DISTRIBUTION OF PARTICULATE SULFUR IN THE WINTER ARCTIC STRATOSPHERE: BALLOON-BORNE MEASUREMENTS OF PARTICLE NUMBER CONCENTRATION

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Abstract: Balloon-borne measurements of the size-number concentration of atmospheric particles were made in December 1994 and January 1995 at Ny-Ålesund, Norway (79°N, 12°E). The measurements showed that the number concentration of stratospheric particles was 2 particles/cm^3 (particle diameter $\geq$ 0.3 μm) and 5×10^{-2} particles/cm^3 (particle diameter $\geq$ 1.2 μm) at the potential temperature height of 370 K (near the height of the background aerosol layer peak) on December 18, 1994, and 2 particles/cm^3 (particle diameter $\geq$ 0.3 μm) and 2×10^{-3} particles/cm^3 (particle diameter $\geq$ 1.2 μm) at the potential temperature height of 370 K on January 17, 1995. The number-size distribution function of the background sulfate particles showed noticeable decrease of particle concentration in the size range of 0.5–0.8 μm in diameter in the heights of 350 K–370 K potential temperature from December 1994 to January 1995, suggesting that sulfate particles growing through up-take of water vapor descended in the winter polar stratosphere.

The winter polar stratosphere functions as one possible sink in the global budget of atmospheric sulfur.

1. Introduction

Recently, the global geochemical cycle of atmospheric sulfur has become a great concern since the sulfur cycle has been recognized as one of the important factors controlling global climate change (Lamb, 1970; Baldwin *et al.*, 1978; Turco *et al.*, 1980, 1982; Houghton *et al.*, 1996).

Severe volcanic eruptions largely disturbed stratospheric aerosols through gas-to-particle conversion of volcanic sulfur dioxide gas injected into the stratosphere. Many investigators suggested that global surface temperature meaningfully decreased owing to radiative forcing of the enhanced stratospheric aerosols following the eruptions of Krakatoa of 1883, Agung of 1963, El Chichón of 1983, and Pinatubo of 1991 (Hansen *et al.*, 1978; Turco *et al.*, 1982; Pollack and Ackerman, 1983; Angell and Korshover, 1985; Hansen *et al.*, 1992; Minnis *et al.*, 1993). Noticeable global depletion of stratospheric

ozone was observed in 1992 and 1993, and the effect of Pinatubo volcanic eruption on ozone chemistry is speculated to have been the most important process causing the ozone loss through heterogeneous reaction including volcanic aerosol particles (PRATHER, 1992; BOJKOV et al., 1993; CHANDRA, 1993; CLEASON et al., 1993; FAHEY et al., 1990; SCHOEBERL et al., 1993; HOFMANN et al., 1994; KOMHYR et al., 1994; McGEE et al., 1994; PLANET et al., 1994; RODRIGUEZ et al., 1994; WMO, 1995).

Those investigations suggested that transport, source and sink, and chemical transformation of volcanic sulfur should be observed much more to obtain better understanding of the volcanic effect on climate.

From field measurements in the polar region, many investigators have suggested that the polar stratosphere functions as a possible sink of stratospheric material (STANFORD, 1973; DELMAS et al., 1982; McCORMICK et al., 1983; KENT et al., 1985; IWASAKA, 1986a, b, c; IWASAKA and HAYASHI, 1991). The behavior of atmospheric particles in the polar stratosphere and diffusion of chemical constituents from the equatorial to the polar region are regarded as important processes affecting the sink of stratospheric material in the polar region (GOLOMBEK and PRINN, 1993).

However, there have been few measurements of number-size and/or mass-size distributions in the polar stratosphere, possibly owing to technical difficulty, and therefore it has been hardly possible to discuss quantitatively the sink of particulate material and related gases in the polar stratosphere.

We describe balloon-borne measurements of polar stratospheric aerosol particles made at Ny-Ålesund, Norway (79°N, 12°E) in the winter of 1994/1995 and discuss changes in the distribution of density of atmospheric particulate sulfate in the polar stratosphere.

