

# DANISH METEOROLOGICAL INSTITUTE

## SCIENTIFIC REPORT

96-11

**European Stratospheric Monitoring Stations in the Arctic:**

**An European contribution to the**

**Network for the Detection of Stratospheric Change**

CEC Environment Programme Contract EV5V-CT93-0333

DMI contribution to the final report

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# European Stratospheric Monitoring Stations in the Arctic ESMOS/Arctic

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CEC Environment Programme Contract EV5V-CT93-0333

Contribution to the final report from the  
Danish Meteorological Institute

This CEC-project has been accomplished in collaboration with  
Alfred Wegener Institute, Potsdam (coordinator)  
University of Rome  
University of Bremen  
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## **Introduction.**

Since the discovery of the Antarctic ozone hole more than 10 years ago a major scientific effort has been imposed to clarify the reasons and the magnitude of the depletion of the stratospheric ozone layer which protects the Earth's surface against harmful ultraviolet radiation from the Sun and plays a significant role in the radiative balance of the atmosphere.

It is now ascertained that the total ozone over the south polar continent each spring is depleted by more than 50 % and the ozone is completely destroyed in the altitude range about 15-20 km. Ozone depletions have also been observed in the Arctic, although ozone holes have not yet developed. Consistent with other measurements the DMI observed ozone destruction over Greenland in the lower stratosphere during the first three months of 1993 at rates exceeding 1 % per day which is comparable to Antarctic conditions. Satellite and groundbased observations reveal that the ozone abundance decreases about 4-7% per decade at midlatitudes and even more in polar regions in both hemispheres, demonstrating the severeness of this global environmental problem.

The increased chlorine and bromine loading of the stratosphere from man made chlorofluorocarbons (CFCs) and halons has been verified to cause the ozone depletions in the polar regions. The reasons for the midlatitude ozone depletion still remain unsolved and cannot be explained by state of the art atmospheric chemistry models. Effects from major volcanic eruptions in combination with increasing chlorine loadings can be discerned, and also evidences of couplings to atmospheric processes in the polar regions appear. The growth rates of several ozone-depleting substances have slowed as a consequence of the restrictions imposed by the Montreal Protocol and its latest Copenhagen Amendment, but peak global ozone losses are still expected to occur during the next several years, and a normalized state of the ozone layer is not expected until the middle of the next century.

Southern hemisphere measurements have shown high levels of ultraviolet surface radiation under clear-sky conditions during periods of low ozone, adding confidence to the ability to link ozone changes to UV-B changes over relatively long time scales. The loss of ozone in the lower stratosphere may cause a decrease in stratospheric temperatures. This itself may induce further chemical ozone destruction, and impose a global-mean negative radiative forcing, leading to an off-set of the positive radiative forcing of well-mixed greenhouse-gases. Increased concentrations of ozone in the troposphere and near the tropopause will enhance the greenhouse-warming. Hence, observing the ozone changes will be an important ingredient in the understanding and ability to predict climate changes and future levels of surface UV-B radiation.

### **Network for the Detection of Stratospheric Change. European Stratospheric Monitoring Stations in the Arctic.**

Realizing the importance of observing and understanding the physical and chemical state of the stratosphere the Network for the Detection of Stratospheric Change (NDSC), comprising a limited set of high-quality remote-sounding research stations, was initiated in the beginning of the 1990's. This effort is supported by national and international institutes and organizations including National Aeronautics and Space Administration (NASA), the United Nations Environment Programme (UNEP), the World Meteorological Organisation (WMO), the EU-commission, and others. A handful of stations in the tropics, at midlatitudes, and in the polar regions in both hemispheres are now established with a number of selected instruments of verified high quality, including UV-visible and infrared spectrometers for measurements of column abundance of ozone and a large number of trace gases, ozone and aerosol lidars, microwave sounders, and facilities to perform balloonborne ozone soundings.

The objectives of the NDSC-stations are to make the earliest possible identification from observations of changes in the ozone layer and to discern the cause of the changes, to provide independent calibrations of satellite measurements, and to obtain observational data to be used to test and improve atmospheric chemical, dynamical, and climate models, thereby enhancing the confidence in the predictive and assessment capabilities of these models.

The DMI stratospheric observatory at Thule Air Base, Greenland (76.5 N, 68.8 W), has been appointed to host one of the arctic NDSC stations. The DMI possess a number of buildings on the South Mountain and at the main base at Thule, fully equipped with communication (telephone, fax, internet) and computational facilities. Presently, the DMI has contributed to the instrumentation of the site by a UV-visible spectrometer for ozone and other trace gas measurements. The DMI has also installed instrumentation for balloonborne measurements of ozone. In collaboration with the DMI the University of Rome operates an aerosol lidar, the National Centre for Atmospheric Research (NCAR), Boulder, Colorado is in the process of installing an infrared spectrometer, and the University of Stony Brook, New York, has performed microwave measurements of ozone and chlorine compounds. The DMI has collaborated for several years with the University of Wyoming on balloonborne stratospheric aerosol and cloud particle measurements from the NDSC station at Thule.

