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Characterization of liquid and solid PSC's by multispectral Lidar

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Abstract. Lidar measurements at 4 wavelengths and two polarizations were performed during the SESAME campaign in Sodankylä, Finland (67.37N, 26.65E). PSC's consisting of spherical (liquid) particles were observed. For this type of PSC we retrieved the aerosol size distribution and the refractive index using the wavelength dependence of the particle scattering. The measured refractive index of 1.36 indicates a high water content of the PSC particles and we assume that this PSC consists of ternary solutions in contradiction to the NAT-hypothesis.
On the other hand we detected layers of solid particles with very low mass densities of frozen background aerosols. Both types of aerosols can coexist within the same altitude region.

Introduction

Polar stratospheric clouds (PSC) play a major role in the process of Arctic and Antarctic ozone depletion due to heterogeneous chemical reactions responsible for chlorine activation and particle sedimentation redistributing nitrogen species in the stratosphere. Therefore the phase, size and the composition of PSC's should be known. PSC can be divided into PSC type I, observed at temperatures some degrees above the ice frostpoint, and PSC type II consisting of water ice particles occurring at temperatures below the frostpoint. PSC type I can be subdivided into aspherical (type Ia) and spherical (type Ib) particles. Measurements of gas phase HNO_3 removal in presence of PSC's and laboratory studies led to the assumption that PSC type I consist of nitric acid trihydrate and the particle shape depends on the cooling rate [Toon et al., 1990]. However, the explanation of PSC I based solely on the NAT-hypothesis can not explain a large amount of data [Toon and Tolbert, 1995], and other compositions like liquid supercooled ternary solutions (STS) of H_2O, HNO_3 and H_2SO_4 are discussed now [Carslaw et al., 1994].
Multiwavelength, 2-polarization lidar measurements give information about the size distribution, refractive index and physical state of the cloud particles.

Experimental Setup

The aerosol lidar used here was designed to measure the particle backscatter in a wide range of wavelengths using a Nd:YAG laser at 355, 532 and 1064 nm together with a Ti:Sapphire laser at 750 nm and simultaneous detection of all wavelengths. Two additional detection channels acquired the cross polarized signals at 532 and 750 nm. The use of a 0.15 nm (FWHM) bandwidth filter at 532 nm, reduces the Rayleigh signal contribution in the cross polarized channel to only 0.36 % by suppressing the rotational Raman lines. This allows a very high sensitivity for nonspherical particles. From the retrieved Mie backscatter coefficients at the four wavelengths we determined the aerosol size distribution of the PSC particles and their refractive index using an algorithm based on Mie theory [Stein et al., 1994].

Observations

1. Liquid PSC's

On Jan. 19, 1995 we observed a PSC with a scattering ratio up to 3.5 at 532 nm in an altitude range from 19.5 to 24 km (Fig. 1). The aerosol depolarization

$$\delta = (R_s - 1) / (R_p - 1) * \delta_{mol}$$

was below our detection limit of 0.36 %. This proves the presence of spherical (liquid) particles. Here δ_{mol} denotes the depolarization of the Cabannes line of the molecular atmosphere with $\delta_{mol} = 0.365$ % [Young, 1980]. R_s and R_p are the scattering ratios at the perpendicular and parallel detection channel. We retrieved the size distribution and the refractive index of the particles assuming a monomodal lognormal size distribution.
Our algorithm scans size distributions with refractive indices from 1.33 to 1.53, median radii from 0.05 to 1 µm and σ from 1.2 to 1.9. Within the PSC, only particles with refractive

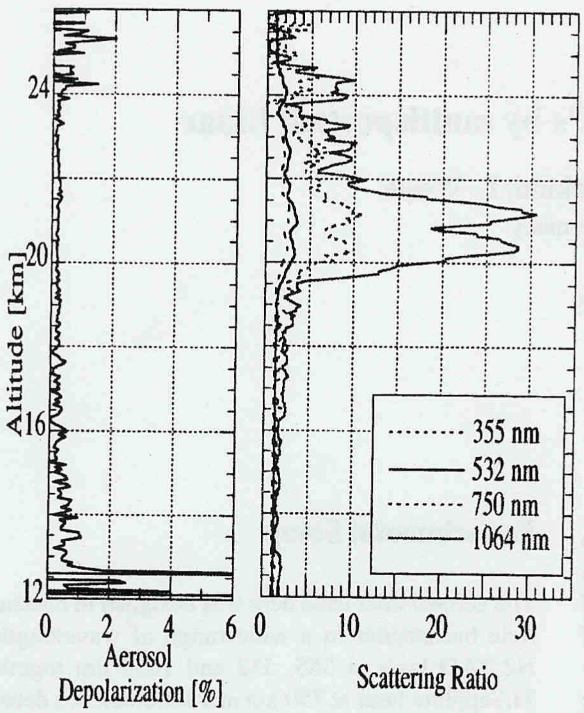

Fig 1: Aerosol depolarization and scattering ratios at 4 wavelengths for a PSC measured on Jan. 19, 1995.