2. Observations of Particle Size and Number Concentration

Atmospheric particle size and number concentration were observed with the balloon-borne particle counter at Ny-Ålesund, Norway (79°N, 12°E) on December 14 and 18, 1994 and January 17, 1995 (Fig. 1).

The main specification of the balloon-borne optical particle counter (OPC) used here is summarized in Table 1. This particle counter has been developed by the atmospheric aerosol research group of Solar Terrestrial Environmental Laboratory, Nagoya University and the engineering group of Sigmatec Co. Ltd. Details of the counter have been described by TSUCHIYA et al. (1996).

Particle size discrimination, as shown in Fig. 1, is made at 0.3, 0.5, 0.8, 1.2, and 3.6 μm (in diameter) on the basis of laboratory experiments for the refractive index of particulate matter, $n = 1.4 + 0\ i$ (for sulfuric acid droplet of about 70 wt%, which is referred from the calculated values of STEELE and HAMILL (1981), and observational values of BAUMGARDNER et al. (1996) in the stratosphere). When we treat other aerosol materials which have different refractive indices, the sizing range of the counter will be easily modified.

For one sampling volume of about 1000 cm^3, the statistical uncertainty in counting is about 3, 10, 32, 100% for particle concentrations of 1, 0.1, 0.01, and 0.001 cm^{-3}, respectively.

In the measurements of December 18, polar stratospheric clouds (PSCs) was found around 18 km in altitude. As described later, the layer of PSCs possibly consisted of nitric acid tri-hydrate (NAT) particles.
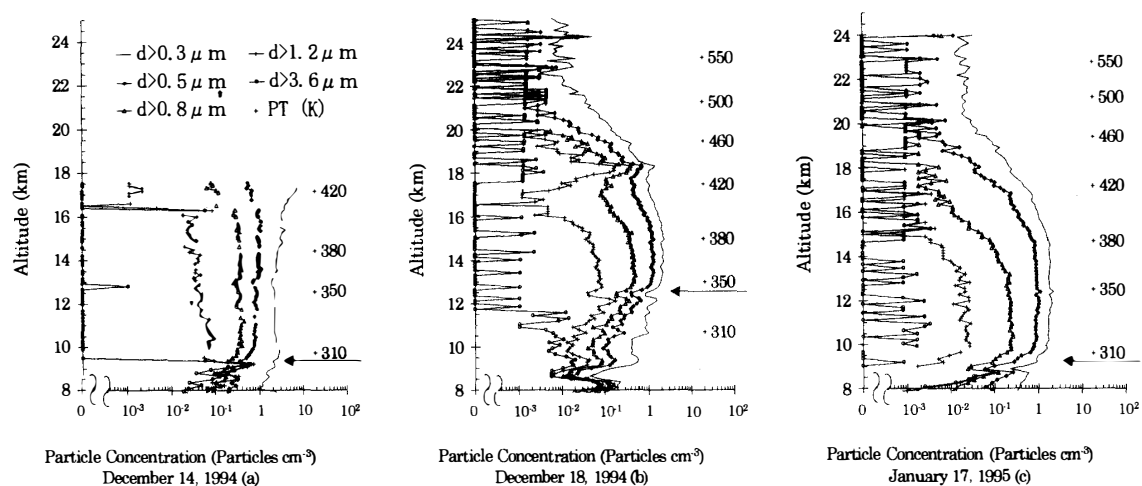
Fig. 1. Vertical profiles of stratospheric concentration of particles with diameter ≥ 0.3, 0.5, 0.8, 1.2, and 3.6 μm measured with a balloon-borne particle counter at Ny-Ålesund, Norway $(79°N, 12°E)$. The right-side axis shows heights of isentropic surfaces (K). Local tropopauses are indicated by arrows.
(a) December 14, 1994, (b) December 18, 1994, (c) January 17, 1995.

Table 1. Specifications of optical particle counter.

