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Lidar observations of liquid and solid PSC at Sodankylä

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INTRODUCTION

Polar stratospheric clouds (PSC) play a major role in the process of Artic and Antartic ozone depletion due to the surface provided for heterogeneus chemical reactions and the removal of NO_2 from the gas phase. Therefore the phase, size and composition of PSC´s should be known. The microphysical structure of the PSC´s depends on the actual temperature and the corresponding airmass thermal history. At temperatures below the ice frostpoint, PSC´s of ice particles (Type II) are observed, while PSC´s seen at temperatures above the frostpoint are classified as PSC Type Ia (anisotropic particles) and PSC Ib (spherical particles). PSC I were believed to consist of nitric acid trihydrate (NAT). NAT should be stable some degrees above the ice frostpoint with a particle shape depending on the cooling rate [Toon et al., 1990]. However, the explanation of PSC based solely on the NAT-hypothesis can not explain a large amount of data [Toon and Tolbert, 1995]. The spherical shape of PSC Ib can be explained with a liquid supercooled ternary solution (STS) consisting of H_2O, HNO_3 and H_2SO_4. Scenarios for the formation of frozen background aerosol (sulfuric acid tetrahydrate, SAT) are now investigated.
The described variance in shape and size of the PSC can be sensed by multispectral 2-polarization lidar, measuring range resolved scattering properties of atmospheric aerosols. Here the lidar observations of PSC´s during the SESAME campaign are compared to the critical formation temperatures of the different PSC types.

EXPERIMENTAL SETUP

Multispectral lidar measurements have been perfomed in Sodankylä (67.37 N, 26.65 E). The lidar was designed especially for polar stratospheric cloud characterization using five wavelengths (355, 387, 532, 750 and 1064 nm) and two simultaneous polarization sensitive detection systems both at 532 and 750 nm. With a narrowband filter on the 532 nm channel the rotational Raman lines were blocked to supress the Rayleigh scattering contribution to the depolarized signal to 0.36 %. This allows a very high sensitivity for nonspherical particles.
From the four wavelengths 355, 532, 750 and 1064 nm we can determine the aerosol size distribution

of the PSC particles, since the depolarization measurement indicates spherical particles. To describe the aerosol depolarization we use here $\delta = (R_s-1) / (R_p-1)*\delta_{mol}$
where R_s and R_p are the backscatter ratios at the perpendicular and parallel polarized detection channels. δ_{mol} is the depolarization from the Cabannes line of the molecular atmosphere, $\delta_{mol} = 0.365$ %.

OBSERVATIONS:

Liquid PSC

On Jan. 19, 1995 we observed a PSC with a scattering ratio up to 3 at 532 nm in an altitude range from 19.5 to 24 km. The aerosol depolarization measured is below our detection limit of 0.25 %, indicating spherical (liquid) particles. The scattering ratio at the four wavelengths and the depolarization at 532 nm are shown in Fig. 1. For this PSC we calculated the refractive index and size parameters for unimodal lognormal distributions using the wavelength dependence of the scattering properties [Stein et al., 1994].

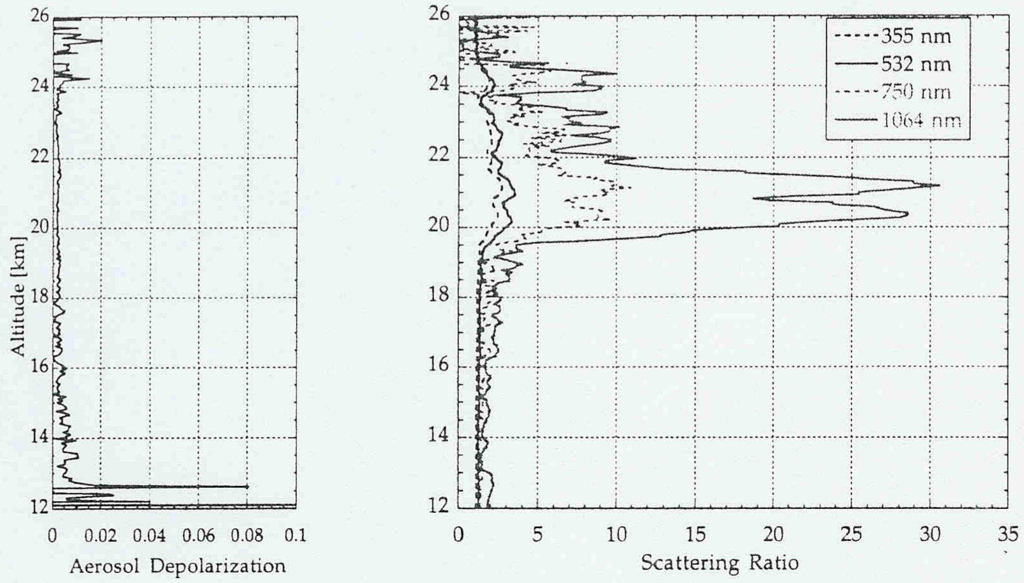

Fig. 1: Aerosol depolarization and scattering ratio for the PSC observed on Jan. 19, 1995.

