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## LIDAR OBSERVATION OF PSCs IN THE ARCTIC AND ANTARCTIC

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### ABSTRACT

Polar Stratospheric Clouds and aerosols are here reported which do not clearly corresponding to present interpretations. They were monitored in the Antarctic and in the Arctic from 1989 to 1995 and therefore refer both to the pre-Pinatubo, the Pinatubo and after Pinatubo period. Such cases comprehend non depolarizing PSCs and warm depolarizing aerosols. Non Depolarizing PSCs have been observed over Dumont d'Urville, Antarctica during the POLE experiment (1989-today) and over Sodankyla during SESAME. For the case of Sodankyla particle size evaluation for non depolarizing PSCs were carried out with interesting results. An index of refraction has been used for Mie calculations which may well correspond to the one expected for a diluted ternary solution of  $H_2O/H_2SO_4/HNO_3$ .

### INTRODUCTION

Multispectral depolarization LIDARs have been used in the Arctic and in the Antarctic by our groups for monitoring and studying optical and microphysical properties of Polar Stratospheric Clouds.

In the Antarctic, at Dumont d'Urville, the French-Italian project POLE (Polar Ozone LIDAR Experiment) has developed since 1989. Continuous monitoring of the Antarctic stratosphere has been accomplished by means of backscatter depolarization LIDARs. Stratospheric profiles are available for every day when LIDAR measurements were possible.

The LIDARs used are described in literature [Sacco et al., 1989; Stefanutti et al., 1991a and 1991b]. The importance of the Antarctic research lies in the long period of monitoring at DDU, which enabled the creation of one of the largest self-service PSCs data banks (South Pole Data Bank of PNRA, [http address www.area.fi.cnr.it/default.html](http://www.area.fi.cnr.it/default.html)).

In the Arctic, at Sodankyla (northern Finland) measurements have been carried out regularly during EASOE and SESAME with multispectral LIDAR. During EASOE the available wavelengths were: 355 nm, 532 nm, 750 nm, 850 nm, while we used the 1064 nm instead of 850 nm wavelength during SESAME

For the EASOE LIDAR measurements the particle size of the Mt. Pinatubo aerosol was computed by different methods [Del Guasta et al. 1994, and Stein et al. 1994].

LIDAR data, at different wavelengths, have been inverted using a modified Klett method or a numerical iterative method to determine the optical parameters. Averaged values of the extinction to backscattering ratios, and their relative distribution, have been obtained for the different wavelengths [Del Guasta et al., 1994]. Also the backscatter coefficients at the different wavelengths have been computed by a different technique [Stein et al., 1994] which permitted to evaluate changes in the size distribution inside the Mt. Pinatubo layer. Mie codes have been run, assuming a given index of refraction for HNO<sub>3</sub> solution. Monomodal size distributions was assumed for the aerosols. From the computation average size distributions were derived, which indicated a mean size of 0.1 microns. During EASOE period no evidence of PSC events were noticed except for two short lasting events.

The same technique has been applied to non depolarizing PSCs during SESAME.

#### ANTARCTIC NON DEPOLARIZING PSCs AND UNEXPECTED DEPOLARIZATION IN WARM STRATOSPHERIC AEROSOLS

Since LIDAR operation at Dumont d'Urville (66°S, 144°E) started prior of the Pinatubo perturbation, it has been possible to study PSCs as well in a time of non perturbed and in strongly perturbed stratosphere. In this paper we will analyze in particular non depolarizing Polar Stratospheric Cloud events, which were relatively frequent. In the Antarctic a very interesting case was monitored for many days during 1990. In figure 1 such case is shown.

Such cloud was observed over Dumont d'Urville for over 8 days during July 1990. Figure 2 shows the scattering ratio and depolarization, defined as ratio of the depolarized signal versus the total signal the total signal. The measurement was carried out on July 11, 1990, at 532 nm, the temperature profile obtained by Rawinsonde is also plotted.

A depolarization peak is evident for a low layer, probably a high cirrus between 10 and 12 km, but there is no sign in the change of the depolarization ratio for the high PSC, ranging between 17 and 24 km. The associated temperature profile indicates a very low temperature at the same altitude, with value of the order of 190°-193°K. The scattering ratio is relatively high, with a peak close to 5.

Mishchenko et al. 1994, Flesia et al, 1994 have shown that particles depolarize relatively high even if their form deviates very little for sphericity and only very small particles smaller than 0.1 micron would not depolarize if illuminated with visible light.

The fact that there is no change in the depolarization ratio, inside and outside the PSC, induces to believe that such cloud should be formed by liquid particles.

Recent theories [Carslaw et al., 1994, Drdla et al, 1994, Middlebrook et al., 1994, Molina et al, 1994] assume the possibility of supercooled particles formed by a ternary system of H<sub>2</sub>O/H<sub>2</sub>SO<sub>4</sub>/HNO<sub>3</sub>.

Unfortunately single wavelength analysis does not permit to extend the analysis to particle size and index of refraction.

If PSCs are most frequently in such state and grow on background diluted  $\text{H}_2\text{SO}_4$  liquid aerosol cores with uptake of  $\text{H}_2\text{O}$  and  $\text{HNO}_3$ , it may be comprehensible why during the Pinatubo period, it was impossible the discrimination between PSCs and volcanic aerosols.

In figure 3 a yearly plot, for 1992, of the scattering ratio for the stratospheric Pinatubo layer is shown. Figure 4 shows the equivalent time plot for the depolarization ratio. From that plots it appears that during the PSCs event period, July-August corresponding to the Julian days 180-245, no evident change occurred in the depolarization, except for few events above 20 km, and for intense clouds in the very low stratosphere, around the tropopause, which form regularly in July and August. In the Pre and Post-Pinatubo periods PSCs were frequently observed at the whole altitude range from 14 to 25 km as shown in figures 5 and 6.

During all of September 1992 PTU sounding over Dumont d'Urville indicated a warm stratosphere, with temperature in the order of 210°K. Jet depolarizing layers appeared frequently during that month. One example is shown in Figure 7. Such a case seems much more difficult to be interpreted. Depolarization peaks could be due to PSCs particles which were melting and coming from colder regions. Unfortunately no back-trajectories were available and it was not possible to confirm such hypothesis.

This hypothesis is not very credible as the stable high stratospheric temperature over DDU induces to believe that the Station was at that time relatively far from the vortex edge. As the scattering ratio presents negligible changes at the depolarizing levels one would assume that the particles should be small, not inducing substantial change in the backscattering profile, or one may assume the presence of a few large non spherical particles embedded in small spherical droplets.

#### ARCTIC NON DEPOLARIZING PSCs

During SESAME, from late December 1994 to the end of January 1995 PSCs were measured very frequently.

Stratospheric temperatures were low during the whole period. Both depolarizing and non depolarizing PSCs were measured. Sometimes during the same day depolarizing and non depolarizing layers were detected, as shown in figure 8.

A first attempt to evaluate particle size of non depolarizing PSCs was carried out. Calculations were carried out for the case of January 19, 1995, that correspond to a long-lasting non-depolarizing PSC, figure 9 and 10. The LIDAR signatures were inverted to obtain the backscatter to extinction ratio at 355 nm, 532 nm, and 750 nm. Histograms of the Extinction to backscatter ratio at the different wavelengths were obtained, figure 11.