Physical dimensions:	$30 \times 35 \times 35$ cm
Weight:	5.5 kg (including battery)
Wave length of source light:	$\lambda = 810$ nm (laser-diode)
Detection:	Photodiode detector; forward-scattered light ($13°$-$44°$ from the beam axis)
Air sampling flow:	50 cm^3/s or 3 l/min (Under the standard atmospheric condition)
Sampling-time interval:	20 s for 1st and 2nd launched OPC. 24 s for the 3rd.
Data transmission wave band:	400 MHz (Vaisala radio-sonde)

All three observations indicate the existence of a "stratospheric background sulfate layer", about between 10 and 20 km. The volume density of the stratospheric background sulfate aerosols is discussed below.

From the observed particle number-size distributions, the volume density of the background sulfate particles (10 km–17 km; 230 hPa–70 hPa) was estimated without the particles with $d > 3.6$ μm since the concentration of particles with $d > 3.6$ μm was low and the relative error of concentration high, as mentioned above (Fig. 2). Figure 2 indicates that less volume density was observed in January than in December. Temperature distributions on the dates of OPC balloon soundings are shown in Fig. 3. The frost point temperatures of nitric acid vapor and water vapor on the surface of nitric acid tri-hydrate (NAT) and water vapor on the surface of ice are estimated, under the assumed atmospheric condition that the mixing ratios of HNO$_3$ and H$_2$O are 10, 5, 1 ppbv and 5, 3 ppmv respectively, on the basis of formulas given by HANSON and MAUERSBERGER (1988). These values are referred from observational values of water vapor (*e.g.* KELLY *et al.*, 1990;
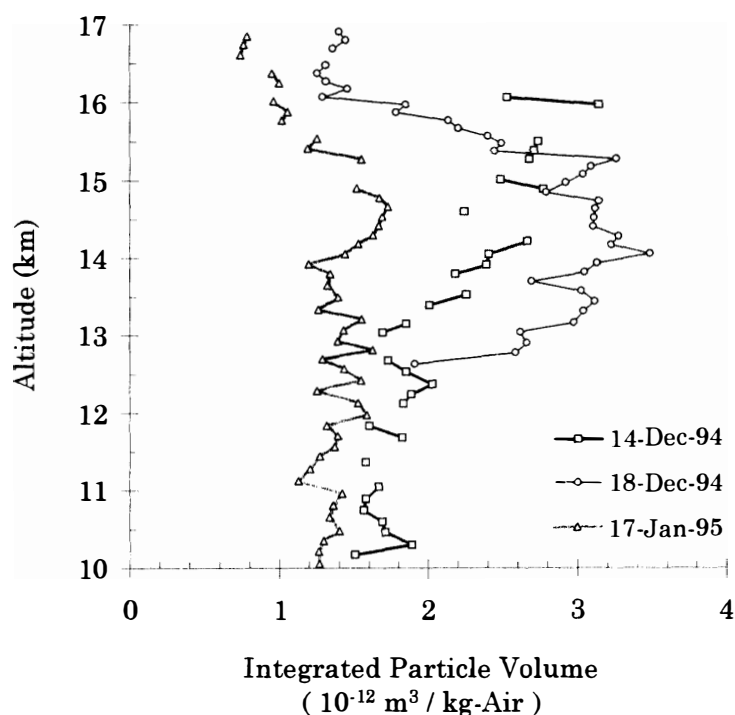
Fig. 2.   *Particle volume per unit mass of air (10⁻¹² m³/kg-Air), integrated in the size range between 0.3 and 3.6 μm in diameter.*
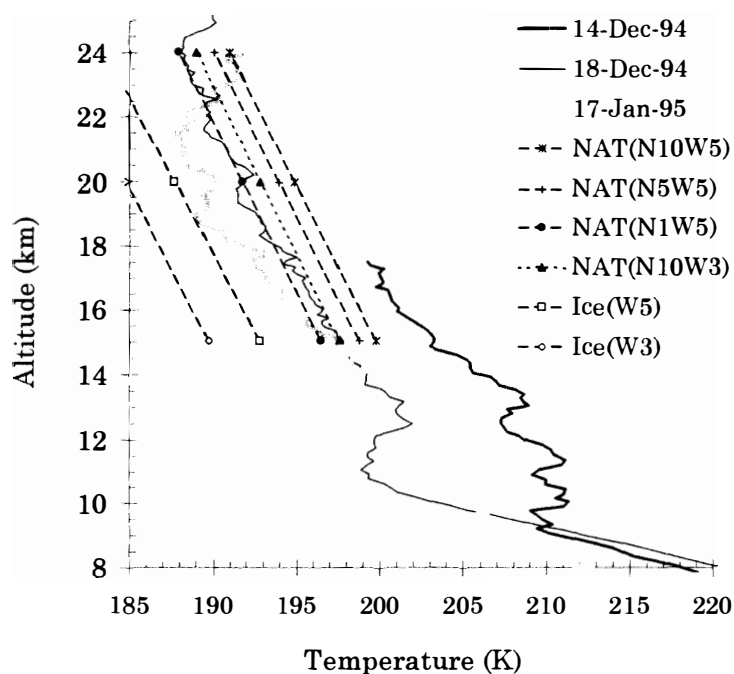