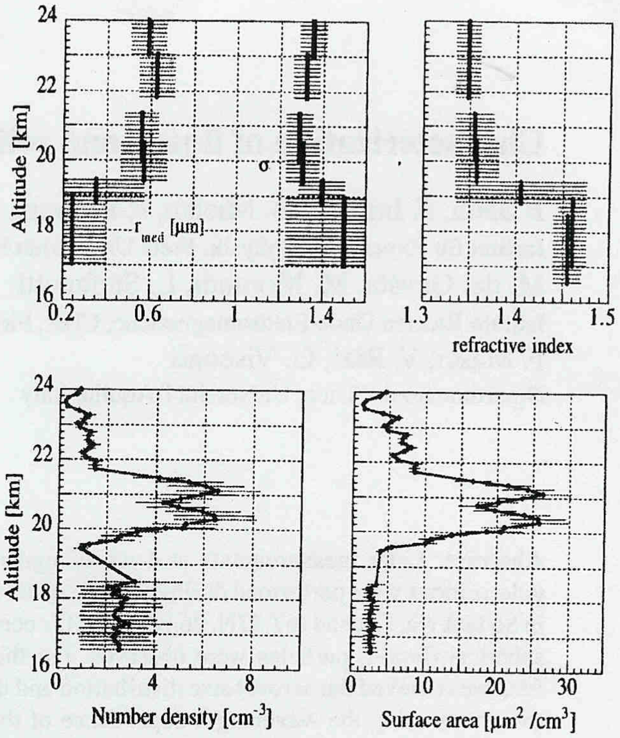

Fig 2: Parameters of the aerosol size distribution, r-median and width σ (upper left), refractive index (upper right), number density (lower left) and surface area density (lower right) for the PSC observed on Jan. 19, 1995.

indices between 1.33 and 1.38 yield backscatter coefficients that match the measured values within our experimental error limit of ± 10 %. Because the refractive index of water is 1.33, this result indicates a high water content of the particles as it would be expected for STS. The median radius of the particles was retrieved to (0.6±0.1) μm and the width of the size distribution was σ = 1.35±0.04. (Fig. 2). Although the real size distribution might be multimodal the description by monomodal parameters is a good approximation since the smaller particle mode (typical r_{med} < 0.1 μm) yield only a small contribution to the optical signal [Rizi et al.]. The surface area density of the PSC was determined to approximately 20 μm² / cm³. Below the PSC we found refractive indices between 1.42 and 1.46, and a median radius of (0.25±0.15) μm. This result is typical for sulfuric acid background aerosol. The local radio sounding (Fig. 3) shows that the temperature drops below the condensation temperature of NAT at 15 km altitude, but no PSC could be observed in the altitude range from 15 up to 19 km. At 19 km the temperature is below the temperature range where formation of supercooled ternary solution particles (STS) is very efficient. This is assumed 3-4 K below T_{NAT}. The existence temperatures used here were calculated using an arctic LIMS profile for the nitric acid content and a water vapor mixing ratio of 5 ppm.

2. Liquid and solid PSC's

On Jan. 12, 1995 we observed three kinds of PSC´s in differ-

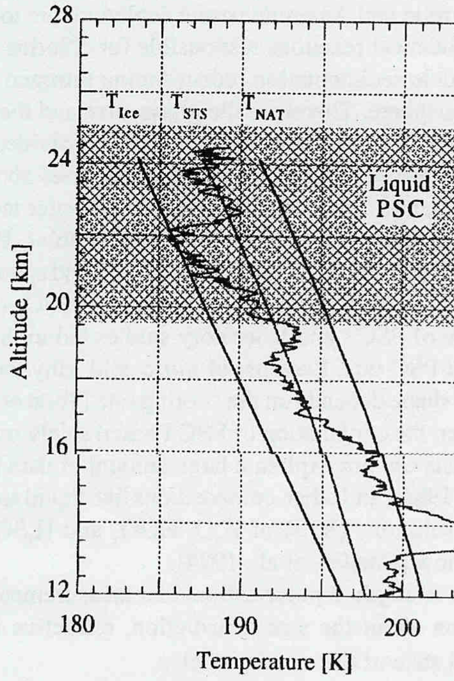

Fig 3: Local temperatures for Sodankylä at 19/01/95 and existence temperatures for NAT, STS and ice

