ENHANCEMENT OF ANTARCTIC STRATOSPHERIC AEROSOL LAYER IN WINTER: POSSIBLE CONTRIBUTION OF AITKEN PARTICLE GROWTH

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Abstract: The lidar measurements at Syowa Station (69°00′S, 39°35′E), Antarctica revealed that the content of stratospheric particles increased noticeably and that particle shape possibly was of ice crystal in winter. The increase in number of particles with large size, in addition to the deposition growth of ice particles, possibly contributes to the stratospheric particulate matter increase during winter.

1. Introduction

Recently MCCORMICK *et al.* (1982, 1985) showed that the content of polar stratospheric particulate matter increased corresponding to decrease in temperature on the basis of satellite (SAM II) measurements. Lidar measurement in Antarctica also revealed that the Antarctic stratospheric aerosol layer enhanced extremely in winter (IWASAKA, 1985; IWASAKA *et al.*, 1985a). These observations showed that the content of particulate matter increased under a very cold stratospheric condition, but we cannot know directly from these results whether the increase in particulate matter is due to an increase in individual particle mass and size or due to an increase in particle number.

From a viewpoint of global budget of stratospheric water vapor and sulfur, it is important to know a chemical composition of particles in the winter polar stratosphere.

The thermodynamical studies on the aerosol particles existing in the very cold polar stratosphere (STEELE *et al.*, 1983) suggested a possibility that active transformation of preexisting aerosol particles from sulfuric acid droplets to ice crystal particles occurred under a very cold condition (-80°C or colder) and that the growth of ice crystals through deposition was an essential factor controlling the increase in particulate matter in the winter polar stratosphere.

In 1983 the depolarization ratio of backscattering light from the stratosphere was measured using the lidar at Syowa Station (69°00′S, 39°35′E) as a clue to examine observationally the existence of nonspherical particles such as ice crystals in the polar winter stratosphere.

The purpose of this paper is to present the lidar measurements suggesting the existence of nonspherical particles in the Antarctic stratosphere and the contribution of spherical particle number increase to the enhancement. The importance of Aitken

particle growth in the early stage of the enhancement will be discussed to understand the winter enhancement of the polar stratospheric aerosol layer.

2. Lidar Measurements

In Table 1, the main characteristics of the lidar used here are summarized. The detailed specifications of this system were described elsewhere (IWASAKA *et al.*, 1985b) and the outline of it is given below.

Table 1. Main characteristics of the lidar system.

Transmitter		
Laser output		>1 J/pulse (694 nm)
		>0.3 J/pulse (347 nm)
Laser pulse length		40 ns
Repetition rate		60 ppm (max)
Laser beam divergence		1.6 mrad
Transmitter optics		Galilean telescope (×4)
Transmitter divergence		0.5 mrad
Receiver		
Optics		Cassegrain telescope
Diameter		50 cm
F number		F/4.0
Field of view		0.5–2.0 mrad
Transmitter/receiver mount		
Vertically detection only		
Detection system		
3-channel detection (typical configuration)		
A-channel	PMT R-943-02	347 nm±1.3 nm
B-channel	PMT R-943-02	694 nm±0.5 nm
C-channel	PMT R-1333	694 nm±0.5 nm
Signal processing		
Analog method		
A/D converter		8 bit resolution
Photon counting method		
Multichannel counter		8 bit resolution
Data processing		
CAMAC data logging system with minicomputer Melcom 70/10		

The lidar system consists of a 694 nm pulse ruby laser, a 50 cm ϕ telescope, dual 100 channels photon counters, an A-scope, an A/D converter with a maximum sampling rate of 20 MHz, and a minicomputer system for data processing. Lidar measurements on the polar stratosphere have been made since April 1983 at Syowa Station (69°00′S, 39°35′E), Antarctica, as part of the international project "Antarctic Middle Atmosphere (1982–1986)".