The Commission of the European Union supports the NDSC activities of observing the climatology of the ozone layer through the project European Stratospheric Monitoring Stations (ESMOS). Beside the DMI-Thule site this network also comprises an arctic station at

Spitsbergen and various midlatitude stations in Europe. The DMI has participated, by contracts to the EU-commission, from the start of the ESMOS-project and through its predecessors ("Experimentation related to polar stratospheric clouds", contract STEP-CT90-0078, and "Investigations of ozone, aerosols, and clouds in the Arctic stratosphere", contract EV5V-CT92-0074), and the ESMOS-project is continued under the 4th framework EU-Environment program with DMI participation ("ESMOS/Arctic II", contract EV4V-CT95-0136).

Part of the work in the project was conducted in connection with the Second European Stratospheric Arctic and Midlatitude Experiment (SESAME) during years 1994 and 1995. It was a major objective of the ESMOS/Arctic project to support this all-European effort with continuous, long-term measurements during these two years and increased observational activities during SESAME phases I and III.

### **Objectives.**

The primary role of the DMI-work within the ESMOS/Arctic project has been to launch a number of backscatter sondes for investigations of volcanic aerosol and polar stratospheric clouds (PSC) from Thule. The cloud measurements has been supposed to be analysed in connection with theoretical microphysical models of the PSC formation processes, based on temperature histories from air parcel trajectories.

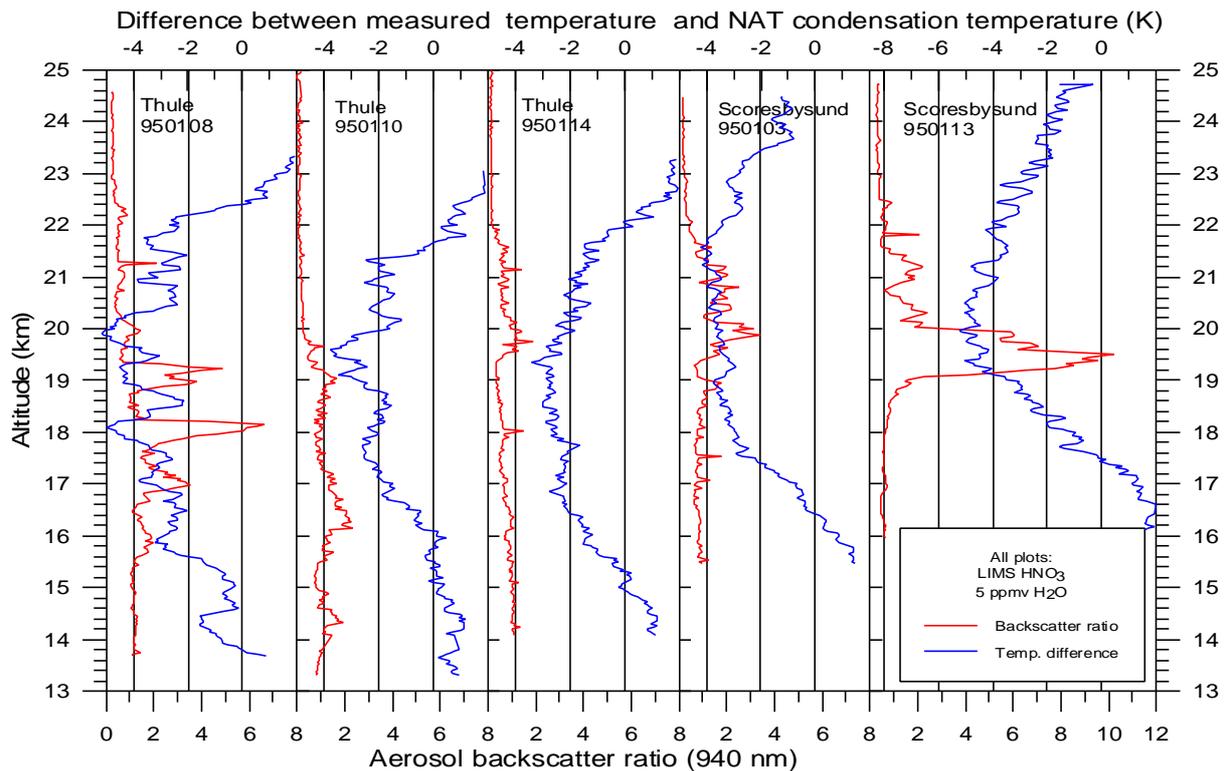
Meteorological analyses of potential vorticity and backward trajectory calculations were supposed to be performed on a daily basis with the intention to calculate atmospheric transport and estimate the effect of a possible Arctic induced ozone depletion at lower latitudes. Ozone soundings and groundbased spectroscopic measurements of trace gases were supposed to be supplemented from related projects.

