N95-11028

STRATOSPHERIC MINOR SPECIES VERTICAL DISTRIBUTIONS DURING POLAR WINTER BY BALLOON BORNE UV-VIS SPECTROMETRY

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ABSTRACT

A light, relatively cheap and easy to operate balloon borne uv-visible spectrometer was designed for investigating ozone photochemistry in the Arctic winter. The instrument was flown 11 times during the European Arctic Stratospheric Ozone Experiment (EASOE) in winter 1991-92 in Northern Scandinavia. The first simultaneous measurements of vertical distributions of aerosols, PSCs, O_3, NO_2 and OClO inside the vortex during flight # 6 on 16 January, in cold conditions are reported, which show that nitrogen oxides were almost absent (lower than 100 ppt) in the stratosphere below 22 km, while a layer of relatively large OClO concentration (15 ppt) was present at the altitude of the minimum temperature.

1. INTRODUCTION

PSCs, nitrogen and chlorine oxides and, in addition, volcanic aerosols injected into the stratosphere by the eruption of Mount Pinatubo in 1991, are the key parameters controlling possible chemical ozone destruction in the Arctic Polar Stratosphere. Their measurement was therefore a primary goal of the European Arctic Stratospheric Ozone Experiment held in Northern Scandinavia in winter 1991-92.

UV-Visible spectroscopy by solar occultation from balloons, already in use for total column observations from the ground in polar regions, offers an opportunity for measuring simultaneously the vertical distributions of several of the above constituents and parameters, namely O_3, NO_2, OClO and aerosol and PSC optical thicknesses, at the latitude of the polar circle in winter and if possible, inside the stratospheric vortex which then becomes a primary target.

However, because of the large displacements of the vortex on one hand and the rather difficult balloon launch conditions prevailing in the Arctic in winter on the other, measurements inside the vortex are rather difficult.

To overcome these adverse conditions and to make frequent flights possible at short notice, a light, compact and highly automated uv-visible spectrometer, was designed and tested successfully in September 1991 in France. Three similar units were then built and used during EASOE at Kiruna in northern Scandinavia for conducting 11 flights from November 1991 until the end of March 1992. The results provide a first insight of the evolution of the vertical distribution of several constituents during the polar Arctic winter of 1991-92.

2. INSTRUMENT CONCEPT

The instrument itself is a uv-visible spectrometer identical to the SAOZ instrument developed at laboratory for ground-based monitoring of the stratosphere (Pommereau and Goutail, 1988). It is a diode array spectrometer working in the 300-600 nm spectral range with a 0.6 nm resolution. The two important features added for balloon flights are: a) hemispherical diffuser and a baffle defining a field of view of 360° in azimuth and + 10°, -5° in elevation, allowing direct sun observations without any tracking or orientation system, and b) an on-board CPU which controls the exposure duration, the duty cycles (alternate observations in the visible, the uv and dark current) and compresses the data transmitted by a simple channel telemetry. There is no remote control. The gondola weights 19 kg.

Besides this, a light flight train, a reinforced 10 000 m^3 balloon (for low stratospheric temperatures and fast wind speed during launch) and simplified launch procedures were designed and tested by the CNES Balloon Division, permitting flights in windy and cold conditions during which orographic PSCs were observed to occur in the past (Pommereau and Schmidt 1991). The gondola is designed to be dragged onto the launch pad, if necessary, and to be protected against hard landings, making frequent flights with the same instrument possible. The period for preparing and launching does not exceed 1h and thus allows flights on alert.

3. MEASURED CONSTITUENTS AND PARAMETERS

The 300-600 nm spectral range allows the simultaneous observations of absorption signatures of various constituents and attenuations by molecular and particle scattering and then, providing the altitude and location of the balloon are known, the retrieval of their altitude distributions. Identified constituents are O_3 in the uv and visible, NO_2 between 360 and 470 nm, OClO between 350 and 390 nm, $(O_2)_2$ at 360, 477 and 576 nm and H_2O in the troposphere at 505 and 590 nm. Mie and Rayleigh attenuations are derived from total attenuation measurements at several wavelengths. Size and number densities of large PSC particles greater than 0.5 μm, can be derived from Mie differential spectral signatures often present in the winter Arctic stratosphere.

4. FLIGHTS DURING EASOE

After a test flight held at Aire sur l'Adour in southern France in September 1991, 11 flights were conducted from ESRANGE at Kiruna in northern Sweden from November 1991 until March 1992, during the EASOE campaign. Figure 1 shows the stratospheric conditions during which these flights took place as measured by a ground-based SAOZ spectrometer (O_3 and NO_2) at Sodankyla in Finland (i.e.

below most of the balloons trajectories), and by meteorological data from the ECMWF (temperatures at 30 and 50 hPa, potential vorticity at 400 K) archived by the data center of NILU at Oslo. The above data, available in real time at ESRANGE, allowed a large diversity of flights: inside and outside the vortex (as shown by PV and total ozone) and at sunrise or sunset, in cold or warm conditions, from the darkest days in winter until equinox in March. Among the 11 flights, number 6 on 16 January, was the one performed in the coldest stratospheric conditions, well inside the polar vortex and in presence of dense orographic PSCs sighted from the launch pad. Its results are described below.

