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Scientific Paper

LIDAR OBSERVED POLAR STRATOSPHERIC CLOUDS OVER SVALBARD IN MID-DECEMBER 1994

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Abstract: Polar stratospheric clouds (PSCs) were observed with lidar at Ny-Ålesund, Svalbard in the winter of 1994/1995. Compared with the temperature record over several years, the stratospheric temperature over Ny-Ålesund was very low in December and January in this winter. In the period when the PSCs were first observed in mid-December, a slight increase of stratospheric aerosols at the top side of the background aerosol layer was found. This increase can be understood as the first stage of PSC formation in the winter. The lidar-observed PSCs in this period are characterized by high variability in space and/or time. As a typical example of these features, an event on December 12, 13 and 14 is presented. In this case two layers of scattering ratio and depolarization were observed within about one km of the vertical separation. The life time of the layers was only a few hours. The characteristics of the PSCs in this period cannot be simply explained by the NAT formation theory which has been widely accepted.

1. Introduction

Stratospheric ozone destruction over the polar region is now believed to be caused through heterogeneous reactions on the surface of polar stratospheric clouds (PSCs). Chlorine atoms, most of which are in inactive molecules, are activated by these reactions (SOLOMON *et al.*, 1986; MCELROY *et al.*, 1986). At the same time, PSCs absorb HNO₃, decreasing NO_x (de-nitrification) that rapidly deactivate active chlorine (CRUTZEN and ARNOLD, 1986; TOON *et al.*, 1986; FAHEY *et al.*, 1989). Thus PSCs play a crucial role in ozone destruction in the polar stratosphere. However, the composition, the physical state and especially the formation processes of PSCs are still uncertain (MOLINA *et al.*, 1993; MACKENZIE *et al.*, 1995). Although it was once widely accepted that a rather simple mechanism by crystals of nitric acid tri-hydrate (NAT) and ice can describe almost all of the aspects of PSC formation, some observed results cannot be explained by NAT (DYE *et al.*, 1992), and re-estimation of observed infrared spectrum over Antarctica strongly suggests that NAT was not included in the observed Antarctic PSCs (TOON and TOLBERT, 1995). In this situation, observational work on PSCs is becoming more important in work on the ozone problem.

To observe aerosols in the stratosphere and in the troposphere, a Nd: YAG lidar system was installed by an atmospheric science team of Nagoya University and Fukuoka University at Ny-Ålesund (79°N, 12°E) in 1993. The observation campaigns which were concentrated to observe PSCs were conducted in three winter seasons from January through the beginning of March 1994, from December 1994 through March 1995, and from November 1995 through February 1996. The PSCs appeared very frequently in the winter of 1994/1995. The frequency of PSC appearance in the winter of 1994/1995 was the highest in these four winters since 1991 according to reports of observations in the winters of 1991/1992, 1992/1993, and 1993/1994 (BEYERLE *et al.*, 1994; BEYERLE and NEUBER, 1994; FUJIWARA *et al.*, 1994). In the winter of 1994/1995, the first detection of PSCs by our observations was on December 13 through 14. In this paper, the lidar profiles on these days will be reported. The formation mechanism of PSCs is still unclear; the first stage of PSC appearance will suggest a possible mechanism.

2. Lidar Observations

In the following discussion, results are presented in terms of the scattering ratio (*R*) and depolarization ratio (δ) at 532 nm. The definitions of these values are as follows: *R* is defined as $R = (\beta_R + \beta_M)/\beta_R$, where β_R and β_M are Rayleigh and Mie backscattering coefficients. *R*-1 is nearly proportional to the mass mixing ratio of the particulate matter. The depolarization ratio δ is defined as $\delta = \beta_\perp/(\beta_\parallel + \beta_\perp)$, where β_\parallel and β_\perp are components of the backscattered light parallel and perpendicular to the polarization plane of the linearly polarized laser beam. δ is a measure of the sphericity of the particles. β_\perp is exactly zero if the particles are perfect spheres as, for example, liquid particles. In this paper δ means total depolarization that includes the depolarization effects by both Mie and Rayleigh scattering. In our system, δ is about 0.5% where there are no aerosol particles (ADACHI, 1996). If δ takes values larger than 0.5%, it means that there are nonspherical aerosol particles.

Lidar observations in the winter of 1994/1995 were made from December 11, 1994 to March 11, 1995. The observations were continued almost all the time when there was no low altitude cloud cover, especially when PSCs were detected.