In order to perform Mie computation information on the refractive index of a ternary solution  $\text{HNO}_3/\text{H}_2\text{SO}_4/\text{H}_2\text{O}$  is necessary. Such information is not obtainable in the literature. A first attempt to evaluate the index of refraction of STS has been carried out. Binary solutions of  $\text{HNO}_3$  and  $\text{H}_2\text{O}$  have been prepared and their index of refraction at ambient temperature has been experimentally measured [M. Pantani, 1996]. The Lorenz-Lorenz formula [Steel, 1981], which contains the index of refraction of the acid at 300K and the density of the acid at the stratospheric temperature  $T$ , was used to compute the index of refraction at 190K. It was therefore necessary to interpolate the density for the  $\text{HNO}_3$  solution, with concentration greater then the 20% of nitric acid, from 230K (at such temperature the data exist) to the temperature of 190K by means of a linear interpolation.

STS aerosol optical parameters were computed by means of a Mie code. Mie simulations were performed for different STS solutions and for the different wavelengths. In figure 12 the extinction to backscatter ratio, for 50% HNO<sub>3</sub> acid solution, are plotted as a function of the median radius and of the width of a lognormal distribution. The plots are carried out for 355 nm, 532 nm, and 750 nm wavelengths. Using the method of the minimum of  $\chi^2$  it resulted that the best matching between the computed and the experimental extinction to backscattered ratio was, for the index of refraction, in the range  $1.41 < n_{355} < 1.47$ ,  $1.39 < n_{532} < 1.43$ ,  $1.38 < n_{750} < 1.42$ . The mean radius is comprised between 0.65 and 0.9  $\mu\text{m}$  and a width of the size distribution between 1.1 and 1.4, for an acid concentration between 30% and 50%, as shown in table I.

Table I

| Acid Concentration | mean radius | distribution width |
|--------------------|-------------|--------------------|
| 30%                | 0.8-0.9     | 1.25-1.4           |
| 50%                | 0.65-0.75   | 1.1-1.15           |

The index of refraction derived from the minimum of  $\chi^2$ , corresponds to a total acid content of about 40%. The total fraction of HNO<sub>3</sub> in weight corresponds therefore to 39.7%, while the amount of H<sub>2</sub>SO<sub>4</sub> should not be larger than only 0.3% [M. Pantani, 1996]. This indicates a strong uptake of water and nitric acid from the background aerosols to form the liquid PSC and consistent with theoretical and modeling results [Carslaw et al 1994].

It is important to note that on the 19 January, PSC was liquid, also if temperature at cloud level was between 190 and 188 K for the whole day. Thus, liquid droplets were detected well below expected NAT freezing point and close to water frost point. This results are well in agreement with the present interpretation of liquid STS PSCs. The temperature time history for the air parcels at 475K are reported in figure 13 and show a relatively rapid cooling of the parcels in the last 60 hours from temperature above the SAT melting temperatures down to temperatures close to the ice freezing in the last 2 hours (190K).

## CONCLUSIONS

The evaluations for PSCs particle size evaluation by means of LIDAR are still in a preliminary stage, but interesting results have been acquired. Such results are in agreement with the more recent theories of PSCs formation. The technique here adopted has shown to be valid both for volcanic stratospheric aerosols, as shown for the Mt. Pinatubo case, as for the case of non depolarizing PSCs. Multispectral depolarization LIDAR technique has shown to be a powerful tool for atmospheric research. In the next future the same procedure will be applied to and tested for depolarizing PSCs. Depolarization is caused by the presence of non-spherical particles, but if the results should be consistent, this could induce to believe that depolarizing particles may be embedded in a larger number of liquid particles.

