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Microlayers of solid particles observed by lidar at Sodankylä during SESAME

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INTRODUCTION

The physical condition of polar stratospheric aerosols is of great importance both for the modelling of surface chemistry reactions and for the understanding of particle production and evaporation in the polar vortex. The particles can be either liquid, supercooled liquids or solid material at different heights and temperatures. Since a solid particle can survive much longer when temperature rises above the freezing point, whereas liquid particles will evaporate quickly at temperatures above the condensation temperature, the knowledge of the physical state is an important parameter to estimate the contribution to heterogenous chemistry of the different aerosol types observed.

MEASUREMENTS

Lidar measurements were performed from October 1994 to March 1995 in Sodankylä, Finland (67.4°N, 26.6°E) during the European SESAME Arctic campaign. The lidar system was designed to measure the size distribution and physical condition of stratospheric aerosols by using a Nd:YAG Laser at 355, 532 and 1064 nm together with a Titanium Sapphire Laser at 750 nm and simultaneous

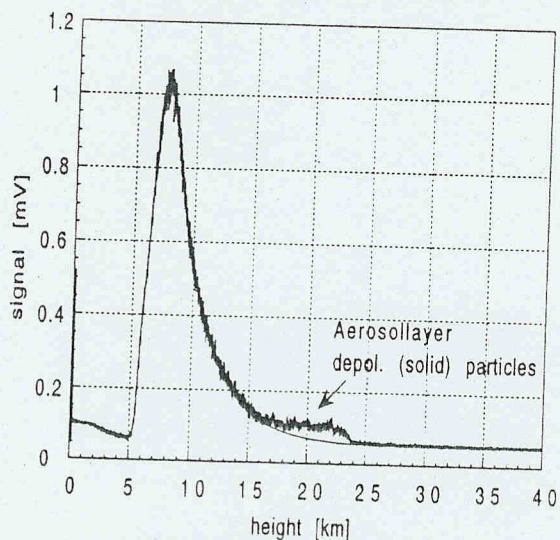


Fig. 1: 532 nm cross polarized signal

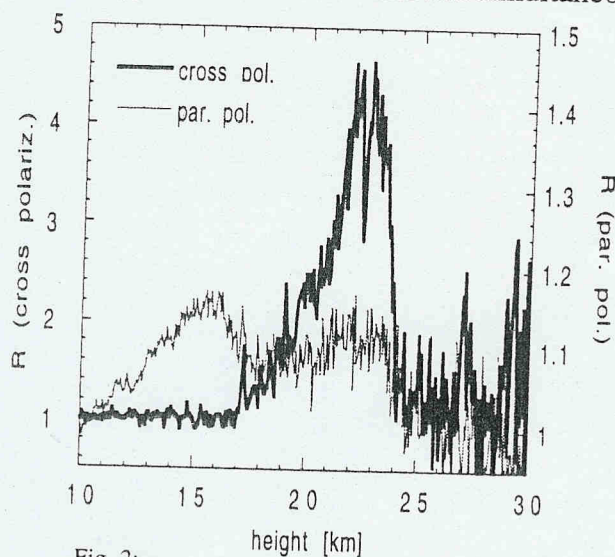


Fig. 2:
Scattering ratios for parallel and perpendicular signals.

detection at all wavelengths. Additional detection channels acquired the cross polarized signals at 532 and 750 nm. In the 532 nm detection channel a 0.15 nm FWHM bandwidth filter was used to block the rotational Raman lines which allows a suppression of the Rayleigh signal contribution to only 0.36% in the cross polarized channel. This signal is therefore extremely sensitive for nonspherical particles.