Fig. 3.   *Vertical profiles of stratospheric temperature measured at Ny-Ålesund, Norway (79° N, 12° E) during the measurements of aerosol concentration.   "NAT" and "Ice" mean the frost point temperatures of nitric acid trihydrate particle and ice crystal particle, respectively.   Assumed mixing ratios are indicated by (NxWy), which means HNO₃ mixing ratio of x ppbv and water vapor of y ppmv.*

Pruvost *et al.*, 1993; Ovarlez and Overlez, 1994), and of nitric acid (*e.g.* Clarmann *et al.*, 1993; Oelhaf *et al.*, 1994; Murcray *et al.*, 1994; Höpfner *et al.*, 1996).

In Fig. 3, the temperature of December 14, 1994 was highest in the three measurements in the altitude region of 10–17.5 km, and higher than the frost point of NAT above 16 km where there was large enhancement in concentrations of particles with diameter between 0.3 and 0.5 μm. A cold layer where NAT particles can form was found in the region of 15–24 km in the measurements of December 18, 1994 where the enhanced layer (in all size-range of detection; larger than 0.3 μm) was detected in some part. On January 17, 1995, large enhancement of aerosol was not detected in those altitude ranges, even though the temperature was lower than on December 18, 1994.

3. Discussion

Many investigations have suggested that stratospheric constituents were transported to the troposphere through diabatic descending motion of the polar stratospheric air-mass inside the polar vortex (*e.g.*, Kent *et al.*, 1985; Loewenstein *et al.*, 1990; Toon *et al.*, 1992; Bauer *et al.*, 1994). Additionally sedimentation of individual particles which grow to large size particles in the cold winter polar stratosphere was suggested (*e.g.*, Stanford, 1973, 1977; Iwasaka, 1986a, b, c; Iwasaka and Hayashi, 1990, 1991).

Descending motion of ice crystal particles grown to super micron size in the Antarctic stratosphere was suggested 25 years ago as a possible process controlling the global budget of stratospheric water vapor by Stanford (1973). In the 1980's, formation processes of polar stratospheric clouds particles were studied and various types of PSCs both in the Arctic and in the Antarctic are now presented. Large denitrification of the polar winter stratosphere in both hemispheres has been suggested by many investigators since PSCs grow through the uptake of nitric acid vapor on its surface (*e.g.*, Fahey *et al.*, 1990; Arnold *et al.*, 1992; Höpfner *et al.*, 1996). On the basis of lidar measurements at Syowa Station, Antarctica (Iwasaka and Hayashi, 1990, 1991), PSC particles descending from the stratosphere to the troposphere have been suggested as a possible mechanism of stratospheric nitrate transport from the stratosphere to the troposphere in the winter polar region.

Previously, there were few observations of atmospheric particle size in the winter polar troposphere and stratosphere. Therefore, discussion of possibility of descending of individual particles has been speculative.