The scattering ratio, $R(Z)$, is defined as follows (*e.g.*, RUSSELL *et al.*, 1976);

$$R(Z)=[B_1(Z)+B_2(Z)]/B_1(Z), \tag{1}$$

where $B_1(Z)$ and $B_2(Z)$ are, respectively, the molecular and particulate backscattering coefficient at altitude Z. To obtain an aerosol backscatter coefficient from the lidar

return, "Matching method" (*e.g.*, RUSSELL *et al.*, 1976) was used here. The molecular backscatter coefficient was estimated on the basis of radiosonde measurements which were routinely made at Syowa Station. The mixing ratio of particulate matter is estimated as follows:

$$R(Z)-1=B_2(Z)/B_1(Z). \tag{2}$$

When we emit the linearly polarized laser light, the backscatter light from atmospheric molecules has a small amount of depolarization due to the anisotropic factor for air molecules. Aerosol particles also can produce some depolarization. The ice cloud particles and the volcanic ash particles loaded into the stratosphere through a severe volcanic eruption produce a large amount of depolarization in backscatter (*e.g.*, PAL and CARSWELL, 1976). Therefore, it must be useful to measure the depolarization ratio of backscatter light in order to detect the nonspherical particles such as ice crystals in the polar stratosphere.

The depolarization ratio of backscatter light is given by;

$$D(Z)=P_{\perp}(Z)/P_{/\!/}(Z), \tag{3}$$

where $P_{\perp}(Z)$ and $P_{/\!/}(Z)$ are, respectively, received backscattered powers in the polarization planes of perpendicular and parallel to the transmitted laser beam which is linearly polarized.

3. Results

The vertically integrated backscatter coefficient in the stratosphere is given by;

$$I=\int_{\Delta Z} B_2(Z)\,\mathrm{d}Z, \tag{4}$$

where ΔZ is the height range from bottom to top of the layer (we chose the tropopause in place of the layer bottom when the tropospheric clouds disturbed a clean level region

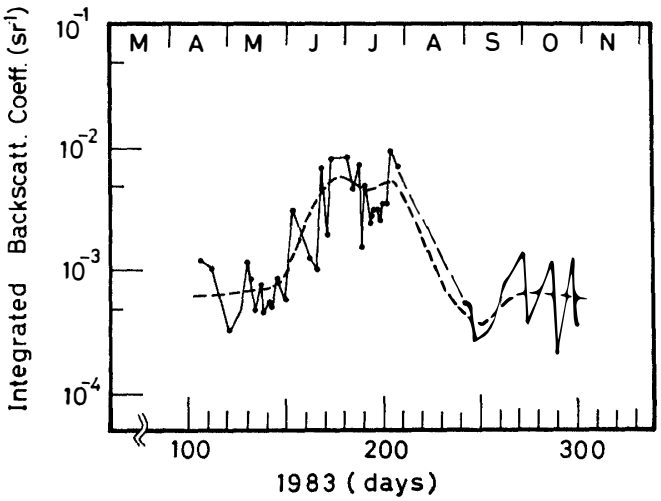

Fig. 1. Integrated backscattering coefficient of particles in the stratosphere measured at Syowa Station (69°00'S, 39°35'E) in 1983.