3. Results

The global distribution and formation mechanism of background aerosols are now very well established (TURCO *et al.*, 1982; SHIBATA, 1995). Before starting to describe the results, we introduce the concept of profiles of the background stratospheric aerosol layer. The vertical profile of R-1 (volume mixing ratio) of the background stratospheric aerosol layer (Junge layer) shows a simple Gaussian-like layered structure with a single peak. Over the Arctic region R-1 has its maximum at about 15 km altitude. The layer width is about 10 km (FWHM). The top side of the background aerosol layer over the polar region is usually at about 25 km altitude, and is at about 20 km if the layer is near the center of the polar vortex because of the downward displacement of air inside the polar vortex (MCCORMICK *et al.*, 1983; ROSENFIELD and SCHOEBERL, 1986). Here we define the top side as the height where R-1 is smaller than about 1/e of the peak value of the Junge layer above this height. δ of the Junge layer takes values smaller than the depolarization ratio of Rayleigh scattering (~ 0.5%), indicating that the background aerosols are

spherical particles. This shows that the aerosol particles of the Junge layer are composed of supercooled sulfuric acid liquid droplets.

The stratospheric temperature over Ny-Ålesund took minimum values from mid-December 1994 to mid-January 1995 in the winter of 1994/1995 in radiosonde soundings by the German Alfred Wegener Institute. This low temperature is understood as the reflection of the polar vortex being over Ny-Ålesund during this period. Figures 1 and 2 shows profiles of R and δ observed by the lidar, and temperature profile observed by a radiosonde on December 12 and 13, 1994.

In Fig. 1 there is no sign of the existence of PSCs in R or δ profiles. In other words, there is no clear difference from the profiles of background aerosols. However, in Fig. 2, R and δ are slightly larger than about 36 hours before (Fig. 1) in the altitude range between about 20 and 27 km. A broad layer of R-1 whose peak is at about 25 km and width about 5 km is superposed on the background aerosol layer. The peak value of R-1 is about 0.15 or about half of that of the Junge layer about 0.3. The top side of the strato-

1994/12/12 Ny-Ålesund (08:43-09:42)



Fig. 1. The lidar observed scattering ratio (R; solid line), depolarization ratio (δ ; broken line), and radiosonde observed temperature profile on December 12, 1995, over Ny-Ålesund, Svalbard. The time is in UT.



Fig. 2. Same as Fig. 1 but for December 13, 1995.

spheric aerosols in the profile of Fig. 2 is at the height of about 27 km. This top side height was much higher than the usual Junge layer taking into account that Ny-Ålesund was within the polar vortex (T. NAGAI; private communication). In Figs. 1–4 the profile of the calculated frost point of NAT is plotted (HANSON and MAUERSBERGER, 1988; REMSBERG *et al.*, 1984; GILLE *et al.*, 1984). At the altitude range from about 20–25 km the temperature observed on these days was lower than the NAT formation temperature.

Figure 3 shows the profiles taken on December 14. In these profiles we find sharp layers of R and δ whose peaks are at about 24 and 23 km, respectively. The widths of both layers are about 2 km. The sharp peaks of this amplitude are never observed in the natural fluctuation of the background aerosol layer as mentioned below. These layers are the first clear indication of PSCs in the winter of 1994/1995 in our observations at the wavelength of 532 nm. These sharp layers were just within the altitude range where a slight increase of aerosols was observed, as in Fig. 2.

Figure 4 shows the aerosol profiles observed about 2 hours after Fig. 3. The sharp peaks in Fig. 3 have disappeared.



Fig. 3. Same as Fig. 1 but for December 14, 1995.



1994/12/14 Ny-Ålesund (03:01-04:00)

Fig. 4. Same as Fig. 3.

4. Discussion

Large fluctuations from background aerosol profiles have been reported soon after volcanic eruptions and PSC events over polar regions. Other sources of large fluctuations need not be taken into account (Turco et al., 1982; Shibata et al., 1984; Shibata, 1995). Fluctuations caused by atmospheric gravity waves are estimated to be only several percent at most of the aerosol mixing ratio or R-1 (CHIU and CHING, 1978). The peak value of the increase in R-1 at altitude 25 km in Fig. 2 is too large to be caused by gravity waves. On the other hand, at 25 km in Fig. 2, the temperature is low enough for NAT formation, and is about the temperature at which PSCs including nitric acid have been observed by direct sampling at the same altitude (Iwasaka et al., 1993). Therefore, the slightly increased layer at this altitude is reasonably thought to be caused by PSCs. The profiles observed in the winter of 1993/1994 and the preliminary results in the winter of 1995/1996 with our lidar show a similar amount of enhancement at the beginning of winter. The airborne lidar observations near Svalbard by Pool et al. (1988) also showed similar enhancement of aerosols in the same altitude range, though they were not discussed in that paper. A broad and slight increase of R (R=1.1-1.2) at the top side of the background layer as in the Fig. 2 is one of the common features in the first appearance of PSCs in a winter season.