Some investigators have suggested that condensation of HNO_3 and H_2O gas on particles is activated under the cold atmospheric condition, and binary-(H_2SO_4 and H_2O) and ternary-(H_2SO_4, HNO_3, and H_2O) solution particles are formed in the polar winter stratosphere (*e.g.*, Steele and Hamill, 1981; Jaecker-Voirol *et al.*, 1990; Tabazadeh *et al.*, 1994; Luo *et al.*, 1995). From Fig. 3, for example at 14 km in altitude, temperatures on December 18 and January 17 were lower than that on December 14. According to these solution models, volume density at 14 km of December 18 and January 17 must be higher than that on December 14 at 14 km. However Fig. 2 indicates a lower volume density on January 17 than that on December 14 and 18. This inconsistency suggests the possible existence of an additional factor controlling sulfate mass distribution in the winter polar stratosphere.

In Fig. 4, the particulate sulfuric acid mixing ratio was estimated on the basis of

volume densities in Fig. 2, according to the equilibrium model of TABAZADEH *et al*. (1994) (with the temperature in Fig. 3 and assumed mixing ratios of HNO_3 and H_2O, of 10 ppbv and 5 ppmv, respectively), between 10 and 17 km in altitude. Figure 4 shows that the two observational results in December are highly consistent, but the amount of particulate sulfuric acid on January 17 was less than those in December.

In Fig. 5, we show stratospheric temperature obtained from radio-sonde soundings at Ny-Ålesund in the winter of 1994-95. Meteorological conditions in the stratosphere above Ny-Ålesund, from the temperature distributions from December 1, 1994 to March 13, 1995, seemed to be relatively stable during OPC sonde soundings.

According to WILSON *et al*. (1992), vertical distributions of particulate sulfate in the polar winter stratosphere showed apparently less mixing ratio inside the polar vortex than outside. They explained this difference as air-mass subsidence owing to diabatic cooling during winter inside the polar vortex. But as we showed in Fig. 4, the mixing ratio in December are higher than that in January, at any altitude. This result cannot be explained only by subsidence.

To test the sensitivity of the HNO_3 mixing ratio to the particulate sulfuric acid mixing ratio (Fig. 4), vertical profiles of H_2SO_4 content were calculated for HNO_3 gas=1 ppbv and 10 ppbv. The difference between both HNO_3 gas concentrations was very small (11% at 14.5 km), and therefore it can be suggested that the decrease in particulate matter in January was not caused only by the decrease in mixing ratio of HNO_3 gas. Descending motion of individual particles which grew up through uptake of water vapor and/or nitric acid vapor possibly affect the variation in density profile of atmospheric sulfate, as mentioned below.
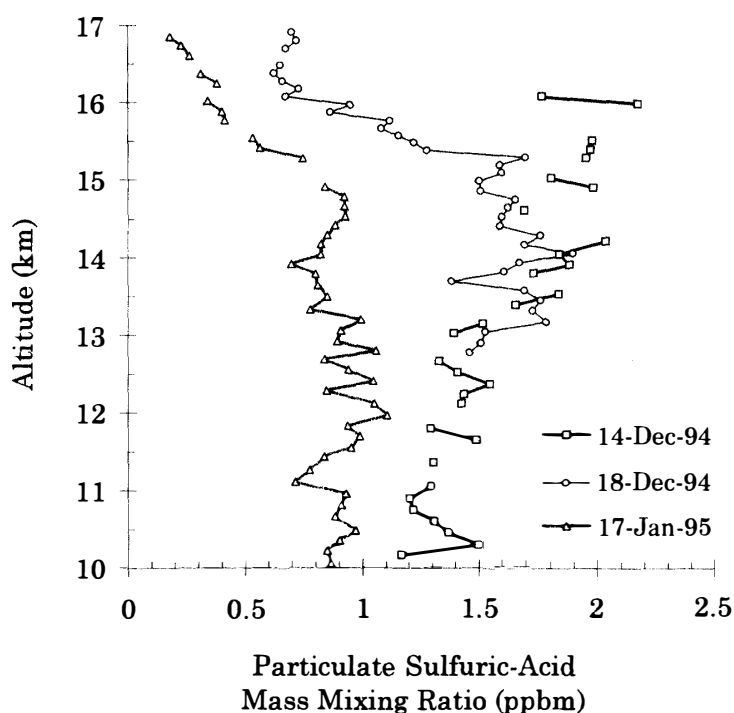