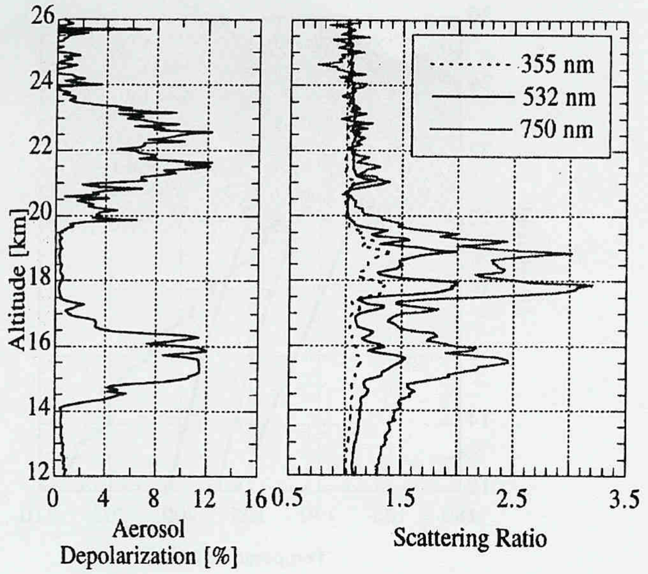

Fig 4: Aerosol depolarization and scattering ratios at 3 wavelengths for a PSC measured on Jan. 12, 1995.

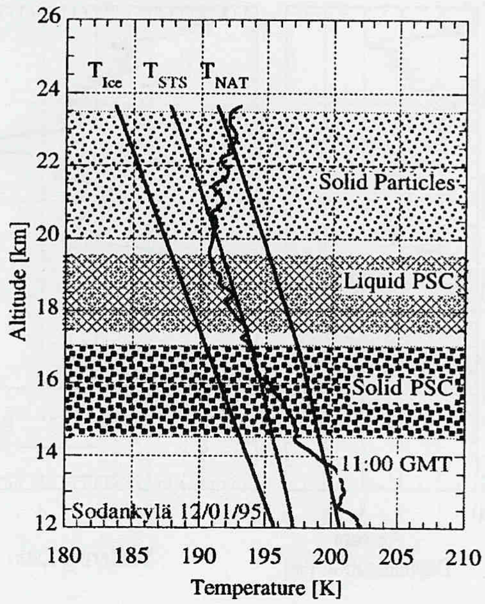

Fig 5: Local temperatures for Sodankylä at 12/01/95 and existence temperatures for NAT, STS and ice

ent altitude ranges, which can be distinguished by their backscatter and depolarization values (Fig. 4):

From 23.5 km down to 20 km we measured an optical thin layer of solid particles with a depolarization ratio of 5 %. From the small backscatter values indicating very low aerosol mass we conclude the presence of frozen background aerosol here. The temperature at this altitude was in the range of $T_{NAT} \pm 2.5$ K (Fig. 5).

A liquid PSC is present in the height range between 19.5 and 17.5 km where the temperature is more than 3 K below T_{NAT}. Mie calculations of the parameters of the aerosol size distribution yield a refractive index in the range from 1.33 - 1.4 and a median radius of the particles between 0.5 and 0.7 µm.

At 17 to 15 km a third layer with scattering ratios of about 1.6 and aerosol depolarization of 10 % is seen which are typical values for PSC Ia.

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 the particles consist of NAT here.

3. Coexistience of different PSC types

On Dec. 21, 1994 we could measure a layer of frozen background aerosol extending from 20.5 up to 26 kilometers altitude with an aerosol depolarization of about 2% and a low scattering ratio in the 532 nm parallel polarized channel (Fig. 6). The temperature is below T_{NAT} at altitudes above 18.5 km (Fig. 7) where no aerosols are detected.

