mie particle concentration and count ratios. This fact indicates that the sonde has good reproducibility in measurements. On the other hand, it also indicates that the observed profile is repre-

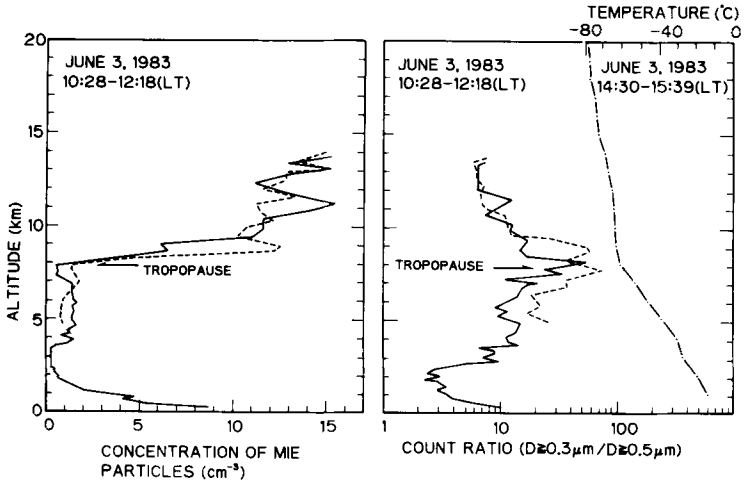

Fig. 2. Vertical profile of concentration and count ratio of Mie particles over Syowa Station on 3 June 1983. The solid and dashed lines indicate the ascending and descending measurements, respectively. The dash-dot line indicates air temperature.

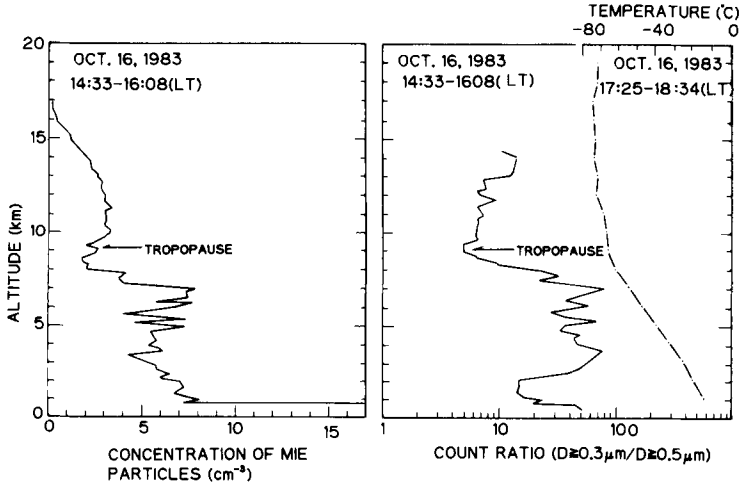

Fig. 3. Vertical profile of concentration and count ratio of Mie particles over Syowa Station on 16 October 1983. The dash-dot line indicates air temperature.

sentative for a horizontal scale of up to a hundred km as evaluated from the time difference between the ascent and descent measurements and the wind velocity.

The most remarkable feature in Fig. 2 is that a very high concentration of Mie particles was observed in the winter antarctic stratosphere. From just above the tropopause to 14 km, a concentration of up to 15 particles cm^{-3} was observed, whereas tropospheric concentrations were lower than 1 particle cm^{-3}.

From the profile of aerosol count ratio in Fig. 2, it can be seen that in the case of enhanced aerosol concentration, there is a difference in size distribution at different heights. Just above the tropopause, small Mie particles prevail but the prevalence of small particles tends to decrease with height so that the count ratio varied from a large value to one similar to that of the stratospheric background value observed at mid-latitude in the northern hemisphere (Hofmann and Rosen, 1981).

4.2. Mie particle sounding on 16 October in 1983

Fig. 3 shows the vertical profile of Mie particle concentration and aerosol count ratio of the two size ranges obtained on 16 October 1983. A very high aerosol concentration in the troposphere of up to 7 particles cm^{-3} was mainly contributed to by small Mie particles as can be seen from the profile of the count ratio. The aerosol concentration in the stratosphere was higher than that reported previously by Hofmann et al. (1973). The count ratio in the stratosphere was similar to the background value observed over mid-latitude in the northern hemisphere (Hofmann and Rosen, 1981).