Fig. 4. Particulate sulfuric acid mixing ratio to in-situ air (ppbm; 10^{-9} kg H_2SO_4/kg-Air).
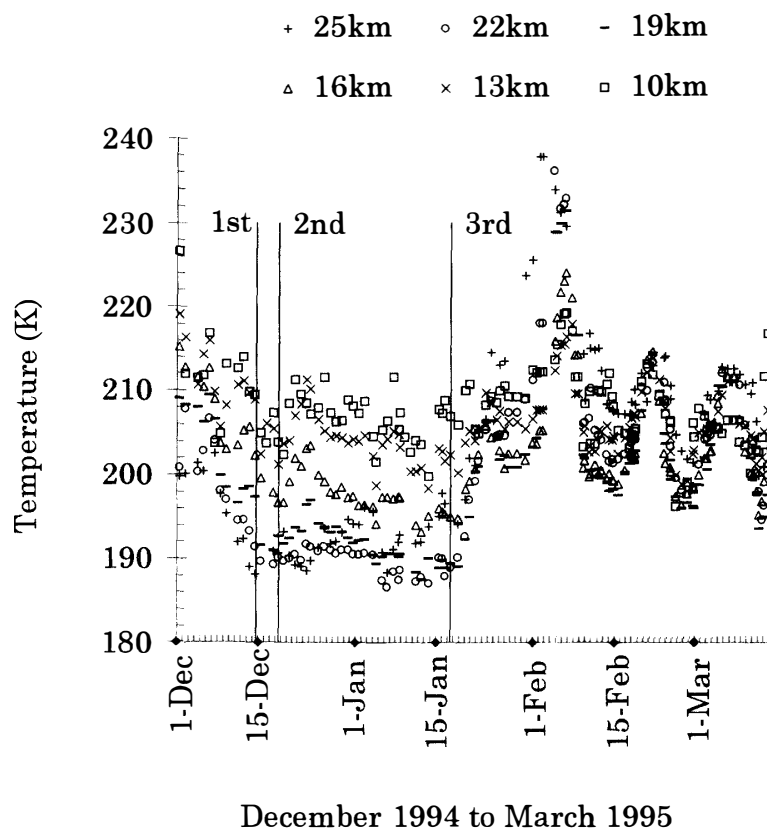
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Fig. 5.   Stratospheric temperature at Ny-Ålesund.   Vertical bars mean the dates when optical
particle balloon-sounding was made.

The concentration on January 17, 1995 was apparently lower than the values on December 14 and 18, 1994, not only in the region where PSC formation is expected but also in the lower aerosol layer where temperature is usually higher than the frost point of PSCs (Fig. 3).

In Fig. 6, the number-size distribution of particles with radii of 0.2–1.0 μm, in the background layer, averaged between 350 and 370 K potential temperature surfaces for December 14, 18, and January 17 (a), and 330–350 K for one profile of January 17 (b). Particle radii are calibrated to that of 70 wt% H_2SO_4 water-solution, calculated from formulas of TABAZADEH *et al.* (1994), to remove the dependence of particle radius on temperature.   The concentration of particles with diameter of 0.5–0.8 μm observed on January 17, 1995 was apparently lower than the values on December 14 and 18, 1994, suggesting the possibility that the decrease in particulate matter in the background aerosol layer is due to decrease in number concentration of aerosol particles of larger diameter.

One possible process is coalescence of background sulfuric acid droplets with descending PSCs particles, since formation of PSCs, as shown in Fig. 1, was very active in the upper aerosol layer.