## Acknowledgments

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Figure

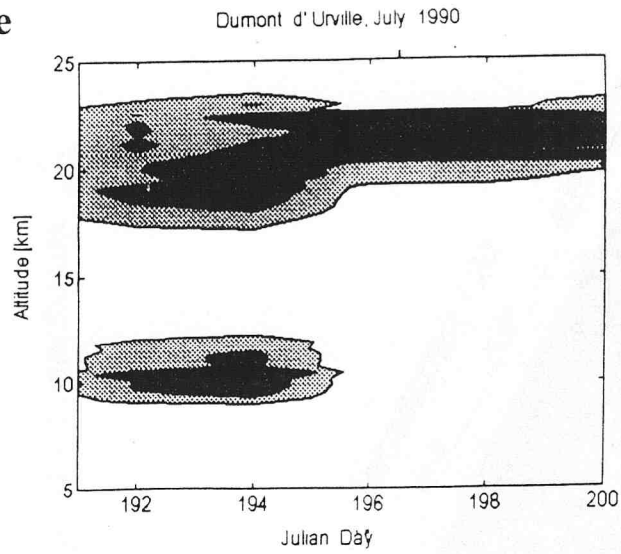


Figure 1: Non depolarizing PSC event at Dumont D'Urville on July 1990

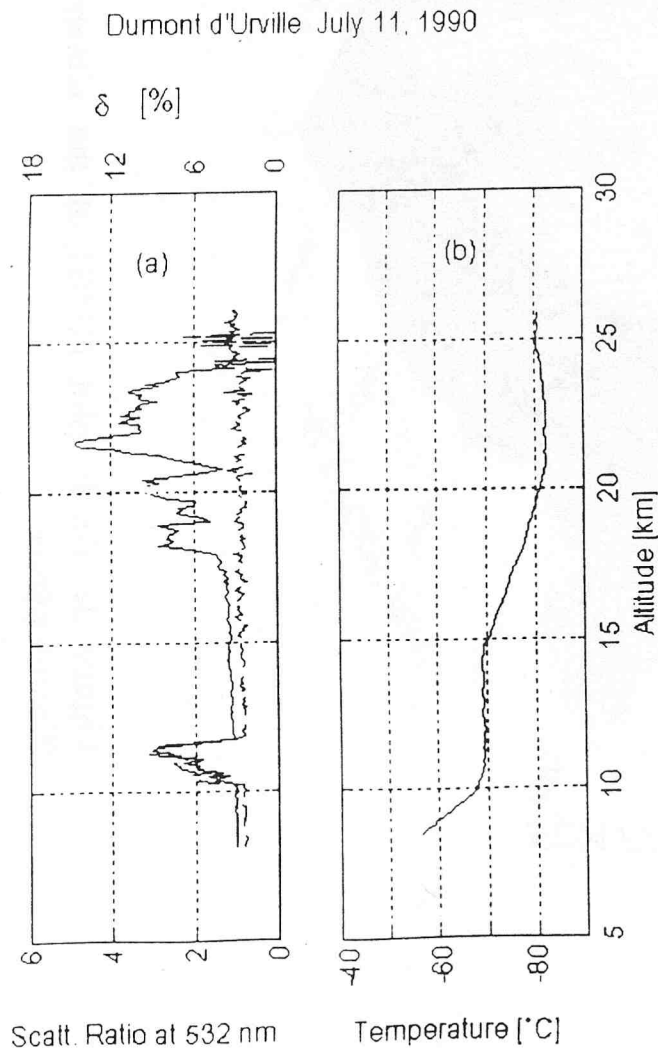
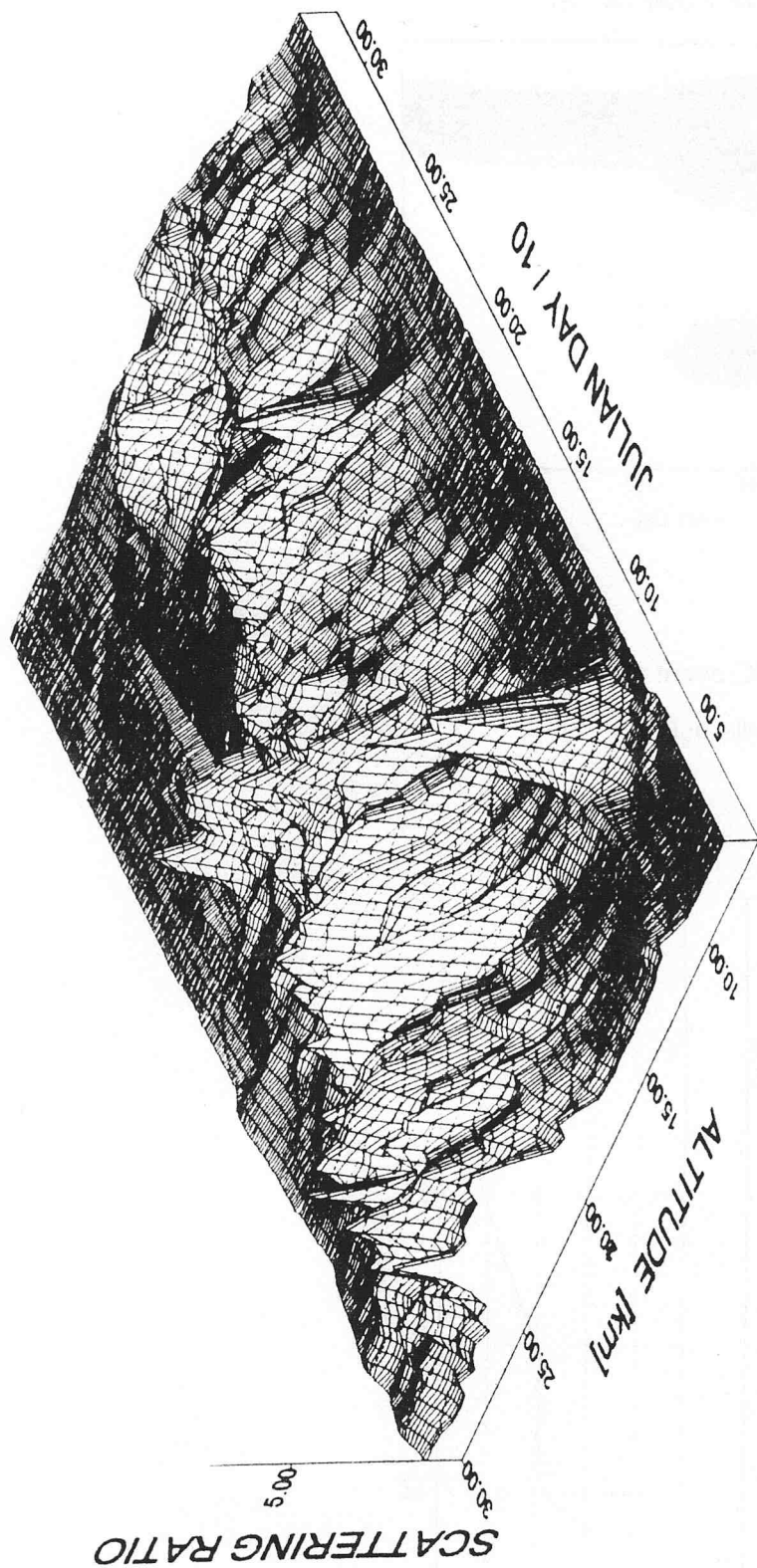
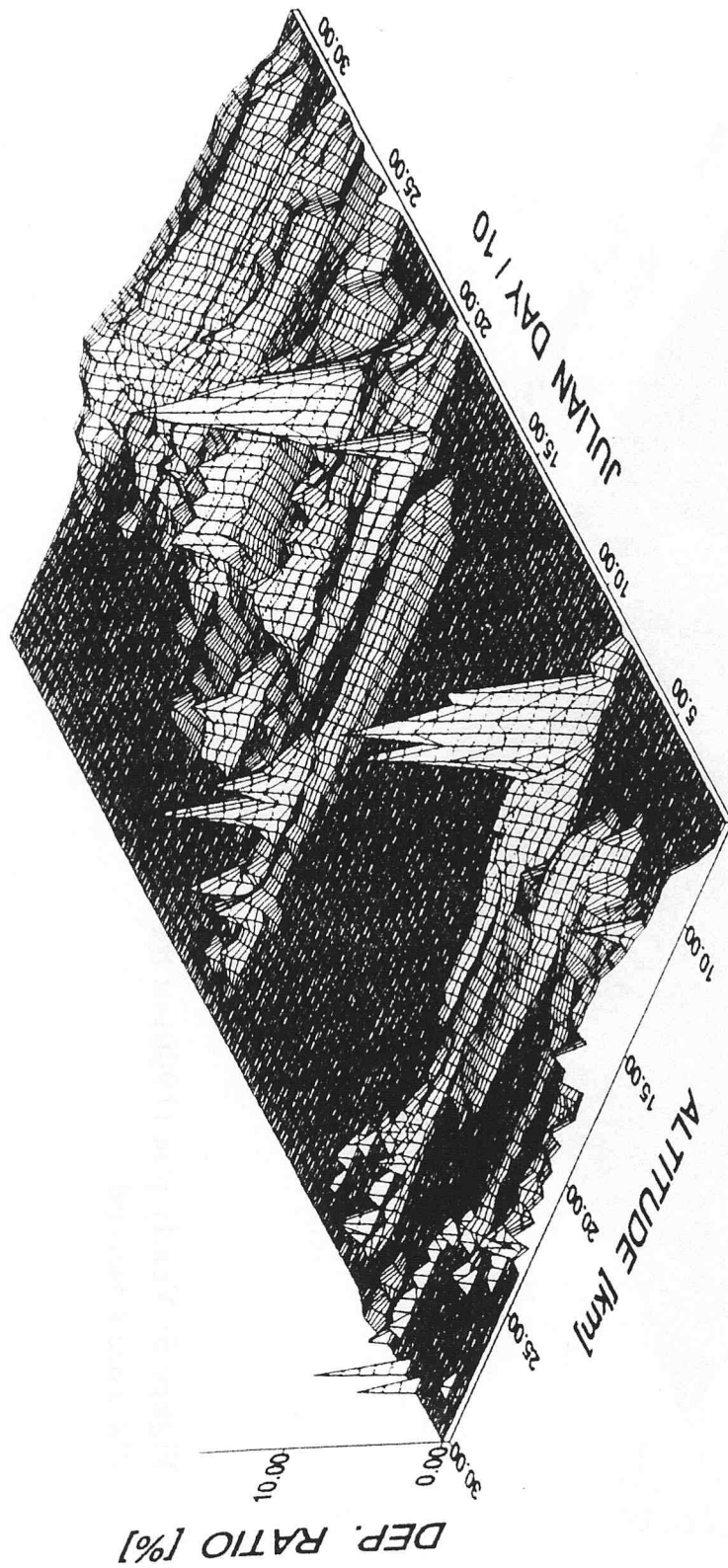


