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LIDAR OBSERVATION ABOVE SVALBARD, NORWAY IN THE WINTER OF 1996/97 —Characteristics of Backscattering Ratio and Depolarization Ratio of PSC Particles—

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Abstract: Lidar observations of polar stratospheric clouds (PSCs) were performed in the winter of 1996/97 at Ny-Ålesund, Svalbard, Norway (79°N, 12°E). Corresponding to the appearance of an area colder than the estimated frost point of nitric acid trihydrate (NAT) above Ny-Ålesund, PSCs were observed intermittently. As the temperature is lower than the NAT frost point, the peaks of scattering ratio (R) tend to increase and most PSC layers with large enhancement in R show negative correlation between R and depolarization ratio (δ). This suggests that the PSC layers were composed of liquid or small particles, which showed small values of δ even though the temperature was very low in that height range. During the period when the temperature profile showed large wave-like fluctuations, the detected PSCs varied remarkably in time and space. Heights of R peaks observed during that period did not coincide with the temperature minima which were measured once a day by rawinsonde.

1. Introduction

Since FARMAN *et al.* (1985) reported the drastic depletion of stratospheric ozone over the Antarctic, scientific interest in the physical and chemical perturbation in the stratosphere above both poles has increased dramatically. Especially, polar stratospheric clouds (PSCs) are considered to play important roles in ozone depletion because of conversion of chlorine atoms from inactive to active forms by the heterogeneous reaction on the particle surfaces (SOLOMON, 1988; SALAWITCH *et al.*, 1989; HAMILL and TOON, 1991).

NAUJOKAT and PAWSON (1996) reported from their daily subjective analyses of radiosonde data that stratosphere in both winters 1994/95 and 1995/96 were extremely cold. Type I PSCs have been frequently observed in the arctic winter in the past several years. Recent laboratory experiments suggest that nitric acid trihydrate (NAT) (HANSON and MAUERSBERGER, 1988; MARTI and MAUERSBERGER, 1993) is the most stable composition of type I PSC and such phases as nitric acid di- or more higher-hydrate, ternary H_2SO_4/HNO_3 hydrate, liquid $H_2SO_4/HNO_3/H_2O$ are metastable compositions (MARTI and MAUERSBERGER, 1993; WORSNOP *et al.*, 1993; CARSLAW *et al.*, 1994; TABAZADEH *et al.*, 1994). Their theories, however, are not supported by enough field observations such as lidar observations and balloon observations (HAMILL *et al.*, 1996; SHIBATA *et al.*, 1997). It is important to have qualitatively and quantitatively results from field observations in order to understand exactly the processes of particle formation, growth and behavior of PSCs.

We installed a YAG lidar system at Ny-Ålesund in September 1993 and have performed observations in every winter since January 1994. In the winter of 1996/97, PSCs were detected intermittently as in former winters.

In this paper, the results of PSC observations in the winter of 1996/97 will be reported. Then they will be compared with those of the former winter 1995/96, considering the particle formation mechanism.

2. Measurements

Our lidar system for the observation of stratospheric aerosol consists of Nd: YAG laser, Schmidt Cassegrainian telescope, and other detecting systems which include photomultipliers, interference filters and some other optics.

In the analysis of lidar data we use the scattering ratio R at 532 nm, atmospheric depolarization ratio δ at 532 nm and Angstrom coefficient α. All profiles are normalized to the molecular backscattering at the height above the background aerosol layer (about 30 km).

R is defined as $R=(\beta_R+\beta_M)/\beta_R$, where β_R and β_M are the molecular and aerosol backscattering coefficients respectively, and R-1 can be considered to be roughly proportional to the mass mixing ratio of particulate matter. δ is defined as $\delta=\beta_{\perp}/(\beta_{\perp}+\beta_{\parallel})$, where $\beta_{\perp}$ and $\beta_{\parallel}$ are the backscattering coefficients for the returns perpendicular and parallel to the polarization plane of the incident laser light. The profiles of δ are normalized to the molecular value of 0.01 between 25 km and the upper end of the lidar profile. If δ takes values larger than 0.01, it means that the particulate matters are composed of the nonspherical particles such as crystallized and solid particles. The Angstrom coefficient α is defined as $\alpha=-\ln(\beta_{M1064}/\beta_{M532})/\ln(1064/532)$ where β_{M1064} and β_{M532} are the aerosol backscattering coefficients at 1064 nm and 532 nm respectively. On the assumption that β_M depends on the wavelength (λ) according to $\beta_M \propto \lambda^{-\alpha}$, α represents the contribution of the size distribution of the particulate matter. Roughly speaking, if the value of α is smaller, the size distribution is considered to be shifted toward larger sizes.