In Fig. 1, a measurement of Jan. 2, 1995 is shown, where the 532 nm depolarized signal shows a

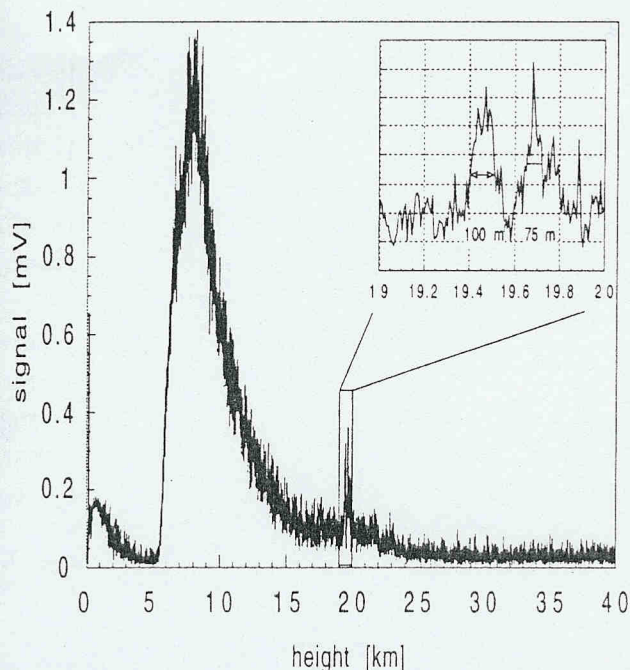


Fig. 3:
Very thin layers of particles appear in the 532 nm cross polarized signal.

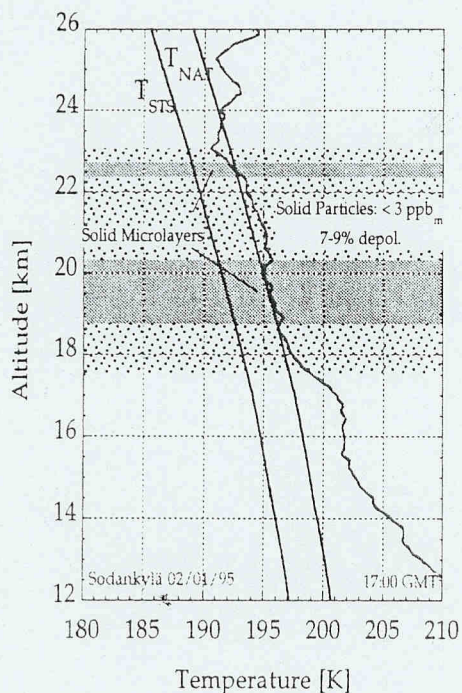


Fig. 4:
Temperature profile by FMI radiosounding for Jan. 2, 1995

significant deviation from the Rayleigh scattering signal between 16 and 24 km altitude. Since liquid particles can be assumed spherical and therefore would show no depolarized backscatter signal, these particles can be identified as solid, nonspherical particles. This can be confirmed by a comparison of the scattering ratios for parallel and perpendicular polarized signals, where the liquid sulfuric acid aerosol from the Mt. Pinatubo volcanic eruption can be seen on the parallel signal between 10 and 16 km without any contribution to the cross polarized signal. The aerosol depolarization ratio of the upper layer can be retrieved from the ratios of R_{par} and R_{perp} to max. 10%. The aerosol mass within the layer is estimated to be $< 3 \text{ ppb}_m$.

While this weak layer of solid particles could be measured for many hours and on several days during January, episodes of additional particle growth within this layer could be observed during a few hours. An example is shown in Fig. 3 where very thin layers of solid particles seem to grow in the coldest part of the broad but weak layer observed before.

A temperature profile for this day is shown in Fig. 4. The temperature is close to, but not necessarily below the NAT-existence temperature in the height range where the broad aerosol layer is observed. We assume this layer to consist of sulfuric acid tetrahydrate (SAT) particles.

The microlayers could either consist of NAT-particles which are formed where the temperature drops below the critical temperature for NAT-formation or even ice particles when temperatures were below the ice frostpoint in lee waves, shortly before the air reached Sodankylä. As winds at 700 hPa in Norway indicate the possibility of lee waves this day, temperatures can fall below the ice frostpoint in small height regions.