Figure 2: Scattering ratio and depolarization for the PSC observed at DDU on July 1990



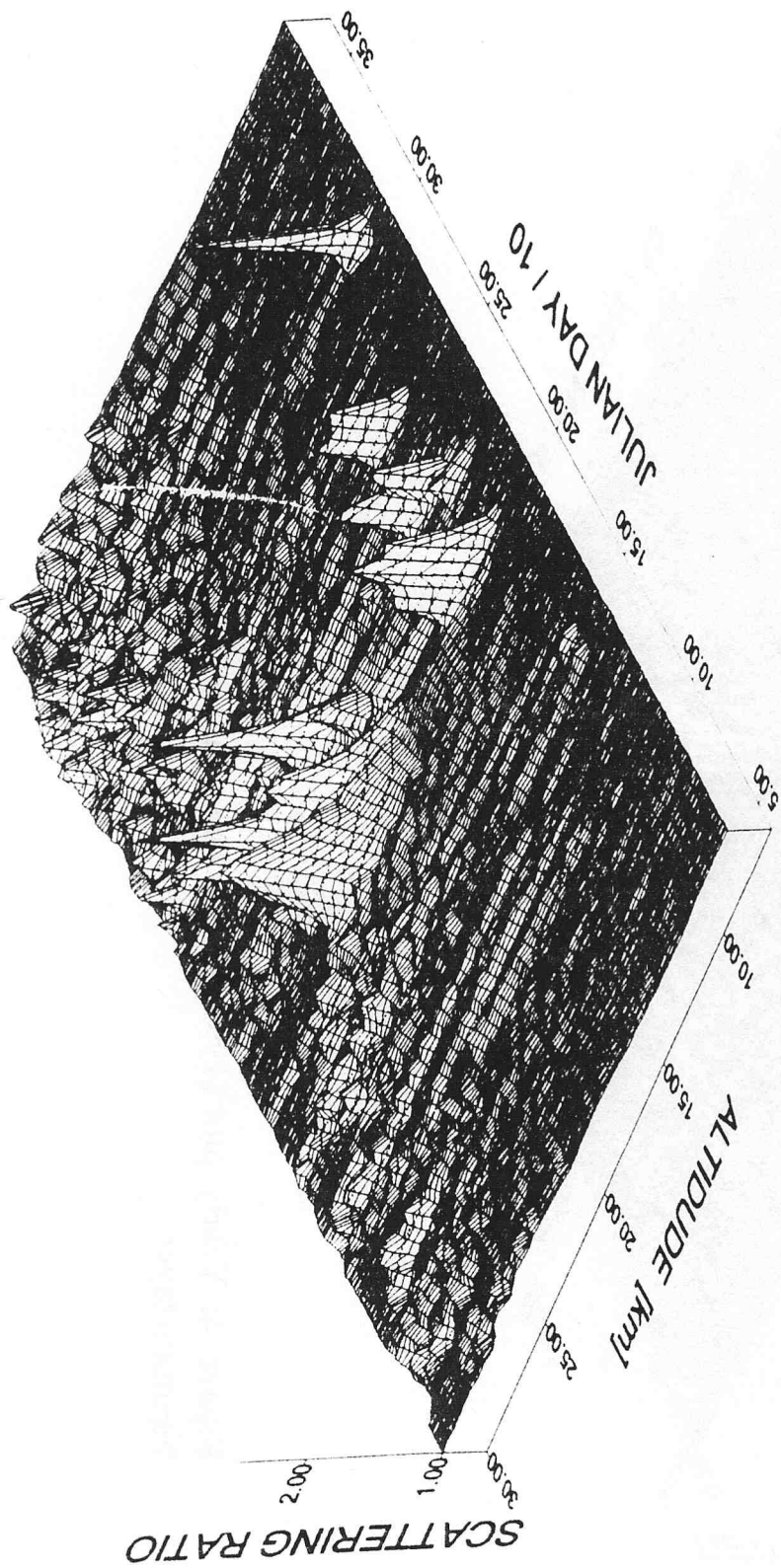


**Figure 3:** Yearly plot (1992) of the scattering ratio for the stratospheric Pinatubo layer

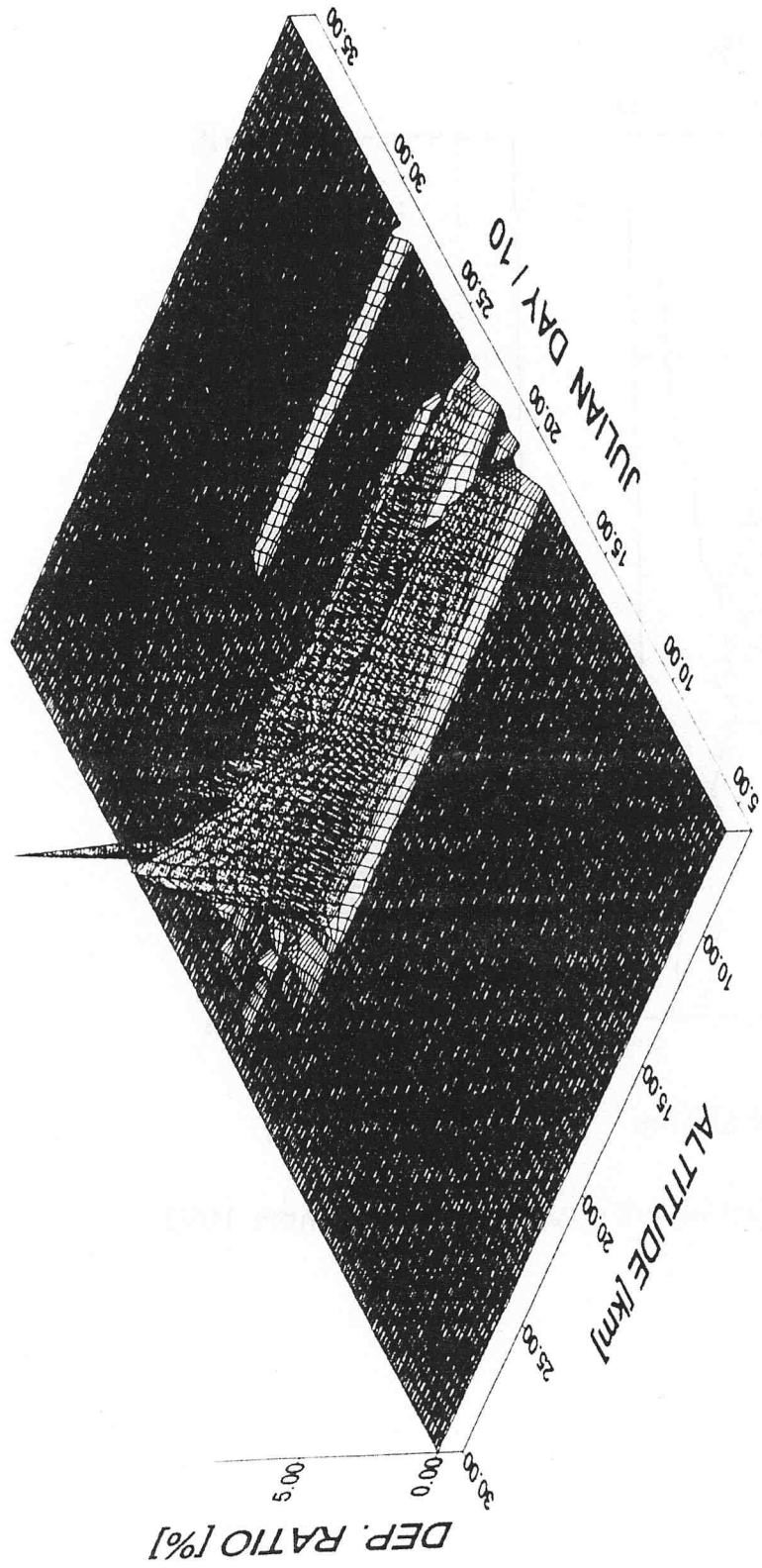


**Figure 4:** Yearly plot (1992) of the depolarization ratio for