The sulfuric acid aerosol particles will grow, following the decrease of the ambient temperature, by the condensation of water and nitric acid molecules (Steele and HAMILL, 1981; CARSLAW *et al.* 1994; TABAZADEH *et al.*, 1994). The temperature on December 13 is a few degrees lower than on December 12 in the altitude range of about 20–25 km. Therefore, the two layers in Fig. 3 would be the results of particle growth following this temperature decrease.

The sharp peak of the depolarization profile is not at the same altitude as the Rpeak. This suggests that there were two altitude ranges of high particle growth (or formation) and the particle phase transition. The two peaks in R and δ cannot be explained simply by NAT formation with a temperature decrease. These two layers contact each other and the peak altitudes are separated by only about one km (or only about 5 hpa). The vertical variation of the synoptic scale horizontal patterns of the polar vortex over the Arctic is not so large in this period that the trajectory and the temperature history of the air parcels that include these layers separated only 5 hpa must be almost the same, though this should be directly confirmed with meteorological data. The peaks in Fig. 3 disappeared in the following observations made a few hours later (Fig. 4). This variation, again, cannot be explained by the synoptic scale difference of the air parcels. There are thermal fluctuations which do not reveal themselves on the synoptic scale temperature map. The observed spatial and temporal fine structures of PSCs suggest that smallor meso- scale dynamical variations have a role in the formation mechanisms of PSCs. The analysis of stratospheric temperature observed by Murphy and Gary also indicates the effect of mesoscale variation for PSC formation (MURPHY and GARY, 1995).

The above case demonstrates that further studies on the formation processes of PSCs are needed.

5. Conclusions

In the PSCs event of the winter of 1994/1995, the first detection of PSCs was at in mid-December 1994. The way of appearance and the relation to temperature cannot be simply explained by particle growth with temperature decrease and phase transition at the critical temperature of NAT formation. Since the mechanism of particle growth and of the phase transition are still not known well, observations of PSC particles are needed with the detailed information of the history of aerosol particle growth and of the thermal history of the air parcels including small- and meso-scale variations. Laboratory experiments are also needed to provide us with the physical properties of stratospheric particles formed mainly by H_2SO_4 , H_2O and HNO_3 . Additionally, it is needed to develop technology to directly sample stratospheric particles and observe them in the cold polar stratosphere.

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References

- ADACHI, H. (1996): External mixture of polar stratospheric clouds: Methodology for analysis by lidar. Master thesis for Nagoya University (in Japanese).
- BEYERLE, G. and NEUBER, R. (1994): The stratospheric aerosol content above Spitsbergen during winter 1991/ 1992. Geophys. Res. Lett., **21**, 1291–1294.
- BEYERLE, G., NEUBER, R. and SCHREMS, O. (1994): Multiwavelength lidar measurements of stratospheric aerosols above Spitsbergen during winter 1992/1993. Geophys. Res. Lett., 21, 57-60.
- CARSLAW, K. S., Luo, B. P., CLEGG, S. L., PETER, Th., BRIMBLECOMBE, P. and CRUTZEN, P. J. (1994): Stratospheric aerosols growth and HNO₃ gas phase depletion from coupled HNO₃ and water uptake by liquid particles. Geophys. Res. Lett., **21**, 2479–2482.
- CHIU, Y. T. and CHING, B. K. (1978): The Response of atmospheric and lower ionospheric layer structures to gravity waves. Geophys. Res. Lett., 5, 539–542.
- CRUTZEN, P. J. and ARNOLD, F. (1986): Nitric acid cloud formation in the cold Antarctic stratosphere: a major cause for the springtime 'ozone hole'. Nature, **324**, 651–655.
- DYE, J. E., BAUMGARDNER, D., GANDRUD, B. W., KAWA, S. R., KELLY, K. K., LOEWENSTEIN, M., FERRY, G. V., CHAN, K. R. and GARY, B. L. (1992): Particle size distributions in arctic polar stratospheric clouds, growth and freezing of sulfric acid droplets, and implication for cloud formation. J. Geophys. Res., 97, 8015-8034.
- FAHEY, J. E., KELLY, K. K., FERRY, G. V., POOLE, L. R., WILSON, J. C., MURPHY, D. M., LOEWENSTEIN, M. and CHAN, K. R. (1989): *In situ* measurements of total reactive nitrogen, total water, and aerosol in a polar stratospheric cloud in the Antarctic. J. Geophys. Res., 97, 11299–11315.
- FUJIWARA, M., IWASAKA, Y., SHIBATA, T., NAGATANI, M., HAYASHI, M., SHIRAISHI, K., NAGATA, K. and UKAJI, K. (1994): Lidar observations of atmospheric aerosols in Svalbard, Norway. 17th International Laser Radar Conference; Abstract of papers, 427–429.
- GILLE, J. C., RUSSELL, III, J. M., BAILEY, P. L., REMSBERG, E. E., GORDLEY, L. L., EVANS, W. F. J., FISCHER, H.,

GANDRUD, B. W., GIRARD, A., HARRIES, J. E. and BECK, S. A. (1984): Accuracy and precision of the nitric acid concentrations determined by the limb infrared monitor of the stratosphere experiment on NIMBUS 7. J. Geophys. Res., **89**, 5179–5190.