All daily profiles of temperature used in the analysis were based on balloon measurements and supplied by courtesy of AWI.

3. Observations

Lidar observations were performed from December 1996 to March 1997. Superposed on the usual aerosol layer, PSCs were observed intermittently in this period.

The stratospheric temperature above Ny-Ålesund fell slowly from the middle to the end of December 1996. The first cold area in which the temperature was lower

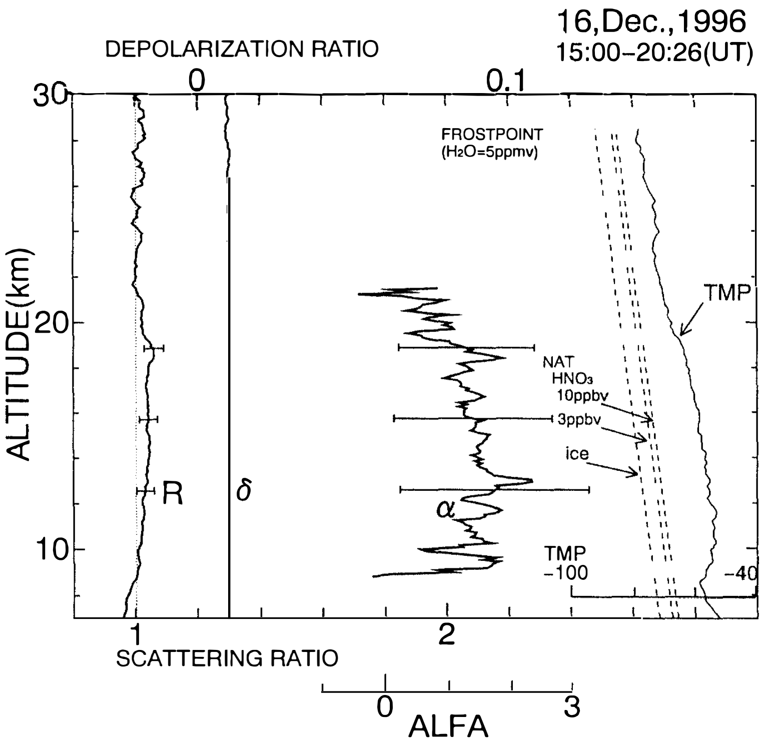

Fig. 1. Vertical profiles of backscattering ratio (532 nm), depolarization ratio (532), Angstrom coefficient, temperature and estimated NAT frost point observed from 1500 to 2026 UT on 16 December 1996.