the stratospheric Pinatubo layer



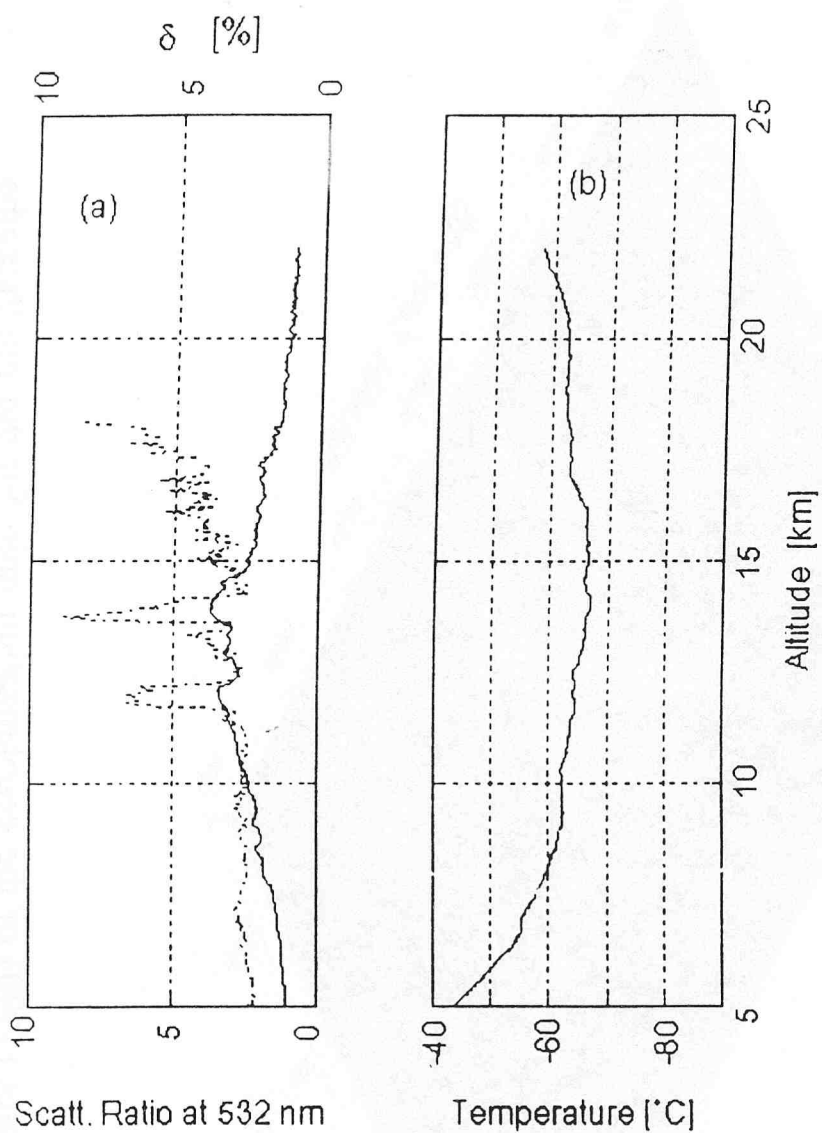


**Figure 5:** Yearly plot (1990) of the scattering ratio for the pre-Pinatubo period. PSCs are observed.



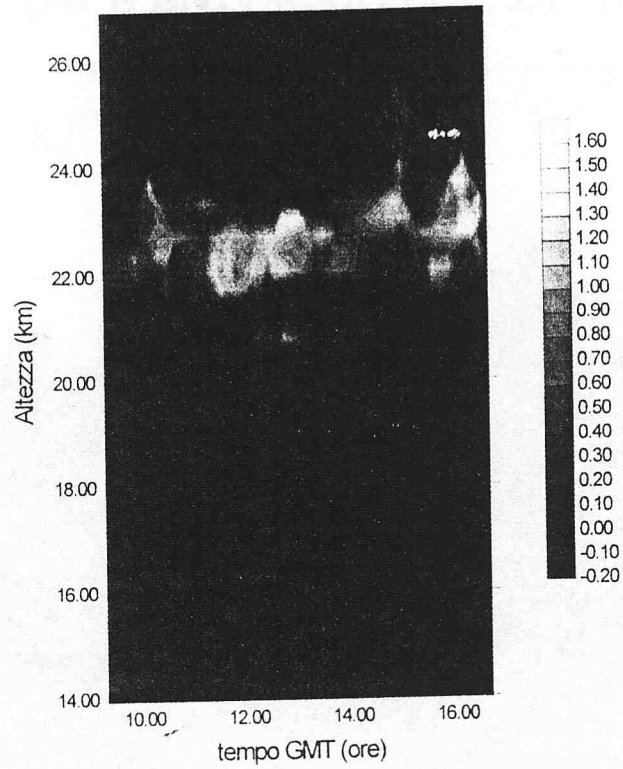
**Figure 6:** Yearly plot (1990) of the depolarization ratio for the pre-Pinatubo period. PSCs are observed.