For the observed PSC only size distributions with refractive indices between 1.33 and 1.38 yield backscatter coefficients which can match the measured values within an error limit of ± 10%. As the index of refraction of water is 1.33 this result indicates a high water content of the particles. The median radius of the particles was retrieved to 0.6 µm.

The local radio sounding (Fig. 2) shows, that the temperature drops below the condensation temperature of NAT at 15 km altitude, while enhanced backscatter could only be observed between 19 km and 24.5 km. This is the height were the temperatures are colder than the existence temperature for supercooled ternary solutions (STS), which is assumed 3-4 K lower than T_{NAT}. [Tabazadeh et al., 1994]. The existence temperatures used here were calculated using a LIMS-Profile for the HNO_3 content of the air and a volume mixing ratio of 5 ppm for water vapor.

The good match of the PSC formation and the cooling below the STS-temperature together with the retrieved high water content indicate particles of ternary solutions in the observed PSC. Although temperatures are below T_{NAT} above 15 km and below T_{STS} from 17-19 km, no aerosols except the weak pinatubo layer can be observed there. The reason for this difference in the particle formation conditions remains unresolved.

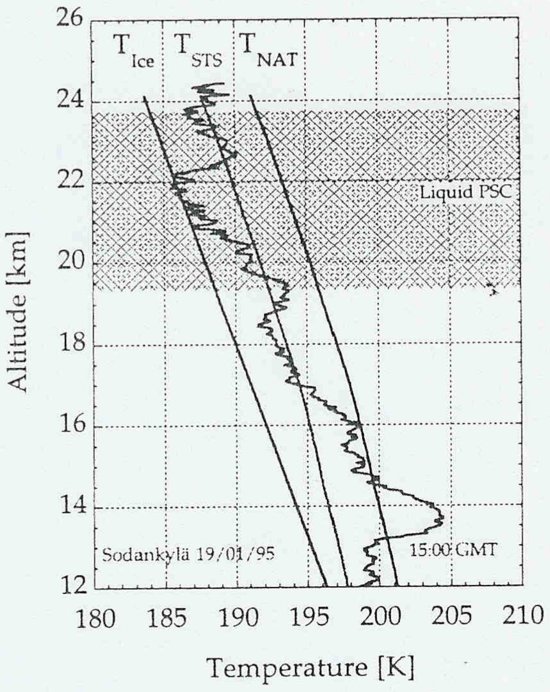

Fig. 2:
Local temperature for Sodankylä at 19/01/95 and existence temperatures for NAT, STS and ice.

Liquid and solid PSC´s

On Jan. 12, 1995 we observed three kinds of PSC´s in different altitude ranges, which can be distinguished by their backscatter and depolarization values (Fig. 3):