- IWASAKA, Y., HAYASHI, M., KONDO, Y., KOIKE, M., YAMATO, M., AIMEDIEU, P. and MATTHEWS, W. A. (1993): Two different type nitrate aerosols in the winter polar stratosphere: morphology of individual particles observed with an electron microscope. J. Geomagn. Geoelectr., 45, 1181-1192.
- HANSON, D. and MAUERSBERGER, K. (1988): Laboratory studies of the nitric acid trihydrate: implications for the south polar stratosphere. Geophys. Res. Lett., 15, 855-858.
- MACKENZIE, A. R., KULMALA, M., LAAKSONEN, A. and VESALA, T. (1995): On the theories of type 1 polar stratospheric cloud formation. J. Geophys. Res., **100**, 11275–11288.
- McCORMICK, M. P., TREPTE, C. R. and KENT, C. S. (1983): Spatial change in the stratospheric aerosols associated with the north polar vortex. Geophys. Res. Lett., **10**, 941–944.
- MCELROY, M. B., SALAWITCH, R. J., WOFSY, S. C. and LOGAN, J. A. (1986): Reductions of Antarctic ozone due to synergistic interactions of chlorine and bromine. Nature, **321**, 759–762.
- MOLINA, M. J., ZHANG, R., WOOLDRIGE, P. J., MCMAHON, J. R., KIM, J. E., CHANG, H. J. and BEYER, K. B. (1993): Physical chemistry of the H₂SO₄/HNO₃/H₂O system: Implications for polar stratospheric clouds. Science, 261, 1418–1423.
- MURPHY, D. M. and GARY, B. L. (1995): Mesoscale temperature fluctuations and polar stratospheric clouds. J. Atmos. Phys., **52**, 1753–1760.
- POOL, L. R., OSBORN, M. T. and HUNT, W. H. (1988): Lidar observations of arctic polar stratospheric clouds, 1988: signature of small, solid particles above the frost point. Geophys. Res. Lett., 15, 867–870.
- REMSBERG, E. E., RUSSELL III, J. M., GILLE, J.C., GORDLEY, L. L., BAILEY, P. L., PLANET, W. G. and HARRIES, J. E. (1984): The validation of NIMBUS 7 LIMS measurements of ozone. J. Geophys. Res., 89, 5161-5178.
- ROSENFIELD, J. E. and SCHOEBERL, M. R. (1986): A computation of stratospheric heating rates and the diabatic circulation for the Antarctic spring. Geophys. Res. Lett., 13, 1339–1342.
- SHIBATA, T. (1995): Numerical modeling of stratospheric aerosols. J. Aerosol Res. Jpn., 10, 197–203 (in Japanese).
- SHIBATA, T., HIRONO, M. and FUJIWARA, M. (1984): The El Chichon volcanic cloud in the stratosphere: lidar observation at Fukuoka and numerical simulation. J. Atmos. Terr. Phys., 46, 1121–1146.
- SOLOMON, S., GARCIA, R. R., ROWLAND, F. S. and WUEBBLES, D. J. (1986): On the depletion of Antarctic ozone. Nature, **321**, 755–758.
- STEELE, H. and HAMILL, P. (1981): Effect of temperature and humidity on the growth and optical properties of sulfuric acid-water droplets in the stratosphere. J. Aerosol. Sci., **12**, 517–528.
- TABAZADEH, A., TURCO, R. P., DRDLA, K. and JACOBSON, M. Z. (1994): A study of Type I polar stratospheric cloud formation. Geophys. Res. Lett., 21, 1619–1622.
- TOON, O. B. and TOLBERT, M. A. (1995): Spectroscopic evidence against nitric acid trihydrate in polar stratospheric clouds. Nature, **375**, 218–221.
- TOON, O. B., HAMILL, P., TURCO, R. P. and PINTO, J. (1986): Condensation of HNO₃ and HCl in the winter polar stratosphere. Geophys. Res. Lett., 13, 1284–1287.
- TURCO, R. P., WHITTEN, R. C. and TOON, O. B. (1982): Stratospheric aerosols: Observation and theory. Rev. Geophys. Space Phys., 20, 233-279.

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