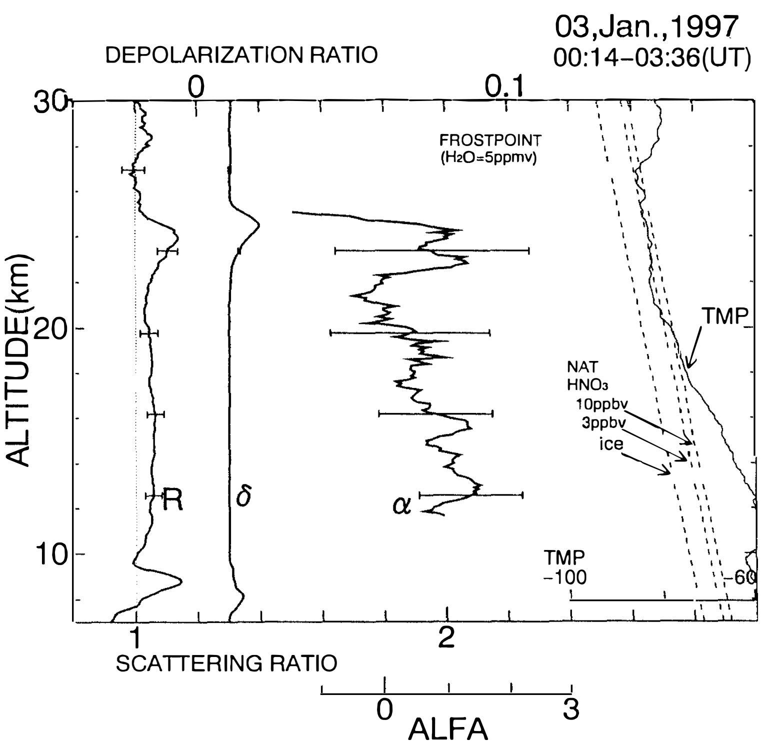

Fig. 2. Same as Fig. 1 except from 0014 to 0336 UT on 3 January 1997.

than NAT frost point (H_2O=5 ppmv, HNO_3=10 ppbv) appeared in the vicinity of height 26 km over Ny-Ålesund on 25 December 1996, and slight enhancement in R was detected in that height range at the end of December. The first distinct PSC layer was detected on 2 January 1997.

Figure 1 shows the profiles of R, δ, α of the typical background aerosols observed on December 16, 1996.

Figure 1 also shows the temperature profiles and the frost points of ice and NAT respectively, which were estimated on the assumption that the air mass of interest has mixing ratios of 5 ppmv H_2O, and 3 ppbv, 10 ppbv HNO_3. The frostpoint of NAT estimated at mixing ratio of 3 ppbv HNO_3 is about 2 K lower than those estimated at mixing ratio of 10 ppbv HNO_3.

The polar vortex was not yet fully formed and the stratospheric temperature above Ny-Ålesund was relatively warm on December 16, 1996. The vertical profile of R takes a maximum at the height of 15 km but shows no prominent peak from about 10 to 22 km altitude, where the temperature is obviously higher than the expected NAT frost point (10 ppbv HNO_3). δ also showed no sign of nonspherical particles. Although a little enhancement in R with no increment of δ at 18.5 km altitude can be seen, a few days of back trajectories show that the air mass was transported from outside to inside the polar vortex. Such a slight enhancement in R with no increment of δ was frequently detected during the observed period. Figure 2 shows the profile observed on January 3, 1997. Superposed on the typical profile of R and δ for the background aerosol layer, the enhancement in both R and δ can be seen in the height range from 21 to 26 km, where the temperature is 1–3 K lower than the NAT frost point. The values of peaks of R and δ are 1.15 and 0.02 respectively at the same height of about 24 km. The enhancement in δ suggests that the air mass in that height range includes nonspherical aerosols such as NAT and it is possible for the NAT to be formed if we assume the appropriate amounts of H_2O and HNO_3 vapor in the air mass. The correlation between R and δ at the height range of PSC is positive. PSC layers with such clearly positive correlation between R and δ were frequently detected in the early stage of the PSC detection period, *i.e.*, the early appearance of cold air mass above Ny-Ålesund. Such a tendency was also seen in the last winter under similar conditions (SHIRAISHI *et al.*, 1997). During January 9 to January 15, we could not detect echoes of such PSCs in spite of the frequent appearance of a cold air mass at a temperature 2–3 K lower than the NAT frost point.