### **Main results obtained.**

#### *Backscatter soundings.*

The observations have been obtained, using the University of Wyoming backscatter sonde which measures in situ the backscattered light from the cloud particles in two wavelengths around 940 and 480 nm with a vertical resolution of 30 meters during balloon ascent and descent. A color index, defined as the ratio between the backscatter in the two wavelengths, provides information of the particle sizes. The ozone concentration, ambient pressure and temperature are measured concurrently with the aerosol backscatter.

Three balloonborne backscatter soundings have been performed from Thule, supplemented by two soundings from Scoresbysund (70.5°N, 22.0°W) Greenland, during January 1995 as part of the SESAME campaign, on days when stratospheric temperatures were low enough for the formation of polar stratospheric clouds (PSCs). The observed vertical profiles of aerosol backscatter at 940 nm, measured on January 8, 10, and 14 at Thule and January 3 and 13 at Scoresbysund, are shown in Figure 1 below as thick curves.

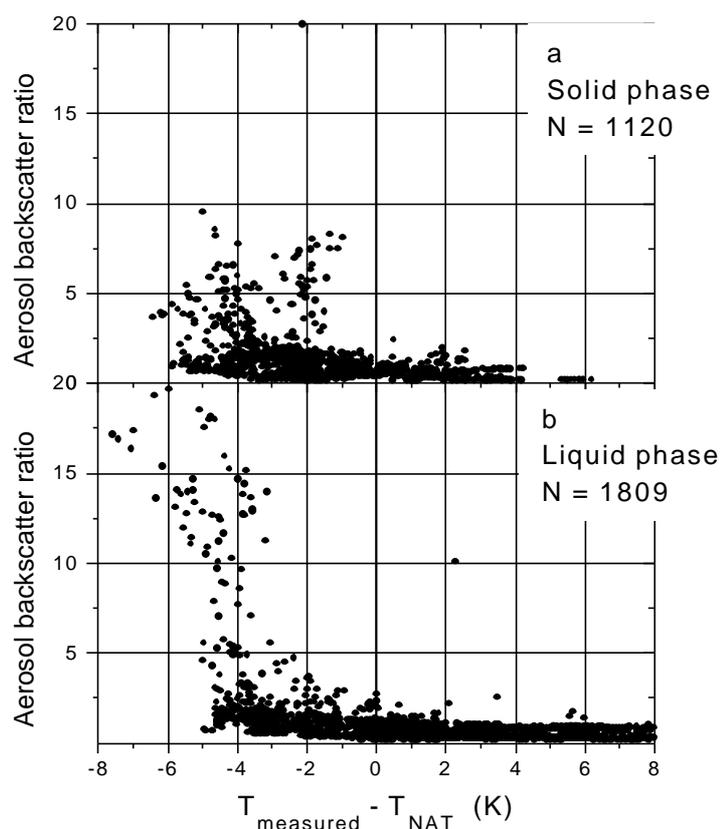


**Figure 1.** Vertical profiles of red-channel aerosol backscatter ratio (thick lines) and difference between air temperature and nitric acid trihydrate (NAT) condensation temperature (thin) in 5 soundings from Greenland, January 1995, during the SESAME campaign.

In the first sounding from Thule strong signals of PSCs were observed in layers around 17, 18, and 19 km altitude with weaker occurrence around 16 and 20 km. On the two subsequent soundings from Thule weak signs of PSCs were observed in broad layers between 16 and 19.8 km and between 19.5 and 22 km. In both soundings from Scoresbysund PSCs were observed in a broad altitude range between 19 and 22 km with a layer in the last sounding around 19.5 km with very high backscatter ratios.

Also shown in Figure 1 (thin lines) are the differences between the measured air temperature and the estimated nitric acid trihydrate (NAT) condensation temperature. In order to calculate the latter a northern hemisphere  $\text{HNO}_3$  LIMS profile has been assumed together with 5 ppmv water vapor mixing ratio at all altitudes. In the first sounding from Thule PSCs clearly form in the layers with the coldest temperatures. However, in all soundings the temperatures fall well below the estimated NAT condensation temperatures in broad altitude ranges between 16 and 22 km without necessarily leading to pronounced PSC formation.