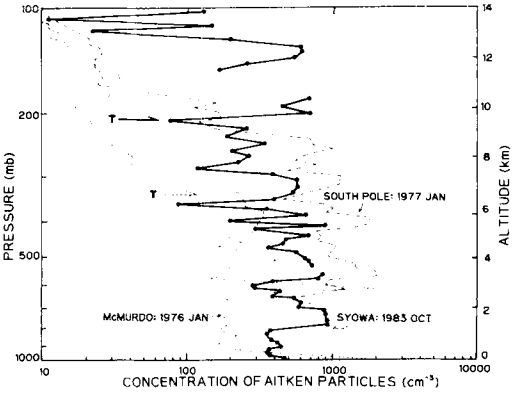

Fig. 4. Vertical profile of the Aitken particle concentration over Syowa Station on 17 October 1983, compared to previous results of Hofmann et al. (1976, 1977). "T" indicates the tropopause.

4.3. Aitken particle sounding on 17 October in 1983

In Fig. 4, the vertical profile of Aitken particle concentration obtained by the sounding on 17 October in 1983 is shown by solid lines with circles. It can be seen that the vertical variation

of the particle concentration measured on 17 October was quite systematic and random fluctuations of individual measurements were rather infrequent. This fact seems to indicate that the whole system of the sonde including the automatic Aitken counter worked properly. The main results can be summarized as follows: a high concentration of Aitken particles of about 1000 particles cm^{-3} was seen around 750 mb, 600 mb, 300 mb and above 200 mb, whereas the surface value was a few hundred particles cm^{-3} only. The maximum mixing ratio is seen in the stratosphere just above the tropopause.

In Fig. 4, the previous results obtained at McMurdo in January 1976 and at South Pole in January 1977 by Hofmann et al. (1976, 1977) are shown by dashed lines. It can be seen that the results at McMurdo, South Pole, and Syowa give a similar vertical profile of particle concentrations in the troposphere. The tropospheric average concentration in the present result is comparable with this in previous studies. The layers with high particle concentrations in the troposphere are also seen in these previous results. A similar high concentration layer has also been found in the aircraft observation by Hogan (1979). Thus the layers with high concentrations of Aitken particles seen in the present result seem to be common in the antarctic troposphere. On the other hand, the observed high concentration of Aitken particles in the stratosphere is peculiar to the present result as compared with previous results by Hofmann et al. (1976, 1977).

5. Discussion

5.1. *Enhancement of antarctic stratospheric aerosols in winter*

There are many papers reporting that the eruption of Volcano El Chichon in Mexico in April 1982 injected a large amount of sulfur-bearing gases into the stratosphere, and massive sulfuric acid aerosols developed and spread world-wide in the stratosphere. On the other hand, apart from the volcanic effect, the SAM II satellite system has sighted a stratospheric phenomenon which suggests that the antarctic stratospheric aerosol mass shows a regular seasonal variation having a remarkable winter maximum (McCormick, 1985). The possible explanation of this phenomenon (the so-called "polar stratospheric cloud or PSC") has been given by the condensational growth of pre-existing particles caused by the extreme temperature drop in the winter stratosphere (Steele et al., 1983).

According to the lidar observation at Syowa Station from April to October 1983, the integrated back-scatter ratio, which is a measure of the column mass of aerosols, showed a marked seasonal variation having a maximum in winter (Iwasaka et al., 1985a); the magnitude of the winter maximum was greater than the value observed in Japan during the enhancement of the stratospheric aerosol caused by the effect of the eruption of El Chichon. The aerosol enhancement on 3 June was observed in a very cold stratosphere, similar to the condition where SAM II sighted the PSC. Thus, the result of the sounding on 3 June gives direct evidence of a winter enhancement of aerosol concentration in the antarctic stratosphere. It is worthwhile noting that the enhancement of aerosol concentrations on 3 June is characterized by a tremendous increase in the number of small Mie particles. For example at 14 km, the concentration of particles larger than 0.3 μm in diameter is about 15 particles cm^{-3}, whereas that of particles larger than 0.5 μm is less than 2 particles cm^{-3}. According to Steele et al. (1983), the best agreement for the theoretical curves to show the relation between extinction by the PSC and stratospheric temperature with the SAM II data shows a total particle concentration of 6–7 particles cm^{-3} at 100 mb (about 15 km). Even the concentration of particles larger than 0.3 μm is much higher than this total concentration. It is suggested that the observed high concentration is partly attributed to the effect of the El Chichon eruption.

5.2. *Volcanic effect on antarctic stratospheric aerosols*

According to Hofmann et al. (1973), the background concentration of stratospheric Mie particles over Antarctica is about 1 particle cm^{-3} during the period of no volcanic effect. The sounding on 16 October 1983 reveals that Mie particles are still present in the lower stratosphere with a concentration higher than this background

concentration. The Aitken particle sounding on 17 October shows the presence of a high concentration of Aitken particles, more than 1000 particles cm^{-3} in the lower stratosphere, giving quite a different figure compared with the previous results obtained by Hofmann et al. (1976, 1977).