From January 19 to January 25, when a cold air mass at the temperature of 4–5 K lower than NAT frost point appeared above Ny-Ålesund, PSCs were detected frequently again. Figures 3 and 4 show the profiles observed continuously at 0043–0403 and 0407–0725 (UT) on January 22, 1997 respectively. PSC layers lie in the same region from 20 to 24 km in both figures. But the peak value of R at 0043–0403 is 2 at the height of 22 km, twice larger than that at 0407–0725. The observed PSC layers varied remarkably in time. Values of δ are low and almost equal to that of background aerosol in the region of enhancement in R, where α took a maximum at the value of about 1.7 which is larger than that in the background aerosol range at 14–16 and 18–19 km in Fig. 3. It is suggested that the detected PSC is composed of small and spherical particles. PSCs observed in this period were highly variable in time and

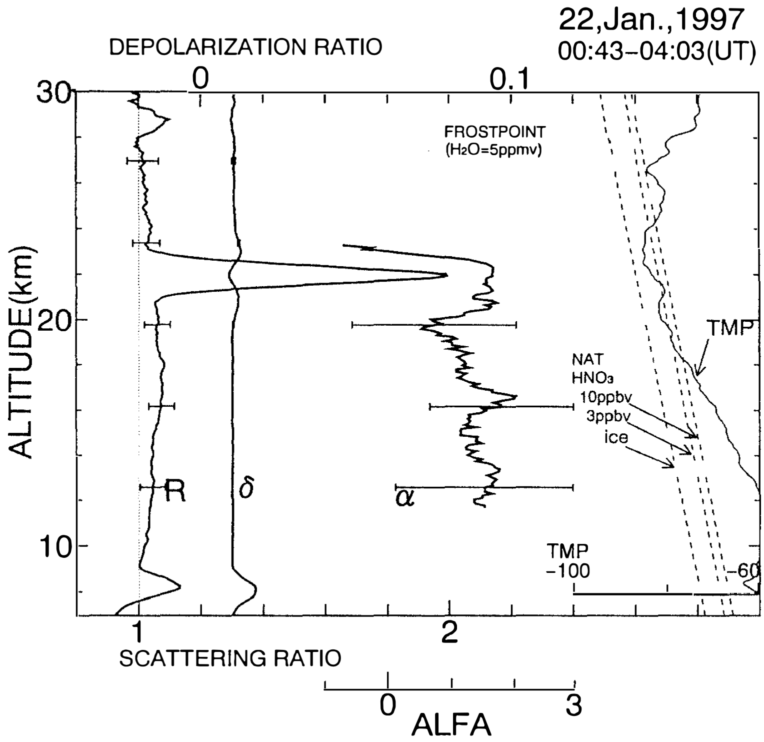

Fig. 3. Vertical profiles of backscattering ratio (532 nm), depolarization ratio (532), Angstrom coefficient, temperature and estimated NAT frost point observed from 0043 to 0403 UT on 22 January 1997.

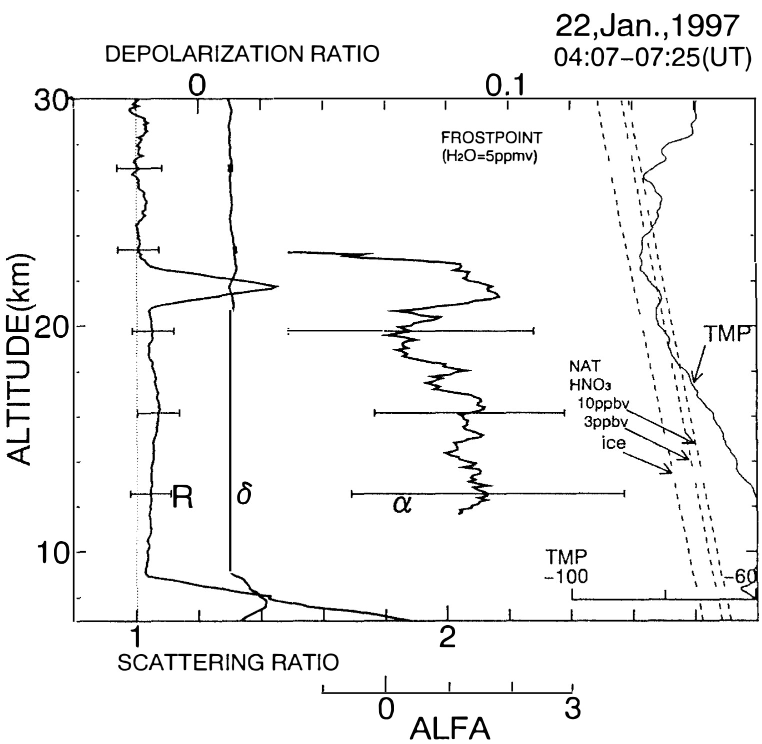

Fig. 4. Same as Fig. 3 except from 0407 to 0725 UT on 22 January 1997.