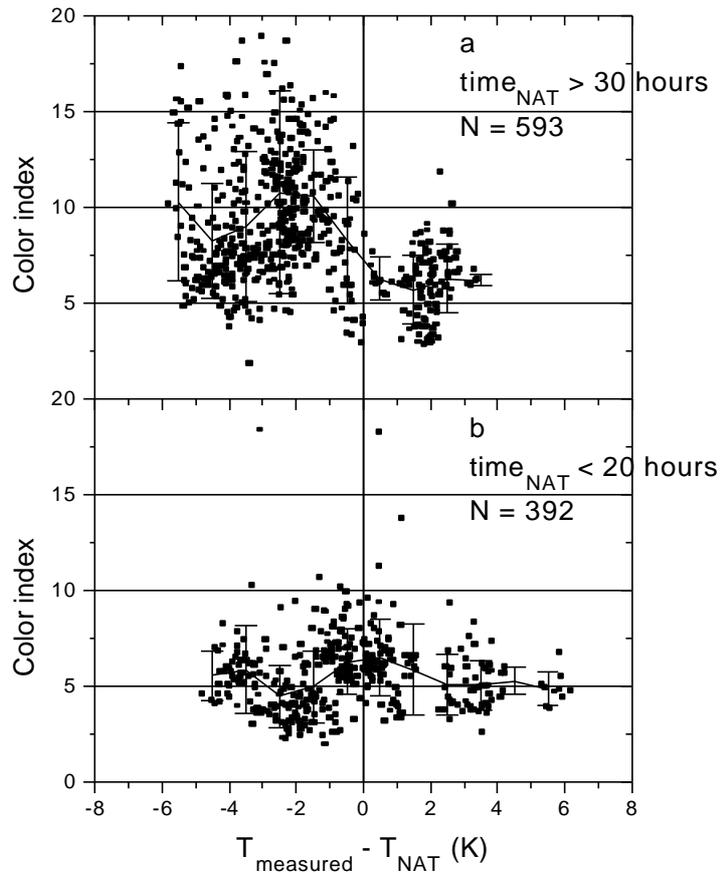
According to a suggested scenario for PSC formation [Tabazadeh et al., GRL 22, 1725, 1995] liquid supercooled ternary solution particles may freeze upon an increase in temperature after previously being close to the ice frost point. The obtained PSC backscatter measurements, supplemented by similar measurements from other Arctic sites during previous years, have been used together with backward air parcel trajectories to access the suggested freezing behaviour and PSC formation theory [Larsen et al., 1996].



**Figure 2.** Aerosol backscatter ratios of PSCs, sorted into liquid and solid phase, and plotted against the difference between the measured air temperature and the NAT condensation temperature.

Liquid particles, identified by the temperature histories according to this scenario, show a relatively compact relationship (cf. Figure 2b) between measured temperature and backscatter ratio, indicating a substantial growth at 3-4 K below the NAT-temperature as expected by ternary solution particles (type 1b PSC). The scatter in the similar diagram among solid particles is larger (Figure 2a).

The color index of the solid particles has been used to estimate the particle sizes. It appears that newly formed solid particles are of small size. Aged solid particles, on the other hand, appear to retain large sizes up to the NAT-temperatures during evaporation, whereas the particles grow by condensation at temperatures slightly below  $T_{\text{NAT}}$ , indicating the composition to be NAT, cf. Figure 3. This result may indicate that solid nitric acid particles form in a metastable phase (e.g. nitric acid amorphous solid solution or nitric acid dehydrate) with a relatively high vapor pressure. These metastable phases may then after a while (app. one day) turn into stable NAT crystals with a lower vapor pressure. This pathway may provide a mechanism for the formation of large solid type 1a PSC particles since large amounts of gas phase  $\text{HNO}_3$  may condense on the few first formed NAT particles simultaneously with the evaporation of the remaining metastable and liquid particles.



**Figure 3.** Color index of solid particles, plotted against the difference between the measured air temperature and the NAT condensation temperature. Solid particles which have excised at temperatures below  $T_{\text{NAT}}$  for more the 30 hours retain relative large color indices (sizes) up  $T_{\text{NAT}}$ , whereas younger solid particles appear to be of smaller sizes.