Dumont d'Urville September 9, 1992

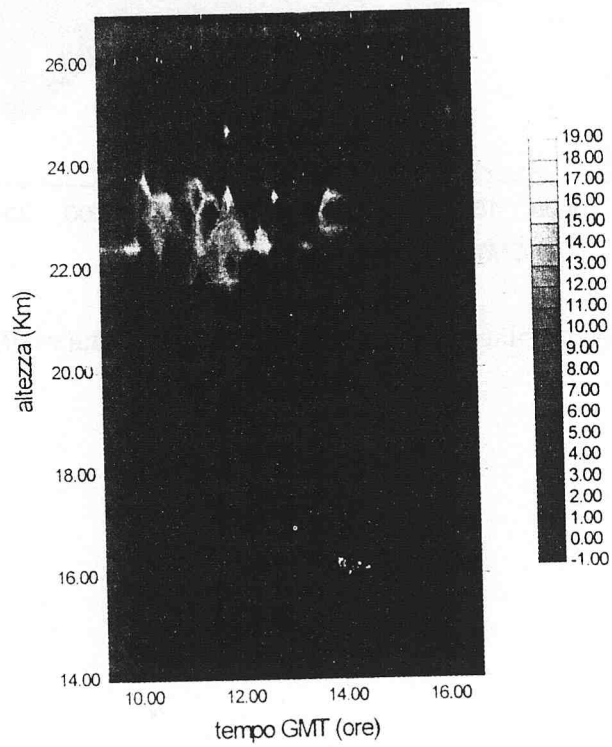


**Figure 7:** Jet depolarizing layers observed on September 1992

Sodankyla 15/01/95, beta\*1E7

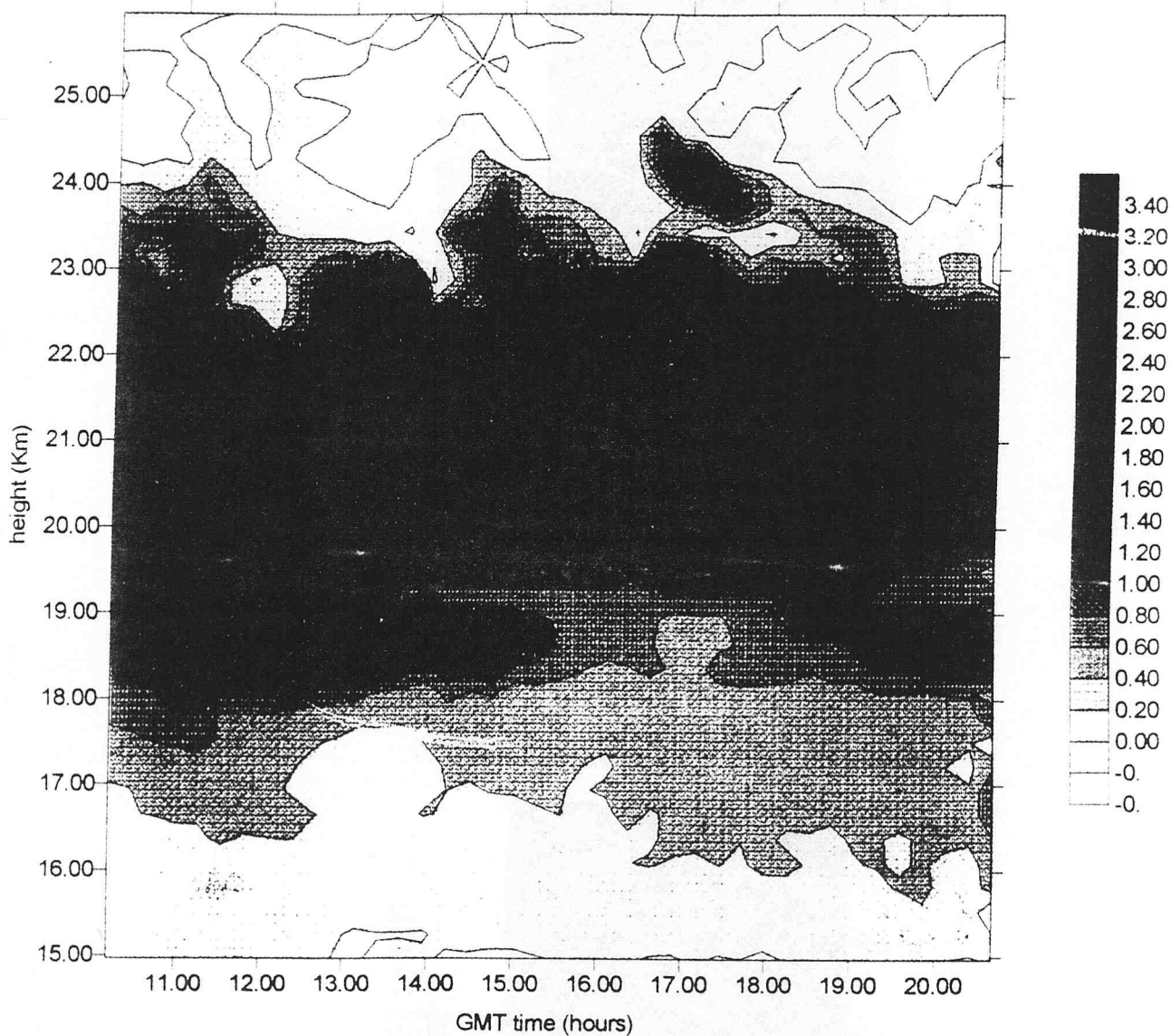


Sodankyla 15/01/95, depolarizzazione %



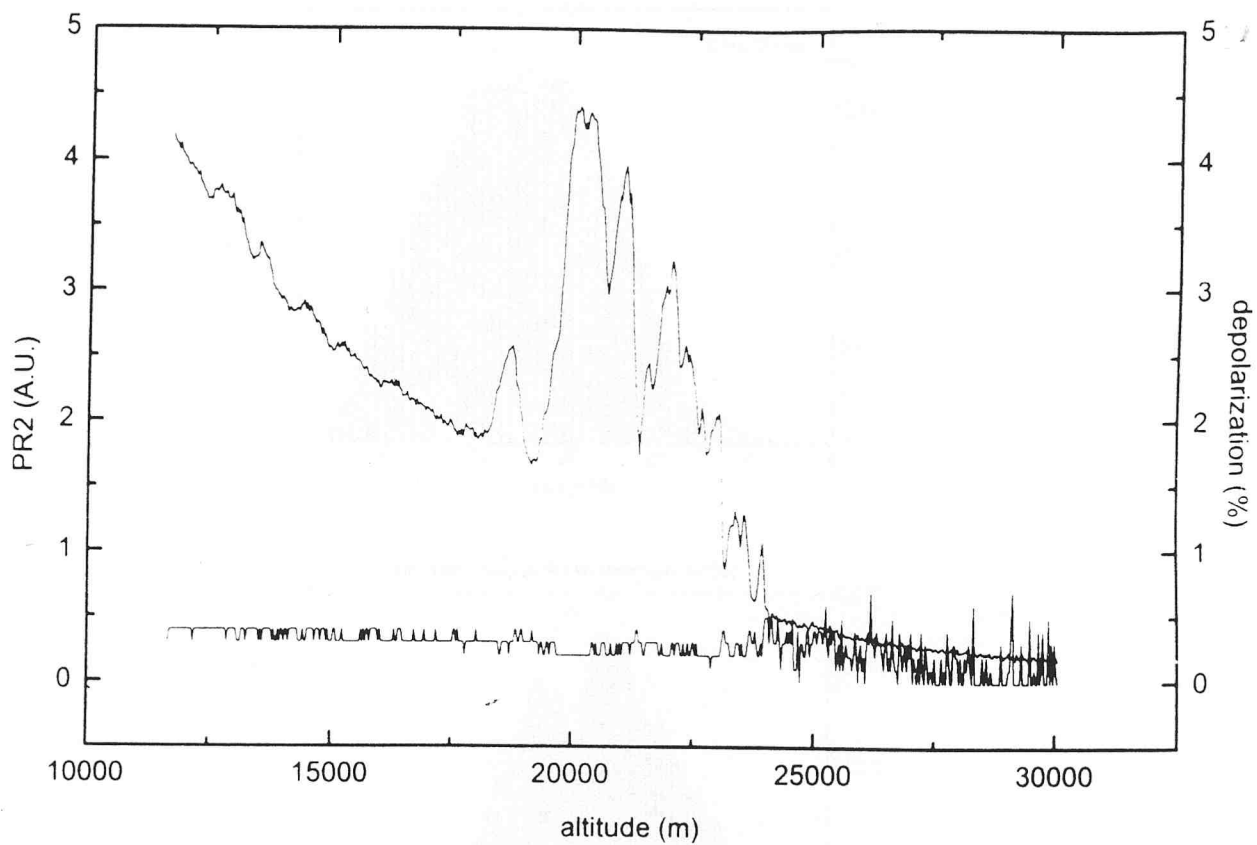
**Figure 8:** Depolarizing and non depolarizing PSC observed on January 15, 1992 at Sodankyla