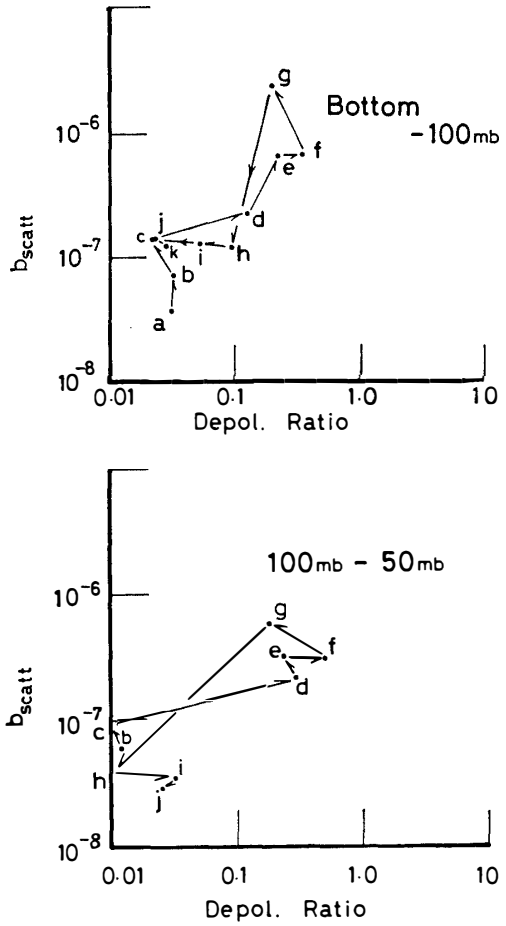

Fig. 2. Correspondence of depolarization ratio (depol. ratio) to backscatter coefficient (b_{scatt}). Measurements in the range from the layer bottom to 100 mb height are summarized in the upper figure and those from 100 mb height to 50 mb height in the lower. Points in figure are half-monthly averaged values as follows;

a: April 16–30, 1983
b: May 1–15, 1983
c: May 16–31, 1983
d: June 1–15, 1983
e: June 16–30, 1983
f: July 1–15, 1983
g: July 16–31, 1983
h: September 1–15, 1983
i: September 16–30, 1983
j: October 1–15, 1983
k: October 16–31, 1983.

near the tropopause). It can be recognized as a parameter corresponding to the column mass concentration of particulate matter (NORTHAM *et al.*, 1974; HOFMANN *et al.*, 1983). In Fig. 1 the change of "I" is shown. The integrated value showed its maxima in winter, 1983. A simillar event had been also found by the satellite SAM II (MCCORMICK *et al.*, 1982, 1985). Therefore, the enhancement of the Antractic stratospheric aerosol layer is not unusual but may be a usual phenomenon seen in winter. Here we call it winter enhancement of the Antarctic stratospheric aerosol layer.

The depolarization ratio *vs.* backscattering coefficient is shown in Fig. 2.

4. Discussion

4.1. Particulate matter content and depolarization ratio in winter

The result shown in Fig. 1 suggests an increase in the content of particulate matter in the winter stratosphere, and confirms the results obtained by satellite SAM II measurements (MCCORMICK *et al.*, 1982, 1985). The results in Fig. 2 suggest that the depolarization ratio also increased to noticeably large values in winter. Such large values, as far as we know, have never been measured in the low and middle latitude stratosphere. Even when many volcanic ash particles were injected into the strato-

sphere by the eruption of the El Chichón volcano, the depolarization ratio of the stratospheric aerosol layer measured at Nagoya was at most about 0.3 (HAYASHIDA *et al.*, 1984). The ice crystal cloud such as cirrus shows frequently the values larger than 0.5.