#### *Meteorological analysis.*

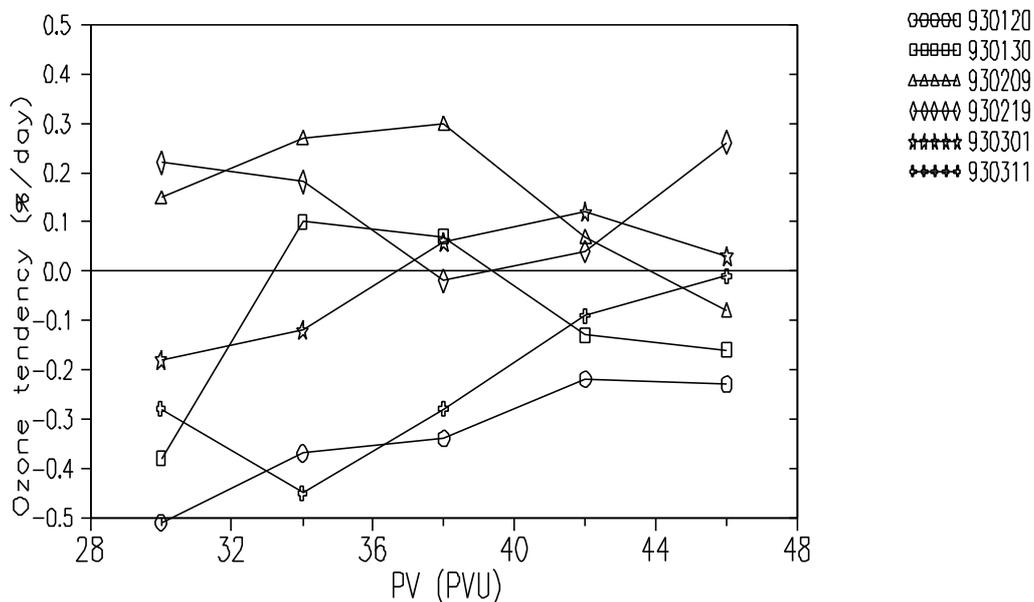
Potential vorticity (PV) and temperature have been calculated in a  $2.5^\circ \times 2.5^\circ$  latitude-longitude grid north of 30N on the 350, 380, 400, 435, 475, 550 and 675K isentropic levels. A 1, 2, 3, 4, 5 and 8 day forecast for 350, 475, 550 and 675K has also been available in real time. These data have been calculated from 1 November to 30 April during the winters 1993/94 and 1994/95 and for campaign activities outside this period. Files with higher-resolution PV data interpolated to each measurement site are also available.

10 day backward isentropic trajectories have been calculated at the 350, 380, 400, 435, 475, 550 and 675K isentropic levels with endpoints on all measurement sites and on an equal area grid with 118 gridpoints north of 30N. These trajectories are calculated for the same period as the other isentropic data mentioned above. In the winter 1993/94 forecasted backward trajectories on the 350, 380, 400, 435, 475 and 550K isentropes ending on ozone sonde launching sites were also calculated. These trajectories were based on the analyses and both 2 and 4 days of forecast.

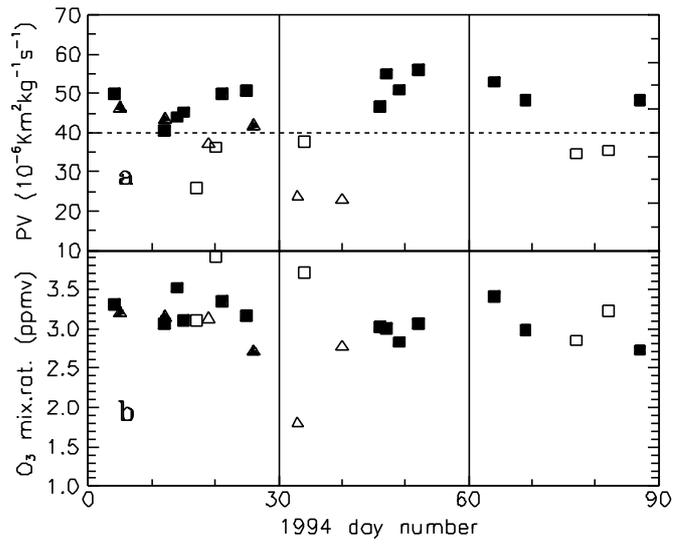
*Ozone soundings.*

For the winter 1992/93 the DMI has been able to detect chemical ozone depletion inside the polar vortex with ozone and backscatter sondes from Greenland (Larsen et al., 1994). Transport into the vortex of air from outside the vortex with relatively low ozone mixing ratios could not be completely ruled out as a cause for the depletion though. Therefore, we have investigated the transport across the edge of the vortex using domain-filling trajectory calculations. Ten day backward 475K isentropic trajectories were calculated in an equal area grid with 10471 points north of 30N. The ozone mixing ratio at the starting points was obtained from ozone soundings using the PV- $\theta$  ( $\theta$  = potential temperature) mapping techniques described in Knudsen et al. (1992). The mixing ratio at the start of the trajectory is then subtracted from the mixing ratio at the end. For both the start and end point of the trajectory the ozone mixing ratios from the start date is used.