# PSC evolution, 19/1/95 Sodandyla (FIN)



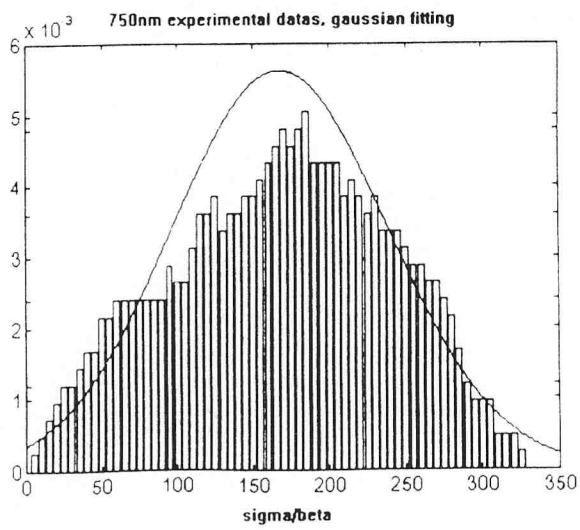
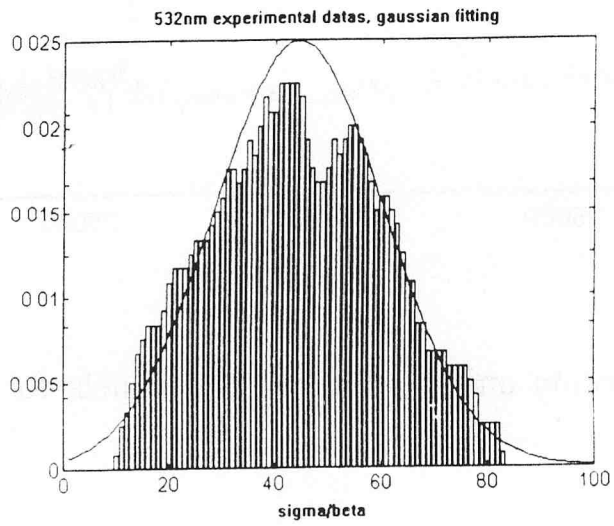
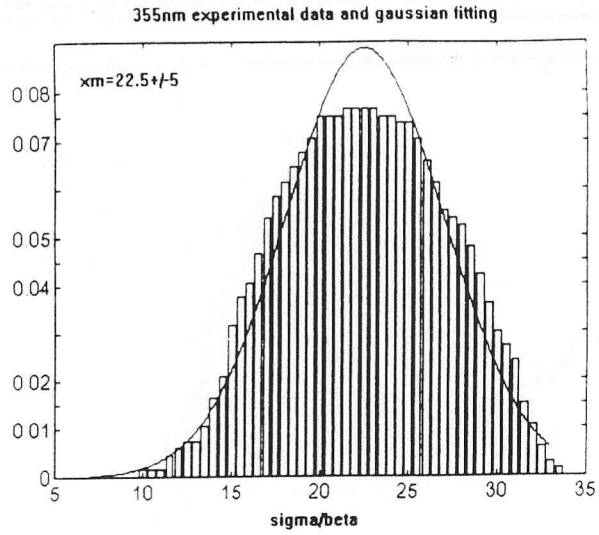
**Figure 9:** Long-lasting non depolarizing PSC observed on January 19, 1992 at Sodankyla

19/01/95 polarization and depolarization channels

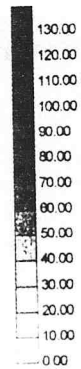
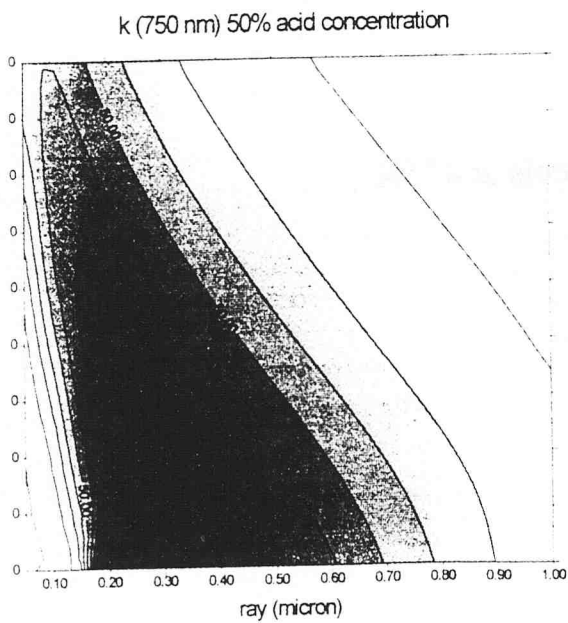
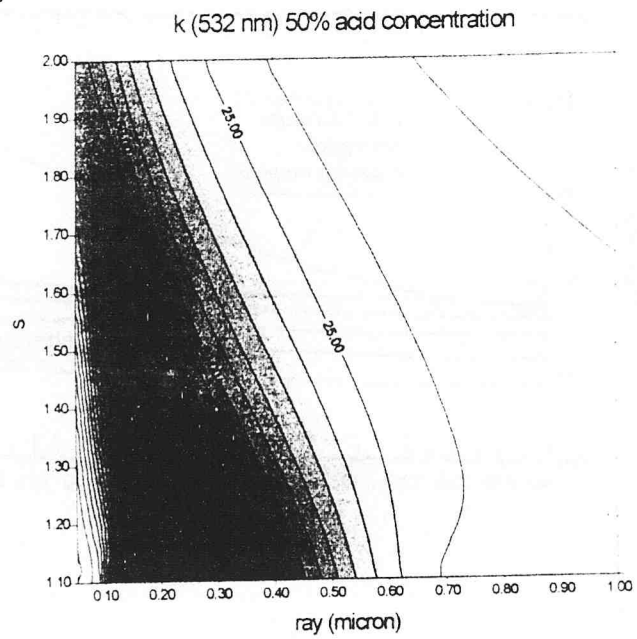
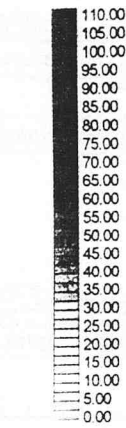
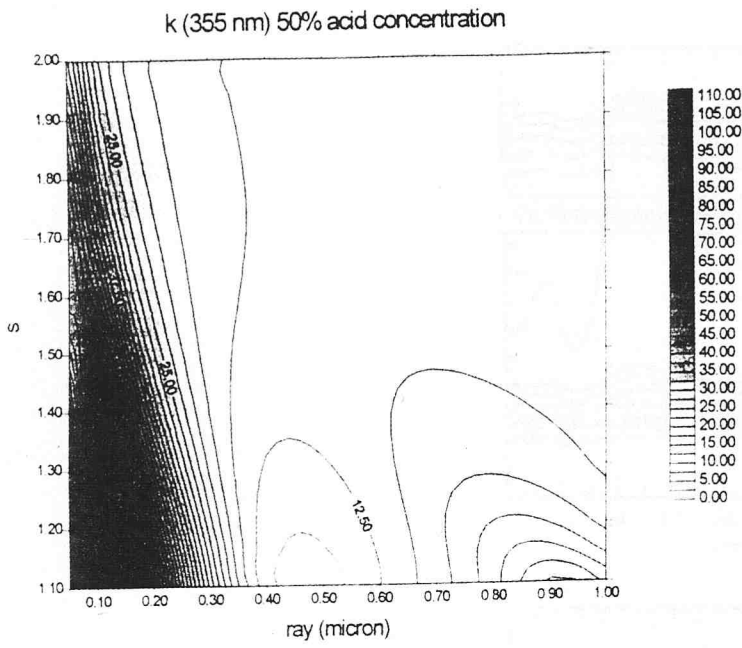


**Figure 10:** Polarization and depolarization channels for the PSC event of January 19,1992





**Figure 11:** Extinction to backscatter ratio at 355nm, 532nm and 750nm obtained from the inversion of LIDAR signatures for PSC event of 19.01.1992



**Figure 12:** Plots of Mie simulations of the extinction to backscatter ratio for 50% acid solution computed for three wavelengths (355nm, 532nm and 750nm).