Within this layer we observed a PSC from 22 to 24 km altitude. The structures of the signals at both polarizations are not corresponding well. We therefore believe that a layer of large liquid particles coexists within a layer of small solid particles. The local radio sounding ends at 21 km altitude, so we don't have information about the temperature within the liquid PSC layer.

Discussion

The altitude range were PSC type Ib were observed does not correspond to the existence temperature for NAT. It corresponds much better to the formation temperature for STS. The depolarization measurement shows spherical particles and the refractive index of 1.35 - 1.4 indicates a high water content, in contradiction to a refractive index of 1.5 expected for NAT. These observations show that PSC type Ib clouds consists of liquid supercooled ternary solution particles (STS).

The presence of solid particle layers is not correlated to the NAT-existence temperature either. The airmass thermal history plays an important role in the formation of solid particles in the stratosphere, but the real formation mechanism is not well understood yet.

Different freezing mechanisms for background aerosol into sulphuric acid hydrates have been discussed recently. 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 from laboratory studies that the formation of solid particles is only possible when the temperature falls below the ice frostpoint. Due to the uncertanties of the airmass thermal history both scenarios are consistent with our observations.

Liquid and solid particles can form independently within the same airmass and then coexist in a mixed PSC. The measure-

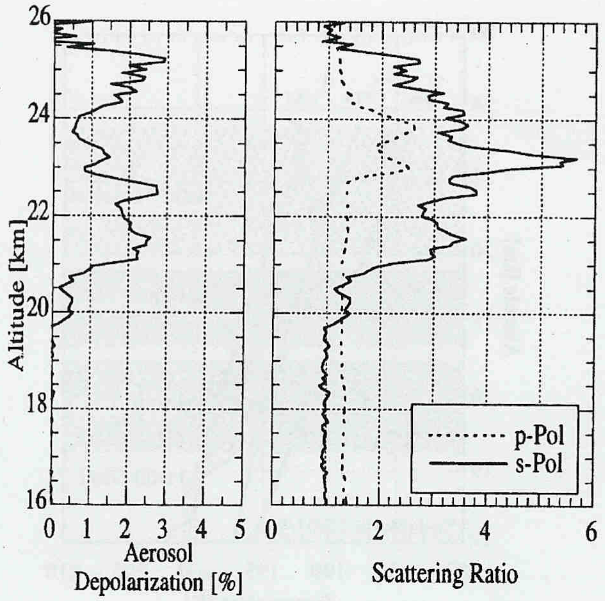

Fig 6: Aerosol depolarization and scattering ratios of the parallel and cross polarization for a PSC measured on Dec. 21, 1994.

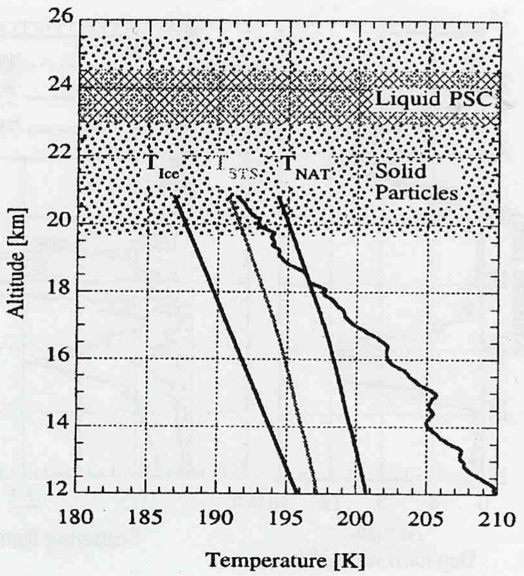

Fig 7: Local temperatures for Sodankylä at 21/12/94 and existence temperatures for NAT, STS and ice

ment of the PSC on Dec, 21 shows that even liquid particles can form in an airmass containing solid condensation nuclei. Either the condensation of nitric acid on the exisiting particles is hindered, or the formation of STS particles is much faster than further condensation on the existing particles.

For modelling the heterogenous chemistry on PSC particles we suggest the use of a microphysical model describing the temperature dependant condensational growth of liquid ternary solutions to describe the surface area of liquid PSC. Significant growth of liquid PSC is observed 3-4 K below T_{NAT}. T_{NAT} can still be taken as a threshold temperature for solid PSC formation, but additional criteria with respect to the airmass thermal histories should be added since temperatures below T_{NAT} are necessary, but not sufficient for solid particle formation.

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