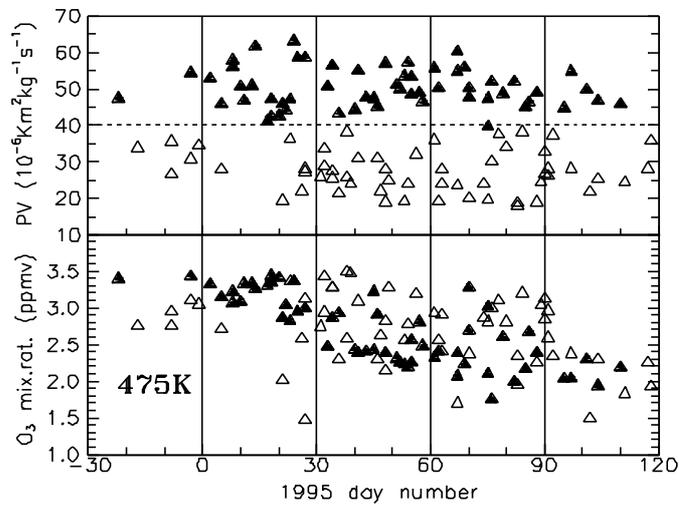
In Figure 4 the average net ozone tendencies is given for 6 ten day periods as a function of PV. It is evident, that the tendencies vary a lot. This could partly be due to intermittent warmings, during which most transport across the edge of the vortex occurs (Plumb et al., J. Geophys. Res., 99, 1089-1105, 1994). It could also be due to shortcomings of the PV- $\theta$  mapping or changes in PV (which on average is considered constant over the 10 day period for this analysis). Nevertheless, it is evident, that the average ozone tendency due to isentropic transport in January to March is much smaller than the ozone depletion found in Larsen et al. (1994), which is listed in Table 1. Therefore, the 475 K ozone depletion found in January to March 1993 cannot be explained merely by intrusions of low ozone air from outside the vortex.



**Figure 4.** 475K Ozone tendencies due to isentropic transport calculated from 10 day backward domain-filling trajectories. In the legend the end date of the trajectories is given.



**Figure 5.** 475 K ozone mixing ratio and PV over Thule (squares) and Scoresbysund (triangles) during the winter 1993/94. Filled symbols indicate PV larger than 40 PVU.

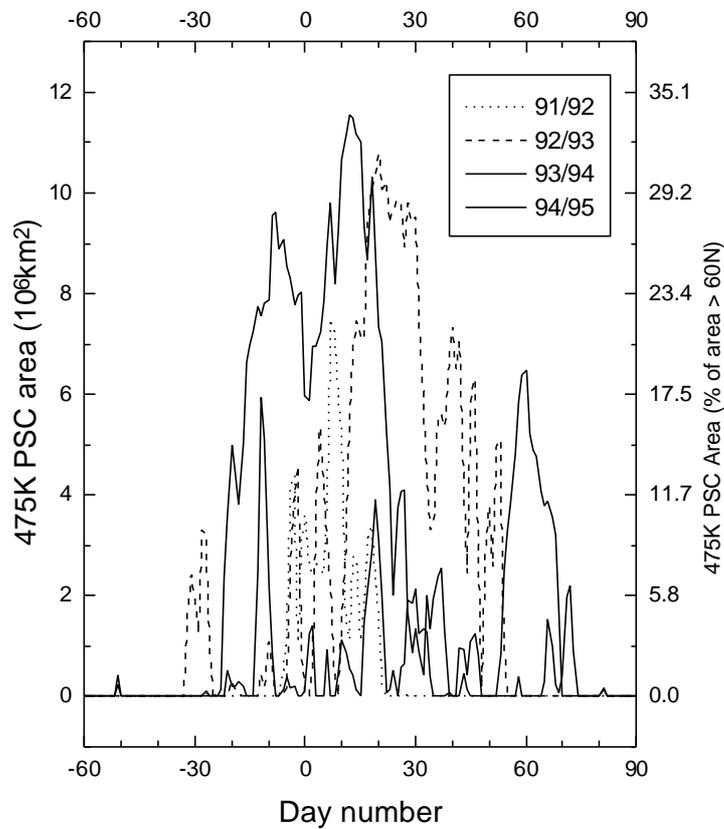


**Figure 6.** 475 K ozone mixing ratio and PV over Greenland ozone sounding stations during the winter 1994/95. Filled symbols indicate PV larger than 40 PVU.