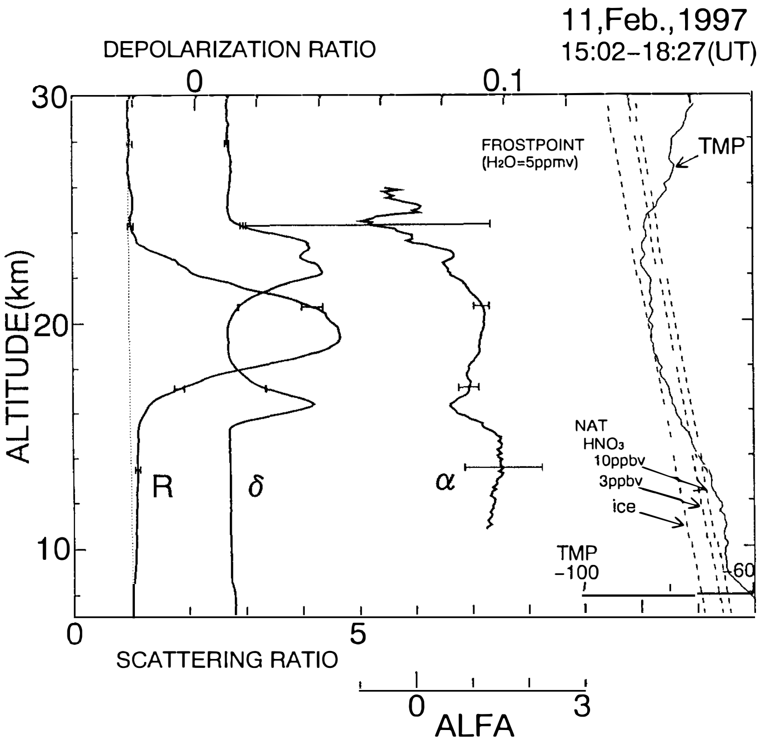

Fig. 5. Same as Fig. 3 except from 1502 to 1827 on 11 February 1997.

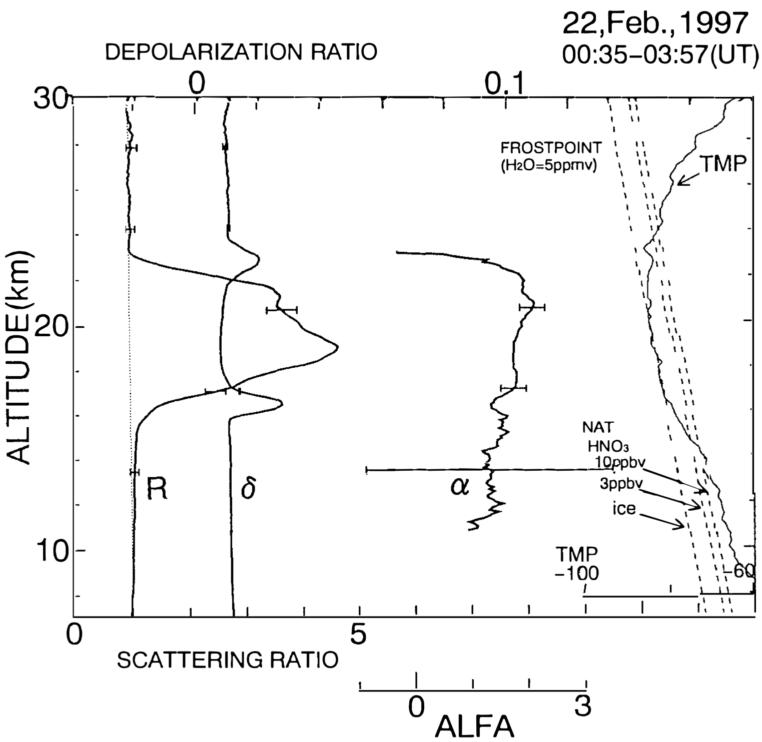

Fig. 6. Same as Fig. 3 except from 0035 to 0357 on 22 February 1997.

space.