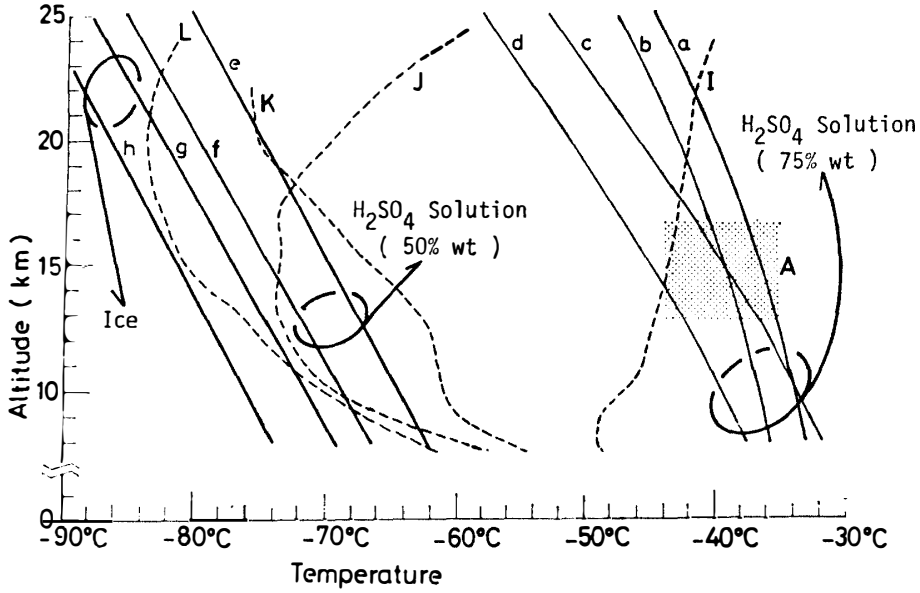

Fig. 3. Comparison of atmospheric temperatures and saturation (or frost) point temperatures of water vapor and sulfuric acid vapor. Curves I, J, K, and L are the atmospheric temperatures which are half-monthly averaged on the basis of the routine radiosonde measurements at Syowa Station (69°00′S, 39°35′E), and show typical feature of each season; I: February 1–15, 1983 (summer), J: September 16–30, 1983 (spring), K: May 16–31, 1983 (fall), L: July 1–15, 1983 (winter).
Curves "a" and "b" are saturation point temperatures of sulfuric acid vapors with 0.2 and 0.1 pptv mixing ratio for sulfuric acid solution with 75% of weight.
Curves "c" and "d" are saturation point temperatures of water vapor of 10 and 5 ppmv, respectively, for sulfuric acid solution of 75% weight, and curves "e" and "f" are for sulfuric acid solution of 50%.
Curves "g" and "h" are frost point temperatures of water vapor with mixing ratio 10 and 5 ppmv, respectively. In winter the atmospheric temperature falls down to the value lower than the frost point temperature, h.
Shaded area "A" indicates the region in which typical stratospheric aerosol particles of about 75% sulfuric acid droplets can exist.

Frost point temperatures are plotted in Fig. 3, and the water vapor pressure on the sulfuric acid solution and the sulfuric acid vapor pressures are compared on the basis of the results by GMITRO and VERMEULEN (1964). The atmospheric temperature profiles are illustrated in the figure to show the characteristics of each season on the basis of the routine radiosonde sounding at Syowa Station. The atmospheric temperature of curve "L" is lower than the frost point temperature "g" for water vapor mixing ratio 10 ppmv, and is comparable with that for about 6 ppmv. In summer, the stratospheric temperature "I" is found in the range where sulfuric acid droplets with 75% of weight can be in equilibrium with the surrounding atmosphere.

STEELE *et al.* (1983) presented the assumption that the increase in particulate matter

content was due to the increase in mass (or size) of individual ice crystals which were formed preexisting sulfuric acid droplets through the dilution caused by temperature decrease. Concerning the presence of the ice crystals, the lidar measurements appear to confirm the assumption by STEELE *et al.* (1983). As described later, according to the lidar measurements and balloon measurements, the possible increase in particle number density is important for the increase in particulate matter content in the polar stratosphere.

4.2. Aerosol content increase in the early phase of winter enhancement

It should be noticed that the lidar measurements showed a large scattering ratio but not a large depolarization ratio in early winter. Typical samples are compared in Fig. 4. The scattering ratio of June 2 in Fig. 4 show a large mixing ratio of aerosol particles (about 30–50 times larger than the levels measured during the quiet period in mid-latitudes). However, the depolarization ratio is not so large as to define the existence of nonspherical particles. Fortunately the size distribution and the number density of large particles with sizes larger than 0.15 μm were observed by a balloon-borne particle counter at Syowa Station on June 3 (MORITA *et al.*, 1984). These results revealed that the number density of particles with radius >0.15 μm was noticeable large, about 15 particles/cm^3, above the tropopause. Previous observations made