Another possible process is descending of background aerosol particles which freeze and grow through uptake of water vapor as temperature decreases.   As shown in Fig. 1, we detected particles larger than 3.6 μm in diameter even in the altitude region below 17
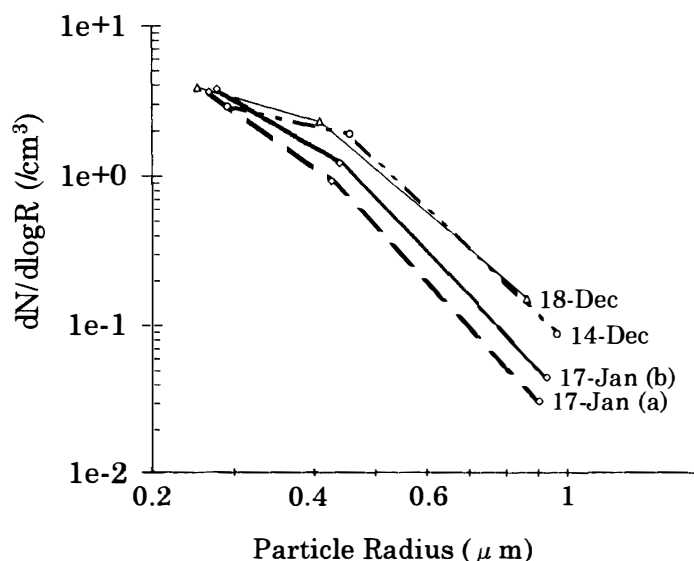
Fig. 6. Number-size distribution of particles observed in the height range of 350-370 K potential temperature, and 330-350 K potential temperature.
◯: December 14, 1994, 350-370 K.   △: December 18, 1994, 350-370 K.
◇(a): January 17, 1995, 350-370 K.   ◇(b): January 17, 1995, 330-350 K.

km.   Such large particles cannot be observed in the calm stratosphere at mid- and lower-latitudes.   However, it is difficult to discuss quantitatively such microphysical processes, owing to lack of measurements of concentration of water and nitric acid vapor, chemical composition of particles, and shape of particles.

From the viewpoint of the global budget of atmospheric sulfur, decrease in particulate sulfur content in the polar winter stratosphere seems as an effective sink since the decreasing rate of particulate sulfuric acid, several tens percent decrease a month (Fig. 4) in the polar region, is not negligible for the global background concentration and estimated influx of sulfur from the troposphere to the stratosphere.

However, the present estimated value is based only on short term measurements at Ny-Ålesund.   The disturbance of the Pinatubo eruption of 1991 may remain.   And if so, the estimated loss could be a characteristic phenomenon for a volcanically disturbed air-mass and not typical values during a volcanically calm period.   It is necessary to continue to observe particle size and number concentration.

The present discussion has assumed homogeneous distribution in the horizontal direction inside the polar vortex on the basis of temperature distribution in the polar stratosphere measured at Ny-Ålesund.   If there is large inhomogeneity of particle concentration in horizontal direction, it causes noticeable changes in particle density on an isentropic surface.

The vertical distribution of particle concentration described here was obtained from balloon-borne measurements only at Ny-Ålesund station. It is impossible to understand the horizontal structure of aerosol particle distribution from the present measurements. Balloon-borne measurements of stratospheric aerosol size and concentration at many stations will be needed to understand the horizontal structure of the aerosol layer.

4. Conclusions

Balloon-borne measurements of atmospheric aerosol particle number-size distribution made in the winter of 1994/1995 at Ny-Ålesund suggest the possibility that stratospheric particulate sulfuric acid was transported from the stratosphere to the troposphere through individual particles descending.

—Volume density of aerosol in January 17 was lower than those in December 14 and 18 above 13 km altitude (Fig. 2).

—Stratospheric temperature on December 14 was higher than that on December 18 and January 17, and temperature on January 17 was lower than that on December 18 above 15 km (Fig. 3).

—Compared with two observational results in December, a several ten percent decrease of particulate sulfuric acid in January 17 was estimated between 10 and 17 km (Fig. 4). These results suggest that the winter polar stratosphere functions as one possible sink in the global budget of stratospheric sulfate.

Acknowledgments

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