The airmass thermal histories for Jan. 2 are shown in Fig. 5.

At 435 and 475 K the temperature of the airmass was close to the ice frostpoint approximately 100 hours before reaching Sodankylä and not warmer than the SAT-melting temperature of 210 to 215 K afterwards.

As SAT-formation in the stratosphere is possible when the temperature is below the ice frostpoint [Koop et. al, 1995], or suggested when the air is cooled below 192 to 194 K and warmed up to more than 198 K afterwards [Tabazadeh et. al, 1995] we would expect a layer of solid SAT particles between 18 and 20 km which is in good agreement with our observations. The temperature history for the 550 K level, according to 24 km altitude shows, that the temperature were not below the ice frostpoint and exposed to the SAT-melting temperature 50 hours before reaching Sodankylä, so the formation of solid particles was not possible in 24 km and no aerosol is observed.

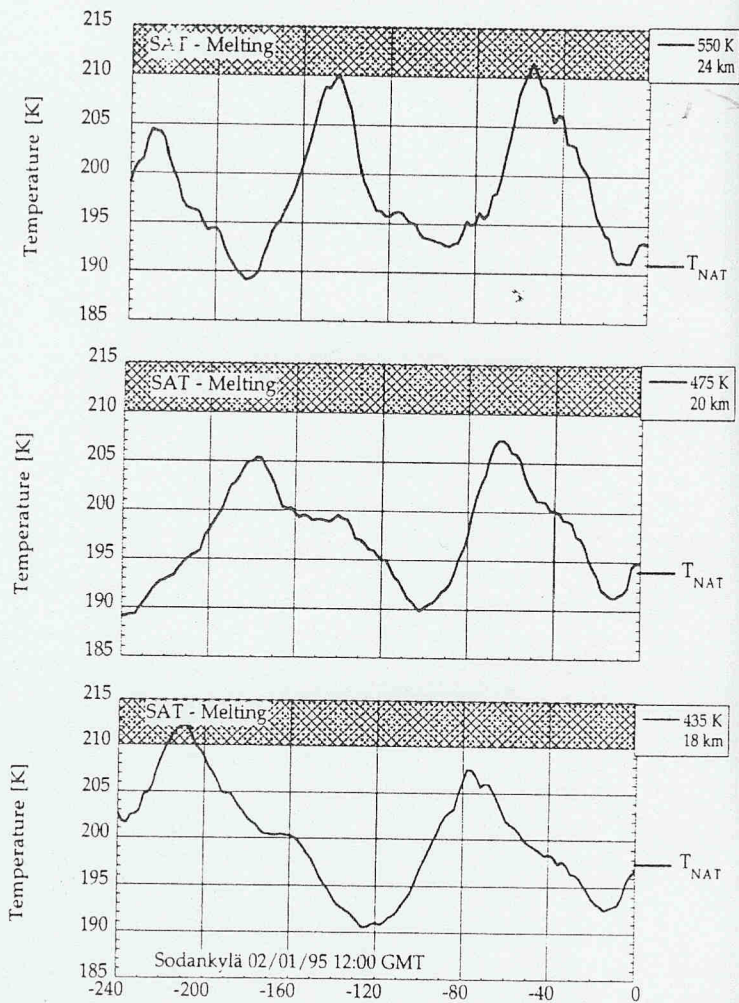


Fig. 5:
Airmass thermal history from ECMWF for Sodankylä at Jan. 2, 1995 12:00 UT

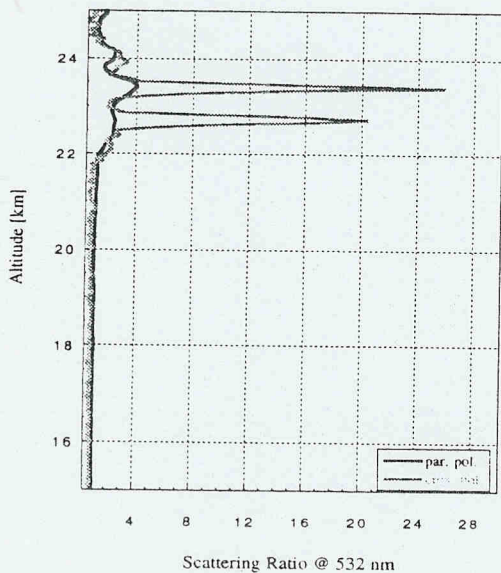


Fig. 6:
Scattering ratios for parallel and perpendicular signals on Jan. 15, 1995.