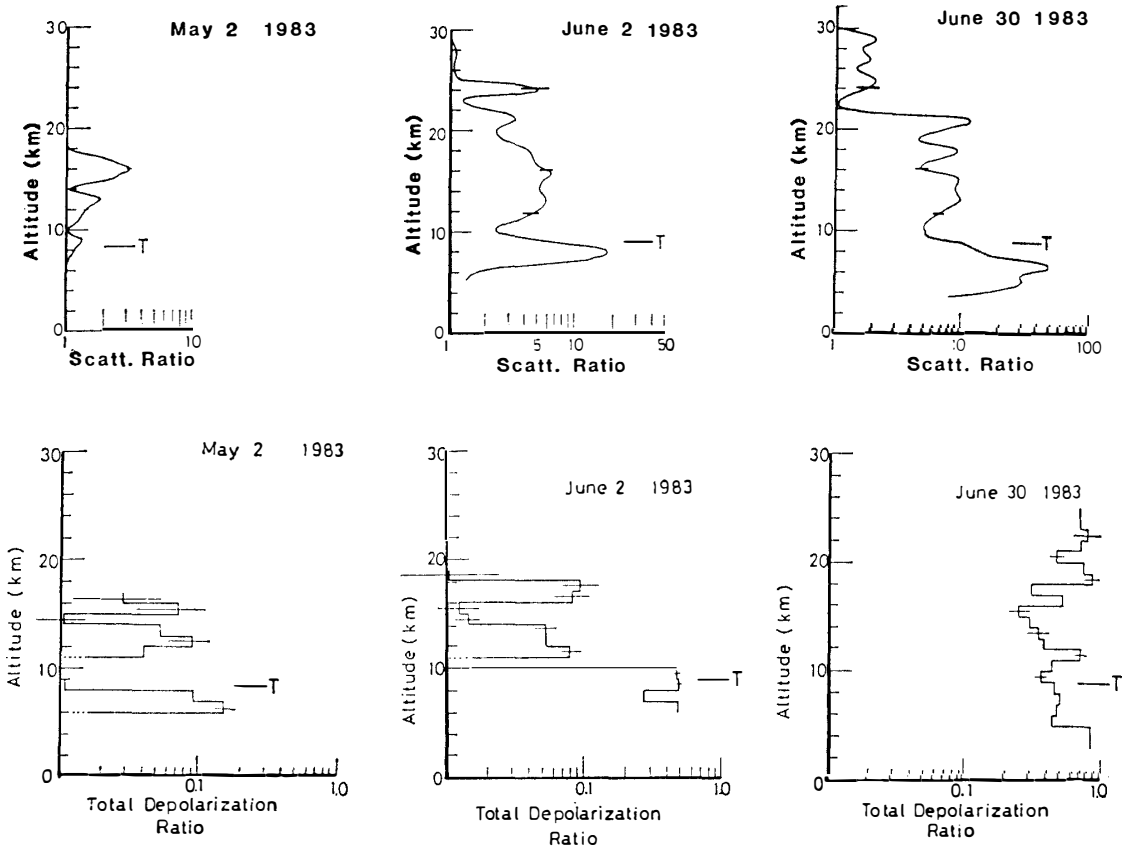

Fig. 4. Typical measurements of scattering ratio and depolarization ratio at Syowa Station. The measurements on May 2, June 2, and June 30, 1983 show the typical feature of pre-enhancement stage, early enhancement, and fully developed enhancement, respectively.

Table 2. Comparison of lidar and balloon measurements at Syowa Station (69°00'S, 39°35'E), 1983.

	April–May	June 1–14	June–August	September–October
Lidar				
Int. backsc. coeff. (sr^{-1})	6×10^{-4}	2×10^{-3}	5×10^{-3}	5×10^{-4}
Backs. coeff. (m^{-1} sr^{-1})	5.2×10^{-8}	1.4×10^{-7}	3.3×10^{-7}	3.3×10^{-7}
*Estimated				
particle a	6.1	16.4	38.8	3.8
density (cm^{-3}) b	11.5	28.8	68.4	6.8
Atmospheric				
temp. (°C) 100 mb	−60.5	−73.5	−76.7	−73.2
50 mb	−65.2	−78.1	−81.7	−70.7
Balloon				
Particle				
density (cm^{-3})		15–20		2–3
Density ratio		8–50		7
		**(June 3, 1983)		(Oct. 16, 1983)

* Particle densities are estimated assuming density ratio 3.5 for "a" and 12 "b". The size distribution function of log-normal is used.