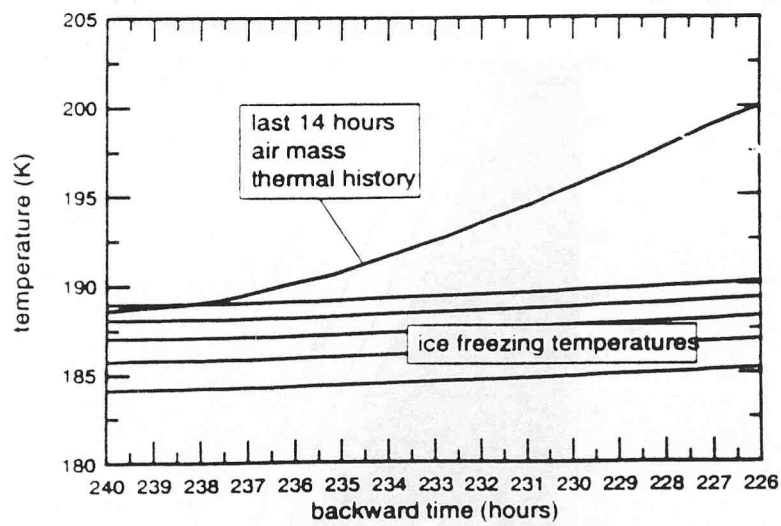
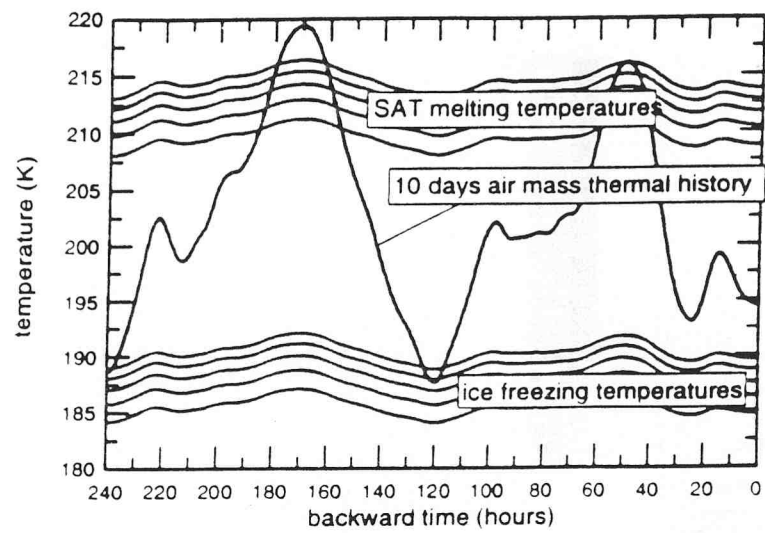


Figure 13: Temperature time history for the air parcels at 475K

## REFERENCES

K. S. Carslow, B.P. Luo, S.L. Clegg, T. Peter, P. Briblecomb, P.J. Crutzen, Stratospheric aerosol growth and  $\text{HNO}_3$  gas phase depletion from coupled  $\text{HNO}_3$  and water uptake by liquid particles, Geoph. Res. Letters, 21, 23, 2479-82, 1994

K. Drdla, A. Tabazedeh, R.P. Turco, M.Z. Jacobson, J.E. Dye, C. Twohy, D. Baumgardner, Analysis of the physical state of one Arctic polar stratospheric cloud based on observations, Geoph. Res. Letters, 21, No 23, 2475-2478, 1994

M. Del Guasta, M. Morandi, L. Stefanutti, B. Stein, J. P. Wolf:, Derivation of Mt. Pinatubo Stratospheric aerosol mean size distribution by means of a multiwavelength lidar, Applied Optics, 33 No24, 1994

Flesia, C., A. Mugnai, Y. Emery, S. Godin, L. de Schoulepnikoff, V. Mitev, Interpretation of lidar depolarization measurements of the Pinatubo stratospheric aerosol layer during EASOE, Geoph. Res. Letters, 21, No 13, 1443-1446, 1994

A.M. Middlebrook, B.S. Berland, S.M. George, M.A. Tolbert, O.B. Toon, Real refractive indices of infrared characterized nitric-acid films: Implications for optical measurements of polar stratospheric clouds, Jour. Geophys. Res., 99, D12, 25655-25666, 1994

Mishchenko M.I., L.D. Travis, Light scattering by polydispersions of randomly oriented spheroids with sizes comparable to wavelengths of observation, Applied Optics, vol.33, NO 30, 20 Oct. 1994.

M.J. Molina, R. Zhang, P.J. Wooldridge, J.R. McMohen, J.E. Kim, H.Y. Chang, K.D. Beyer, Physical Chemistry of the  $\text{H}_2\text{SO}_4/\text{HNO}_3/\text{H}_2\text{O}$  system, implication for Polar Stratospheric Clouds, Science, 261, 1418-1423, 1993

M. Pantani, Studio di Nubi Polari Stratosferiche mediante Lidar Multispettrale, Tesi di Laurea in Fisica, Università degli Studi di Firenze, Aprile 1996

Sacco, V.M., F. Castagnoli, M. Morandi, and L. Stefanutti, Elastic backscattering lidar system for stratospheric measurements in Antarctica, Opt. and Quantum Electr., 21, 215, 1989

B. Stein, M. Del Guasta, J. Kolenda, M. Morandi, P. Rairoux, L. Stefanutti, J.P. Wolf:, Stratospheric Aerosol Size Distribution from Multispectral Lidar Measurements at Sodankylä

Stefanutti, L., F. Castagnoli, M. Del Guasta, M. Morandi, V. M. Sacco, L. Zuccagnoli, S. Godin, G. Megie, J. Porteneuve, The Antarctic Ozone LIDAR system, Appl. Phys. B, B55,(1),3-12, 1992

Stefanutti, L, F. Castagnoli, M. Del Guasta, M. Morandi, V.M. Sacco, V. Venturi, L. Zuccagnoli, J. Kolenda, H. Kneipp, P. Rairoux, B. Stein, D. Weidauer, J.P. Wolf, A four wavelength depolarization backscattering lidar for polar stratospheric cloud monitoring, Appl. Phys. B, B55,(1),13-17, 1992  
during EASOE, Geophys. Res. Lett., 21, 13