From early to mid February 1997 the cold area appeared again just below the height of the previous cold area and cooling progressed rapidly to the fully developed stage. Figures 5 and 6 show the typical profiles observed in this period. Large enhancement of both R and δ in both profiles can be seen in the height range at a temperature lower than the NAT frost point. Layers with enhancement of R in both profiles are sandwiched between two peaks of δ, that is, a layer composed of spherical particles is sandwiched between two layers of non-spherical particles. Such PSC layers of "sandwich structure" were frequently observed also in the latter two winters when the cold area lower than ice frost point appeared above Ny-Ålesund. α at the peak height of R took a maximum in Fig. 5, while α observed on 22 February, 1997, took a minimum. The difference came from the difference of size distribution at the peak heights of R in the two figures.

4. Discussion

PSCs observed in the winter of 1996/97 varied in space and time in a manner remarkably similar to the variations observed in the winters of 1995/96 and 1994/95 (SHIBATA *et al.*, 1996, 1997; SHIRAISHI *et al.*, 1997). We frequently detected PSCs of "Sandwich structure" when the polar vortex developed fully and the temperature was lower than the frost point of NAT.

Figure 7a shows the variation of correlation coefficient between R and δ with the temperature difference dT (dT=T-estimated frost point of NAT) at the peak height of R. The correlation coefficients were estimated over the height range where the values of R exceed those of background aerosols. Figure 7b shows the behavior of peak values of R in all PSC profiles versus dT at the same height. The tendency for the increase in the value of R, which was accompanied by the decrease of dT, is obvious from Fig. 7b, and that tendency is remarkable below dT of -6 K. Most PSCs detected at dT from -5 K to -7 K show a negative correlation between R and δ in Fig. 7a, that is, PSCs with large enhancement in R indicate low δ. In other words, it is suggested that the detected PSCs were composed of liquid or small particles which showed small δ even though the temperature was very low. TOON *et al.* (1990) mentioned that small nonspherical particles exhibited a small depolarization ratio. Meanwhile SHIBATA *et al.* (1997) have pointed out that the PSC layer with enhancement in R and small δ may be mainly composed of ternary solution ($H_2SO_4/HNO_3/H_2O$). As the temperature lowers below the NAT frost point, supercooled sulphuric acid droplets rapidly grow by absorbing HNO_3 and H_2O vapor, and the droplets remain liquid at a temperature close to the ice frost point. They can yield larger aerosol volumes than would have been the case for solid NAT particles (CARSLAW *et al.*, 1994). In this case, α in the height range may be smaller than that in the other height regions in which PSCs have different optical features as shown in Fig. 6.

PSCs with positive correlation between R and δ as shown in Fig. 1 were frequently detected in the region where dT is slightly lower than the NAT frost point at the beginning of appearance of the cold air mass above Ny-Ålesund in early January 1996 and early February 1997. The positive correlation means that the PSC layer with