** Balloon did not reach the peak height of the aerosol layer.

Model calculation: Particle number is estimated for various H_2O mixing ratio under the condition that size distribution is log-normal and total particle density fixed (STEELE *et al.*, 1983).

Best fit for SAM II data

Total particle density, 6–7 cm^{-3}

Large particle, $R>0.10$ μm, density, 0.14–0.16 cm^{-3}

Density ratio, $N(R>0.15\ \mu m)/N(r>0.25\ \mu m)$, 4.68–6.14

Balloon measurement: At McMurdo Station during summer (HOFMANN and ROSEN, 1984).

Large particle density, $R>0.15$ μm, 7 cm^{-3} at layer peak (Oct. 27, 1983).

[High CN concentration was found above the layer peak (about 100 cm^{-3}).]

mainly in summer showed that the stratospheric aerosol number concentration was about at most 1 particles/cm^3 during the calm stratospheric period (*e.g.*, ROSEN *et al.*, 1975) and was 7 particles/cm^3 during the disturbed period (HOFMANN and ROSEN, 1984). In Table 2 we compared the measurements at Syowa Station with the values by HOFMANN and ROSEN (1984), and by STEELE *et al.* (1983). It should be noticed that the duration when we observed the event having a high density particle content and a low depolarization ratio is very short, at most half month. Therefore, it is necessary to take continuous measurement in order to catch this event. Previous measurements, concerning this, were far from satisfactory.

The averaged backscatter coefficient is about 1.4×10^{-7} m^{-1} sr^{-1} in the early stage. This value corresponds to about 50 particles/cm^3 with $R>0.15$ μm for the particle density ratio measured on June 3 using the estimation by SWISSLER *et al.* (1982). The measurements by MORITA *et al.* (1984) showed about 15 particles/cm^3 near the layer bottom, where a large concentration than that in the layer center can be expected. Considering the difference in measurement time (about 10 h) and unknown details

of size distribution and composition, we can conclude that the lidar data and the balloon data are in good agreement with each other.

The important point here is that the increase of particulate matter in the early stage can be due to the increase of large particle number density.

One possible interpretation for the characteristics found in the early stage of the enhancement is that many large water droplets were formed from Aitken size particles corresponding to the decrease of the stratospheric temperature. To clarify the problem, it is necessary to know size distribution, chemical composition, and activity spectra of Aitken particles. However, there are few data about Aitken particles in the polar stratosphere, and we cannot give a definite conclusion.

In 1982 there was found a severe disturbance by the eruption of El Chichón in the stratosphere on a global scale (*e.g.*, MCCORMICK *et al.*, 1985). The volcanic eruption disturbed also the Antarctic stratospheric aerosol layer (HOFMANN and ROSEN, 1984). Therefore, there is a possibility that many Aitken particles acting as condensation nuclei and ice nuclei had been transported into the Antarctic stratosphere by the early winter in 1983 and caused the noticeable increase in density of large particles under a very cold stratospheric condition.

5. Summary and Conclusion

Summarizing the measurements at Syowa Station (69°00′S, 39°35′E), we can conclude that formation of nonspherical particles, possibly ice crystals, is very active and that the results confirm the ice crystal growing model in the polar stratosphere presented by STEELE *et al.* (1983). Additionally it should be noticed that many nonspherical particles were observed in the early winter and they contribute to the increase of the particulate matter content.

To understand physical processes controlling the winter enhancement of the polar stratospheric aerosol it should be essential to obtain information about size distribution of aerosol particles, nature of particles, content of related gases (H_2O, H_2SO_4, and so on), and their detailed temporal changes.

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