The high concentration of stratospheric aerosols observed on 16 and 17 October may give an indication that the effect of the eruption of El Chichon still remained in the antarctic stratosphere about 15 months after the eruption. Recently the result of a sounding conducted at McMurdo, West Antarctica was reported by Hofmann and Rosen (1985). The sounding was made on 27 October, 10 days after the soundings at Syowa Station, East Antarctica. They also observed a high concentration of up to 7 particles cm^{-3} of Mie particles in the lower stratosphere, whereas the number of condensation nuclei (which is equivalent to our Aitken particles) was as low as about 20 particles cm^{-3} in the lower stratosphere, although a remarkably high concentration was observed above 20 km in height. They also recognized the presence of the volcanic effect in the Antarctic stratosphere at that time. The observed differences in the particle concentration over McMurdo Station and over Syowa Station may indicate that the stratospheric aerosol was not distributed uniformly but exhibited large spatial and day-to-day variations. The stratospheric backscatter observed by lidar also exhibited large day-to-day variation over Syowa Station (Iwasaka et al., 1985a).

Aitken particles do not have a long residence time in the atmosphere in the presence of a large number of Mie particles, due to their coagulation with large particles. Thus the high Aitken particle concentration as observed on 17 October is considered to indicate the possible production of Aitken particles in the antarctic stratosphere. It is hard to believe, however, that SO_2 from El Chichon, which is oxidized to produce a sulfate aerosol, could survive more than 15 months after the eruption to be present with a significant amount in the antarctic stratosphere. It seems reasonable to suppose that the high concentration of Aitken particles in the stratosphere detected by the sounding on 17 October is probably attributed to the re-nucleation of sulfuric acid vapor produced by the evaporation of large particles. The drastic temperature change and large upward and downward motion in the spring antarctic stratosphere may play some rôle in these aerosol processes.

5.3. Tropospheric aerosols over Antarctica

It has been well established by a series of investigations by Cadle et al. (1968), Maenhaut et al. (1979), Parungo et al. (1981), and Delmas et al. (1982) that the antarctic tropospheric aerosol is dominated by sulfate particles. As to the chemical form of these sulfate particles, a decisive conclusion has not been obtained. Gras (1983) showed that sulfate aerosol at South Pole was nearly fully neutralized by ammonium. The electron-micrograph of antarctic aerosol particles shown by Bigg (1980) seems to support Gras's finding, in which particles having a morphology peculiar to sulfuric acid particles (Frank and Lodge, 1967) could not be seen. On the other hand, Cadle (1968), Meszaros and Vissy (1974), Ono et al. (1981), and Iwasaka et al. (1985b), showed the presence of sulfuric acid in antarctic aerosols by electronmicroscopy of aerosol particles. Furthermore, Delmas et al. (1982) showed that excess sulfate in the antarctic snow, which comes from sulfate aerosol, is most generally in the form of sulfuric acid.

From the extensive examination of seasonal and day-to-day variation of the concentration, size distribution and composition of antarctic aerosols, Ito (1983, 1985) concluded that there are two types of sulfates of different origin in antarctic aerosols. One is aged sulfates other than sulfuric acid, which prevails in winter. The other is sulfuric acid produced within the antarctic troposphere which prevails in summer. The conclusion by Ito (1983, 1985) seems to combine inclusively the inconsistent observational results given by Gras (1983) and Delmas et al. (1982), which show a discrepancy with each other on the chemical form of the antarctic sulfate. Aerosol samples taken during a rather short time like those of Bigg (1980), and Gras (1983), may tend to show a relatively uniform chemical form coming from a single origin.

The knowledge on the route through which sulfate particles or sulfur bearing gases would be transported into Antarctica is also far from determined. The volcanic component detected in