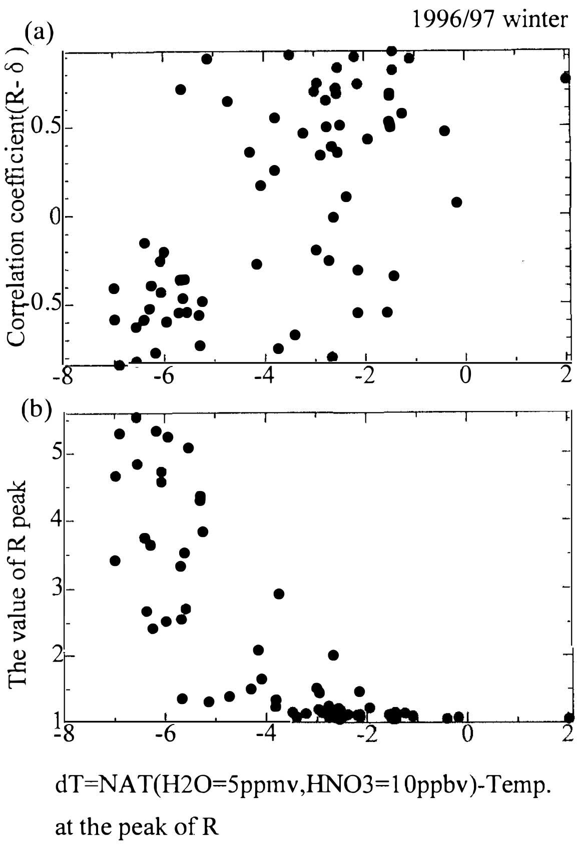

Fig. 7

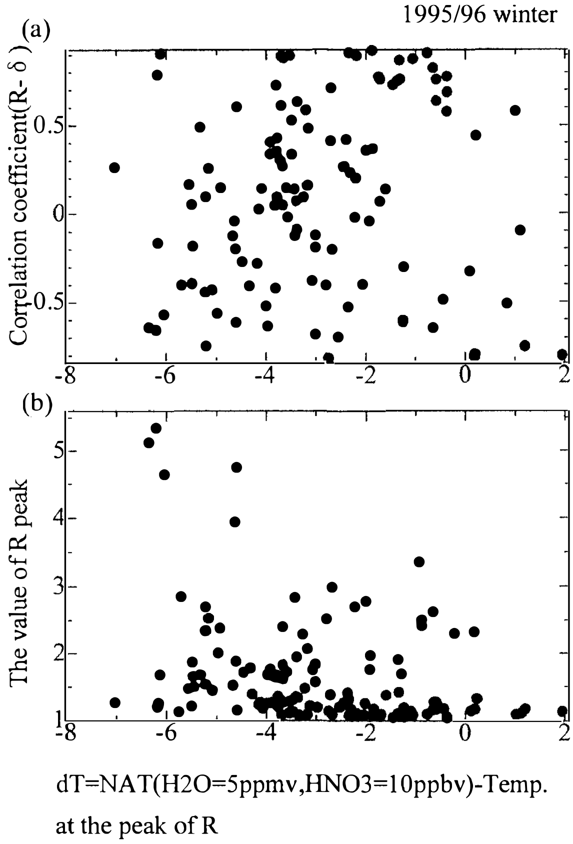

Fig. 8

Figs. 7, 8. The variation of correlation coefficient between R and δ with the temperature difference dT (dT=T-estimated frost point of NAT) at the peak height of R (a). The correlation coefficients were estimated over the height range where the values of R exceed those of background aerosols. The behaviors of peak values of R in all PSC profiles are plotted versus dT at the same height (b).

enhancement of R was not composed of liquid but nonspherical particles such as crystallized particles. Such a tendency can also be seen in the former winter of 1995/96, when PSCs were observed at the beginning of appearance of cold air mass above Ny-Ålesund (SHIRAISHI *et al.*, 1997).

To compare with the former winter, we draw the profiles observed in the winter of 1995/96 in Figs. 8a and 8b on the same scale as in Figs. 7a and 7b. In Fig. 8b, no trend as mentioned above in Fig. 7b was found; we detected relatively larger values (2–3) of R at the temperature near the NAT frost point. For instance, a dot located at $dT=-1$ K and $R=3.3$ shows the PSC profile observed on January 17, 1996 in Fig. 9. The detected PSC layer had a peak in R at the height of 20 km, where the temperature was about 1 K lower than the NAT frost point. Considering small δ (<0.07) and large α of the detected PSC layer, it can be interpreted that the observed PSC layer was composed of small particles (TOON *et al.*, 1990). Considering the temperature history of the air mass which included the enhancement in R, the temperature estimated by back trajectory analysis on the synoptic scale was 7 K lower than the NAT frost point at 20 km altitude at the time of observation. The temperature esti-