The Figures 5 and 6 show the ozone mixing ratios measured by Greenland ozone soundings at 475 K in the winters 1993/94 and 1994/95. The 475 K PV is shown in the plots as well and soundings with a PV of more than 40 PVU ( $10^{-6}\text{kg}^{-1}\text{s}^{-1}$ ) (well inside the vortex) are marked with filled symbols. Regression analysis of these data showed an ozone depletion at 475 K from January to March 1994 of  $-0.14\pm 0.14$  %/day for soundings inside the polar vortex. Including Ny Ålesund soundings in the regression leads to a trend of  $-0.26\pm 0.16$  %/day. Here only 95 % confidence limits are given. In the winter 1994/95 the downward trend was much larger. In Table 1 the trends are given for the set of all ESMOS/Arctic soundings (soundings from Thule, Scoresbysund, Søndre Strømfjord, Ny Ålesund and Sodankyla). The trends are compared to trends in the winters 1991/92 and 1992/93 derived from Greenland ozone soundings alone (Larsen et al., 1994). These trends are not corrected for diabatic cooling, that generally increases the trends. In 1992/93 the correction for cooling, inferred from the descent of the Pinatubo aerosol layer measured by backscatter sondes, increased the 475, 435, 400 and 380 K absolute trends more than a factor 1.8. Table 1 gives evidence for a substantial ozone depletion in 1994/95, that is comparable to the depletion seen in 1992/93.

Table 1. Vortex ozone depletion rates. 95% confidence limits are given. Number of measurements used in the regression analyses is given in parenthesis.

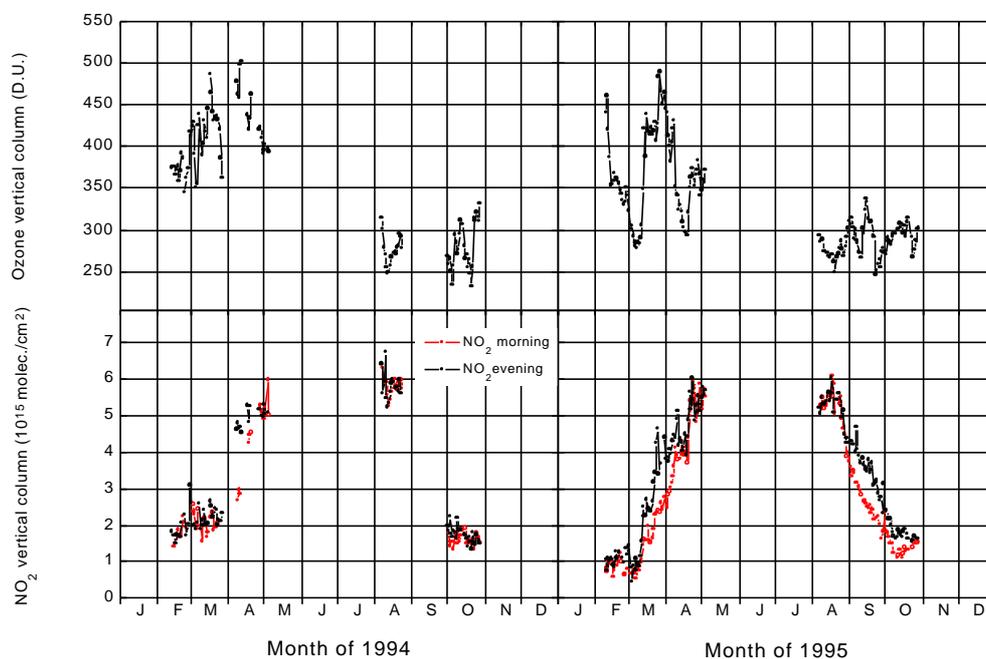
| level | vortex<br>PV-limit | Ozone depletion rates January-March (%/day) |              |              |
|-------|--------------------|---|--------------|--------------|
|       |                    | 1994/95                                     | 1992/93      | 1991/92      |
| 550K  | 80 PVU             | +0.2±0.1(75)                                | -0.1±0.2(12) | +0.2±0.1(31) |
| 475K  | 40 PVU             | -0.5±0.1(88)                                | -0.6±0.2(14) | +0.1±0.1(44) |
| 435K  | 26 PVU             | -0.9±0.1(97)                                | -0.6±0.3(15) |              |
| 400K  | 17 PVU             | -0.9±0.1(91)                                | -0.7±0.2(16) | -0.4±0.3(20) |
| 380K  | 13 PVU             | -0.5±0.2(85)                                | -0.6±0.3(13) | -0.1±0.6(15) |
| 350K  | 8.7 PVU            | +0.5±0.2(53)                                | -0.2±0.3(10) | +0.7±0.7(13) |



**Figure 7.** Temporal development of the 475 K PSC area in the northern hemisphere in the winters 91/92, 92/93, 93/94 and 94/95 predicted from ECMWF analysed temperatures.