antarctic snow by Delmas and Boutron (1980) could be transported through the stratosphere, to be sure, as demonstrated by the soundings on 16 and 17 October, but their intrusion into the antarctic troposphere from the stratosphere does not necessarily occur over Antarctica. Cadle et al. (1968) inferred that the sulfuric acid droplets observed at McMurdo Station originated in the stratospheric Junge layer. They also described that one way to test their inference was to confirm the high concentration of sulfuric acid particles in winter when downward mixing is the greatest among all seasons. According to Ito (1983), at Syowa Station sulfuric acid droplets existed in summer but did not exist in winter, contradicting Cadle's inference. As shown in Fig. 2, even in winter, the large discontinuity of the Mie particle concentration in the vertical profile was observed at the tropopause, suggesting only weak coupling between the troposphere and the stratosphere over Antarctica. A similar discontinuity in the aerosol concentration at the tropopause was commonly seen in the soundings at Syowa Station on 16 and 17 October and soundings made by Hofmann et al. (1973, 1976, 1977). Thus, the coupling between the troposphere and the stratosphere over Antarctica seems to be weak and air mixing between troposphere and stratosphere, if any, would not be continuous but intermittent over Antarctica. As the most plausible explanation, it can be said that the stratospheric component of sulfate as detected by Cadle et al. (1968) and Maenhaut et al. (1979) and also the volcanic component as detected by Delmas and Boutron (1980) would be mixed into the troposphere through a tropopause gap at mid-latitude or by the dynamical effect of the strong cyclone around the subantarctic region (Kida, 1983) and thereafter transported into the interior of Antarctica through the meridional air exchange mainly caused by the migration of cyclones into antarctica.

According to Delmas and Boutron (1980), the main contribution to sulfate deposition in central Antarctica has been attributed to the oxidation of marine gaseous compounds. This sulfate produced from sulfur-bearing gases of marine origin has been thought to be produced in the atmosphere outside of Antarctica and transported into Antarctica in a form of particles (Bigg, 1980; Parungo et al., 1981; Hogan et al., 1982; Shaw, 1982). Ito et al. (1982) found, however, that the seasonal and latitudinal variation of the number concentration of Aitken particles has a strong relation to the variation in intensity of solar radiation in the antarctic atmosphere, suggesting the possible production of aerosol particles in the antarctic troposphere. A similar fact was also found by Bigg et al. (1984) for the seasonal variation of Aitken particle concentration over the southern hemisphere. The size distribution of Aitken particles observed at Syowa Station in the sunlit season showed a bimodal distribution, having a significant concentration in a size range smaller than 0.01 μm in radii (Ito, 1985). This also suggests the possible production of Aitken particles over Antarctica, contradicting the discussion given by Shaw (1982).

The sounding of Mie particles on 16 October showed a higher tropospheric concentration than the Mie particle sounding on 3 June. The Aitken particle concentration observed in the sounding on 17 October was higher than the annual mean concentration at Syowa Station (Ito, 1985). The sounding on 3 June was made in a northern wind regime, indicating the air in the troposphere to be essentially of maritime origin, whereas the soundings on 16 and 17 October were made in a southern wind regime indicating the air to be essentially of antarctic origin. Thus the aerosol soundings made at Syowa Station in 1983 also seem to suggest the possible production of aerosol particles in the Antarctic troposphere. It can be inferred that the sulfate particles produced and/or grown in the antarctic troposphere make up a significant portion of the number concentration of antarctic aerosol particles, and probably are even important to the mass concentration in the sunlit season.

6. Summary

Balloon soundings of aerosols up to 15 km in height were carried out in 1983 at Syowa Station (69°00′S, 39°35′E), Antarctica. Vertical distributions of the concentration and size distribution of Mie particles (diameter larger than 0.3 μm) were obtained on 3 June and on 16 October. The vertical distribution of the concentration of Aitken particles (diameter larger than 0.004 μm) was obtained on 17 October.

A very high concentration of Mie particles of up to 15 particles cm^{-3} was observed in the lower stratosphere in June. This is the first direct evidence of the winter aerosol enhancement in the antarctic stratosphere. The observed enhancement is characterized by a tremendous increase in a concentration of small Mie particles.

A concentration of Aitken particles as high as about 1000 particles cm^{-3} is observed in October around 750 mb, 600 mb, 300 mb, and above 200 mb, whereas the surface value is in the order of a few hundred particles cm^{-3}. The maximum mixing ratio is seen in the stratosphere just above the tropopause. From a comparison of the present result with previous results, it is concluded that the layer of high particle concentration in the troposphere seems to exist commonly over Antarctica, whereas the high concentration in the stratosphere is peculiar to the present result. The high concentration in the troposphere may be evidence for the production of new particles through the photochemical gas-to-particle conversion process in the antarctic troposphere. The high concentration of Aitken particles in the stratosphere, which was accompanied by the high concentration of Mie particles, may be attributed to the effect of the eruption of the volcano El Chichon which occurred about 15 months earlier.

A marked discontinuity in aerosol concentration at the tropopause was seen in a vertical profile of the concentration of Mie and Aitken particles, suggesting that the particle transport from the stratosphere to the troposphere over Antarctica is not significant compared with the horizontal transport of particles to the antarctic troposphere. The concentration in the winter troposphere for a northern wind regime was lower than that in the spring troposphere for a southern wind regime, suggesting the significance of particle production in the antarctic troposphere.

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