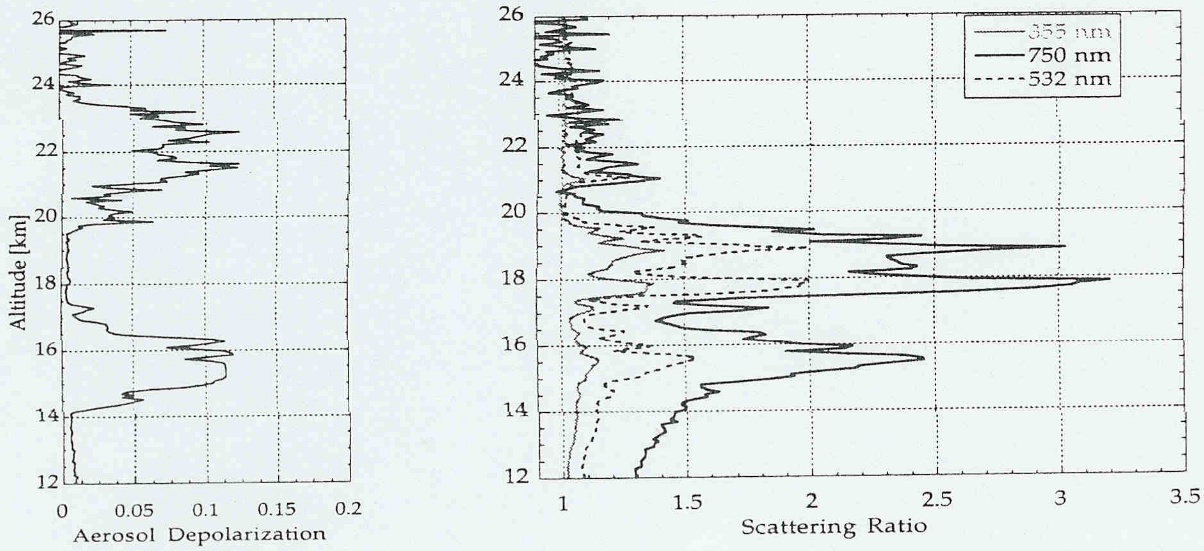

Fig. 3:
Aerosol depolarization and scattering ratio at 3 wavelengths for the PSC´s measured on Jan. 12, 1995 at 21:30 GMT

From 23.5 km down to 20 km we measured an optical thin layers of solid particles which shows a depolarization of about 5 %. Temperatures in this altitude were $T_{NAT} \pm 1.5$ K (Fig. 4). A liquid PSC (aerosol depolarization is lower than 0.25 %) is present in the height range between 19.5 and 17.5 km where the temperature is more than 3 K below T_{NAT}. At 17 to 15 km a third layer with scattering ratios of about 1.6 and aerosol depolarization of 10 % is seen.
For the liquid PSC we found again a coincidence of temperatures lower than T_{STS} and the presence of the PSC. Mie calculations indicate a refractive index of 1.33 - 1.4 and again a median radius between 0.5 and 0.7 µm. We therefore assume that this liquid PSC also consists of ternary solutions.
For the solid aerosol layer in 23 km height we assume particles of frozen background aerosol, SAT. Tabazadeh et al. [1995] described a possible mechanism for SAT-formation in the stratosphere, by cooling below 192 to 194 K and subsequent warming to 198 K. Koop et al. [1995] suggest that SAT formation is only possible when the temperature falls below the ice frostpoint. Temperature histories for this height show that it is consistent with the proposed cooling warming cycle of Tabzadeh et al. Since the temperature of the frostpoint is some degrees uncertain, depending on the water content of the air, and mesoscale temperature variations are not included in the airmass thermal histories of ECMWF, also the latter formation mechanism is possible. A more detailed study of the temperatures histories and their influence on PSC formation is going on.

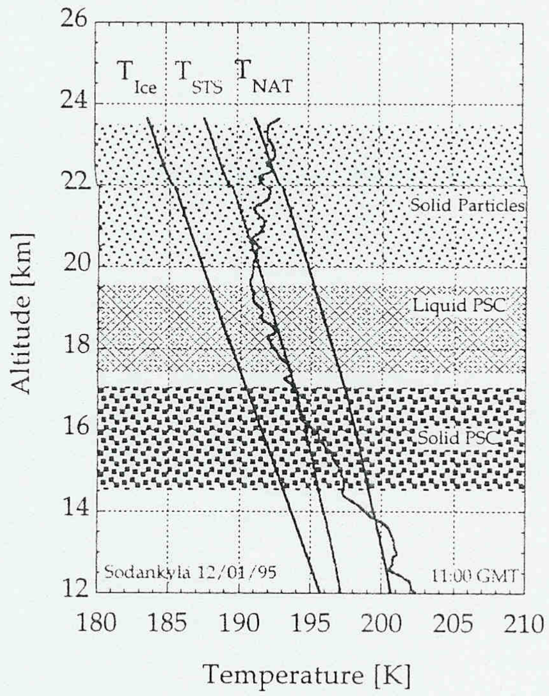

Fig. 4:
Local temperature for Sodankylä at 12/01/95 and existence temperatures for NAT, STS and ice.

Particles of the solid PSC observed are different from the particles in the aerosol layer at 23 km, as they show a higher scattering ratio indicating a larger particle surface but similar depolarization values. In this altitude region the temperature is between 1 and 3 K below T_{NAT}. We assume here NAT-Particles.

CONCLUSIONS

The PSC observations described here cannot be explained by assuming only NAT- particles. The altitude range where PSC Ib are observed corresponds to the height were the temperature is below the formation temperature for liquid supercooled ternary solutions. The formation of solid PSC observed above the frostpoint seems to be more complicated and is not well understood in detail yet. From our data we can not yet verify wether the SAT-mechanism proposed by Tabazadeh et al. can work, or if it is necessary to cool the air below the ice frostpoint to form initially solid PSC particles.

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