The variation of the ozone depletion from winter to winter can be explained by the occurrence of PSCs. Figure 7 shows the 475 K PSC area for all the winters predicted from ECMWF analysed temperatures using a LIMS HNO<sub>3</sub> profile and 5 ppmv H<sub>2</sub>O. It is evident, that there was much more PSC activation predicted in the winters 1992/93 and 1994/95, than in the other two winters.

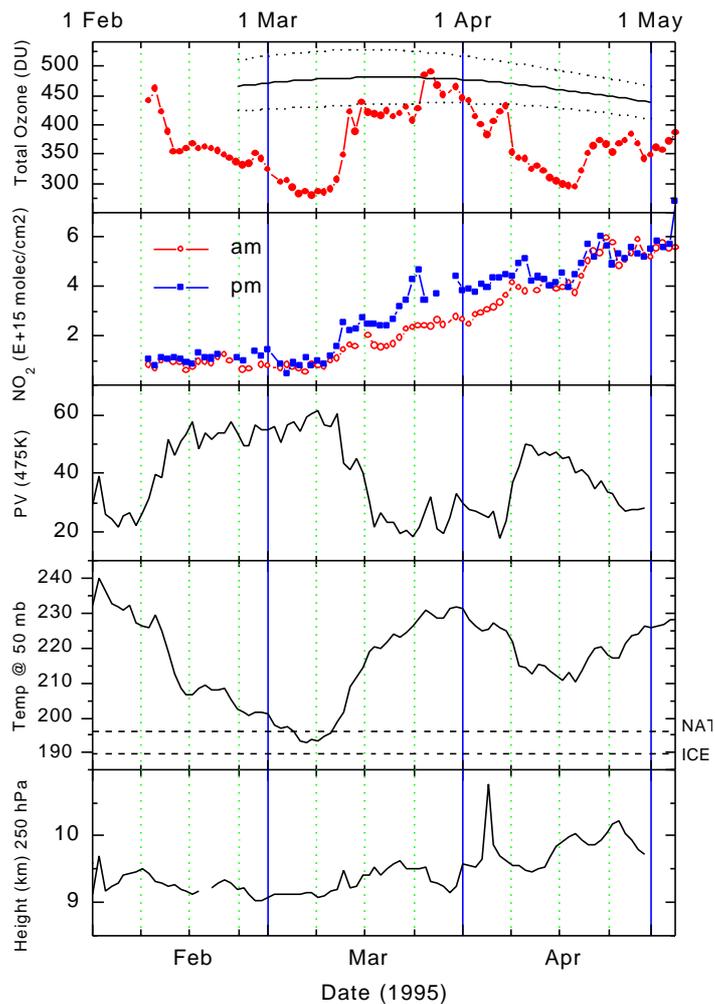
## Groundbased spectroscopic measurements.



**Figure 8.** Thule, NO<sub>2</sub> and Ozone vertical columns 1994 and 1995.

A SAOZ UV-vis spectrometer has been in operation in Thule, Greenland (76.5 N, 68.8 W) since September 1990. During this period total columns of ozone and NO<sub>2</sub> have been measured by observing the sunlight scattered from the zenith sky at sunrise and sunset. The measurements with the standard SAOZ method are only reliable in the solar zenith interval 91°-87°, this means that reliable measurements can only be made in the periods February -April and August - October. Figure 8 shows the ozone and NO<sub>2</sub> columns measured in 1994 and 1995. The missing observations in spring 1994 are due to a program malfunction and the missing observations in September 1994 are due to participation in the intercomparison of UV-vis spectrometers in Camborne. Results from the intercomparison will be published by Vaughan et al. (1996).

Figure 9 shows the vertical columns of ozone and NO<sub>2</sub> measured by in spring 1995 in more detail, together with ECMWF potential vorticity at 475 K, indicative of the presence of the vortex over the station, and the temperature of the 50 mb level. Finally the height of the 250 hPa pressure level is shown in order to distinguish variations of the vertical columns due to variations in pressure. In the period mid-February to mid-March the vortex was over Thule and a continual decrease in ozone is seen. Temperatures low enough for polar stratospheric clouds to form are only seen in the vortex in a shorter period in March, where also NO<sub>2</sub> values are very low indicating that processing of nitrogen oxides on PSC's may be taking place.



**Figure 9.** Vertical columns of ozone and NO<sub>2</sub> spring 1995, together with ECMWF potential vorticity at 475 K, the temperature of the 50 mb level and the height of the 250 hPa pressure level.

### Acknowledgements.

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