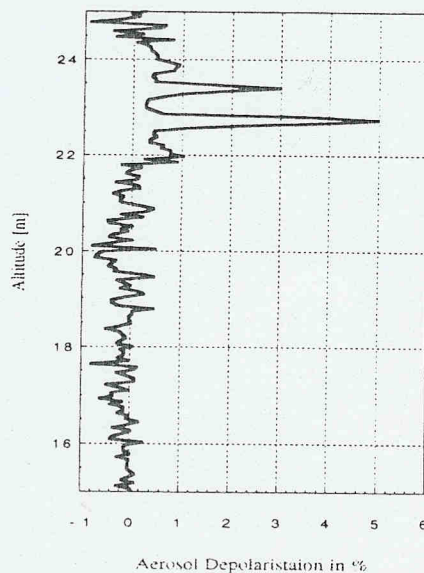


Fig. 7:
Aerosol depolarization for a microlayer on Jan. 15, 1995

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In Fig. 6 an observation of solid microlayers from Jan. 15, 1995 is shown. The microlayers here are seen within an PSC consisting of spherical particles as the aerosol depolarization measured is below 0.5%, while the depolarization in the microlayers is about 5%. Temperatures for this day (Fig. 8) are below T_{NAT} from 17 to 19 km and from 21 km to the burst height of the balloon. The PSC can only be observed in an altitude >22 km where the temperatures fall below the existence temperature for supercooled ternary solutions. On Jan. 15 the air was exposed to the SAT-melting temperature at all height levels before reaching our lidar and was not below 194 K afterwards, so we would not expect solid particles on this day. The observed solid microlayers can be explained by meso- or microscale temperature variations in lee waves over the scandinavian mountains.

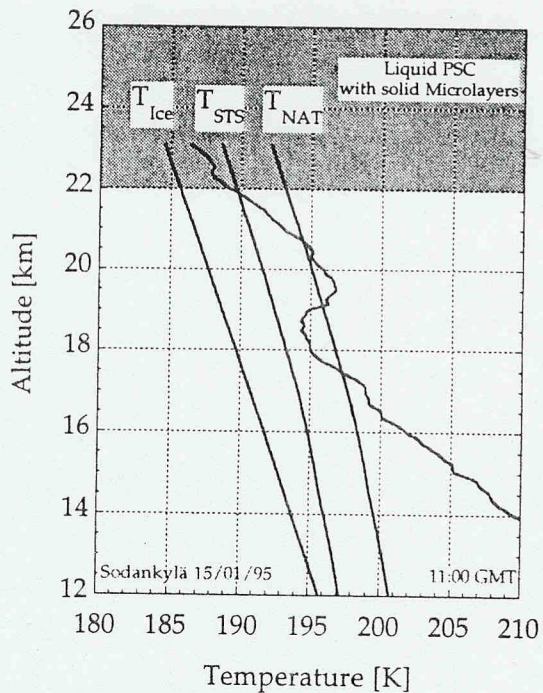


Fig. 8:
Temperature profile by FMI
radiosounding for Jan. 15, 1995

CONCLUSIONS

By using 2-polarization detection the sensitivity for lidar measurements of solid aerosol particles can be greatly enhanced.

This allows observation of very low concentrations of solid particles as for example frozen background aerosol, transformation of supercooled droplets into crystals and condensational particle growth. By correlation of these observations with temperature the necessary conditions for formation of the various PSC types can be observed.

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