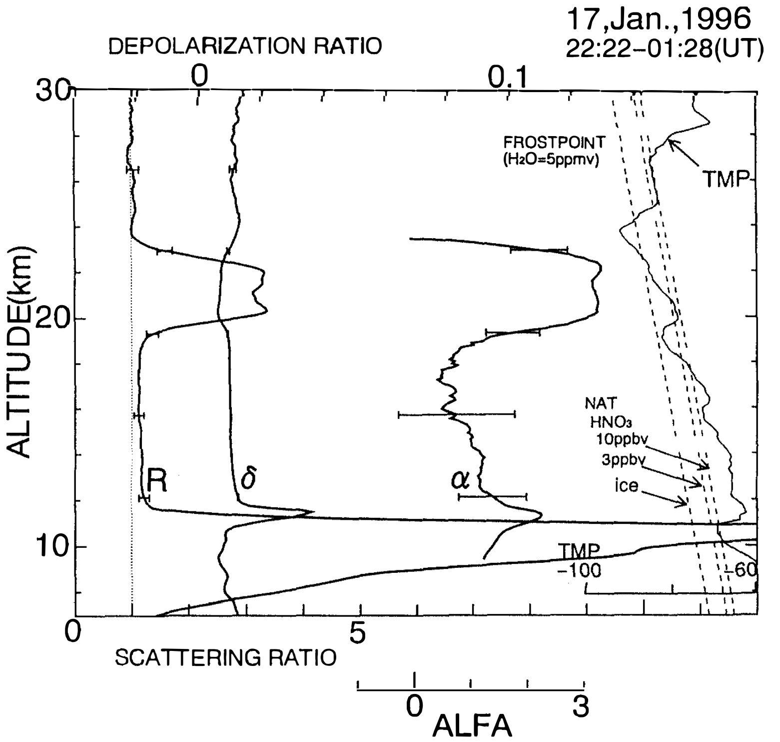

Fig. 9. Vertical profiles of backscattering ratio (532 nm), depolarization ratio (532), Angstrom coefficient, temperature and estimated NAT frost point observed from 2222 to 0128 UT on 17 January 1996.

mated was 6 K lower than that at the dot noticed above in Fig. 8 or at the height of 20 km in Fig. 9. All temperature profiles in Figs. 1 to 9 were measured once a day by rawinsonde observation. The difference of the temperature may show high temperature fluctuation in the height range where PSC layers were observed. In Fig. 9, the temperature profile on 17 January, 1996 had large wave-like fluctuations. The wavelength is about 5 km. In the winter of 1995/96, such wave-like fluctuations were frequently detected above Ny-Ålesund, when PSCs varied remarkably in time and space. The appearances of peak of R of PSCs observed during that period were not strongly dependent on dT.

SHIBATA *et al.* (1996) indicated that the coexistence of different types of PSC layer in a small altitude range (about 1 km) was detected and it was difficult to explain the connection to the synoptic scale thermal history of the air mass. They also suggested that the possible effect of small- or meso- scale dynamical variations on the formation mechanisms of PSCs must be considered. During the period when the PSC layer was detected, temperature profiles with such large wave-like fluctuations were observed, too. Such wave-like fluctuations may have strong influence on the stratospheric temperature fluctuation and particle formation of PSCs.

5. Conclusions

Lidar observations of PSCs were performed from December 1996 to March 1997 at Ny-Ålesund, Svalbard, Norway (79°N, 12°E). PSCs were observed frequently, corresponding to the appearance of a cold area at temperature lower than the NAT frost point. As the temperature is lower than the NAT frost point, peaks of R tend to increase clearly and most PSC layers with large enhancement in R show a negative correlation between R and δ. This suggests that the PSC layer was composed of liquid or small particles, which showed small δ at the height range of very low temperature. PSCs with positive correlation between R and δ were frequently detected at the beginning of appearance of the cold air mass above Ny-Ålesund in early January 1996 and early February 1997. Such a tendency was also observed in previous winters. When the vertical profile of the temperature had large wave-like fluctuation, detected PSCs were characterized by high variability in time and space. Heights of R peaks observed during that period did not coincide with the temperature minima observed by rawinsonde once a day.

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