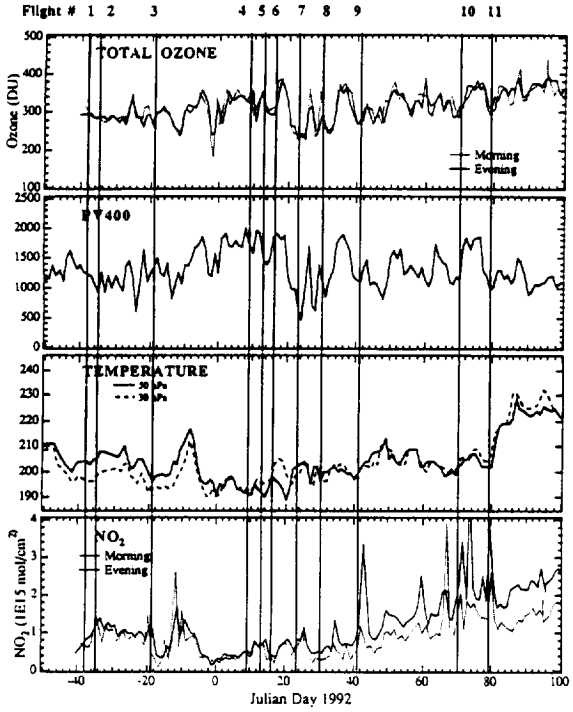

Fig. 1. Total ozone and NO_2 as observed during EASOE by a ground-based SAOZ at Sodankyla in Finland, together with potential vorticity at 400 K and temperatures at 50 and 30 hPa above the station. The above information available in real time, allowed us to explore a large variety of stratospheric conditions as indicated by the vertical bars at the dates of the flights. Among these, the 6 th on 16 January which is presented in greater detail here, was the one performed the deepest inside the vortex.

5. IN-FLIGHT OBSERVATIONS

As well as many observations from orbit since summer 1991, solar occultation measurements from balloon at twilight have been made difficult because of the large tangent optical thickness of the Pinatubo aerosol cloud. Measurements at twilight are impossible for tangent heights below about 19 km and therefore observations below this altitude can be only achieved at lower sun zenith angle (SZA), that is during the ascent of the balloon. This is illustrated in figure 2 where the altitude of the balloon during flight # 6 and the tangent height and SZA are displayed in panel a), observed fluxes at 350, 450 and 550 nm in panel b) and a color index defined as the ratio between the 550 and 350 nm fluxes corrected for absorption by constituents in panel c).

The balloon is launched so that it reaches float at approximately 90° SZA. This gives enough sensitivity to perform measurements during ascent and gives the maximum duration for observations during sunset. The first 5 spectra were recorded below a cirrus cloud layer. Fluxes are constant as well as the Color Index (CI) which is shifted toward the blue. These are cloudy zenith sky measurements which cannot be used in the following. As soon as the balloon crossed the cirrus layer at 8 km, all fluxes increase rapidly and the CI

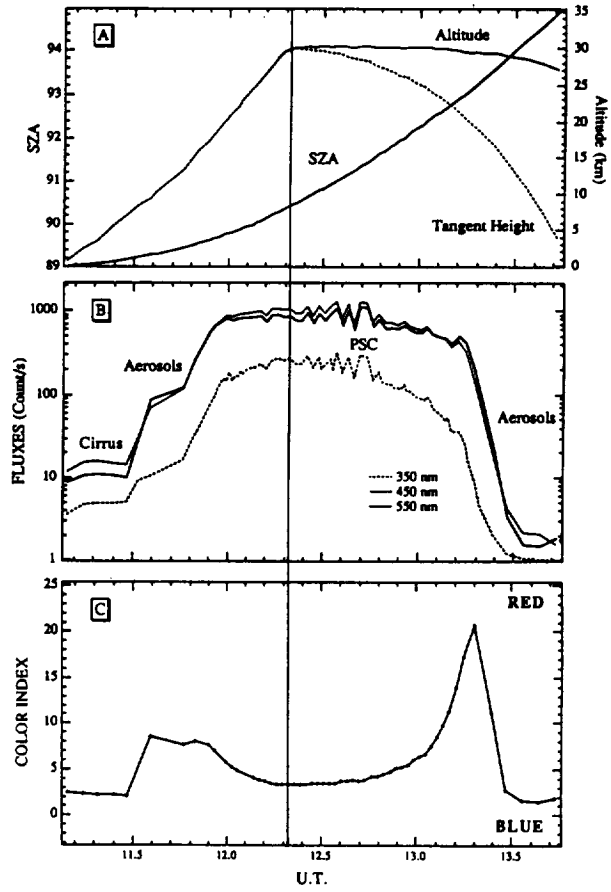

Fig. 2. Data from flight # 6. Part 1. Panel a): Balloon altitude, tangent height and sun zenith angle; Panel b) Fluxes at 350, 450 and 550 nm in arbitrary units; Panel c) Color index defined as the ratio between the 550 and 350 nm corrected for absorptions, mostly by ozone in the Chappuis bands (see text).

shifts toward the red. Direct sun observations begin, but fluxes are still reduced by a factor 20 compared to what was observed at float. This is a result of the aerosol layer, which is not crossed until the balloon reaches 20 km. Above, and up to float at 30 km, visible fluxes are almost constant while the progressive reduction of molecular scattering makes the CI decrease slowly. At float from approximately 90° SZA until 93° during sunset, where the tangent height reaches 20 km,

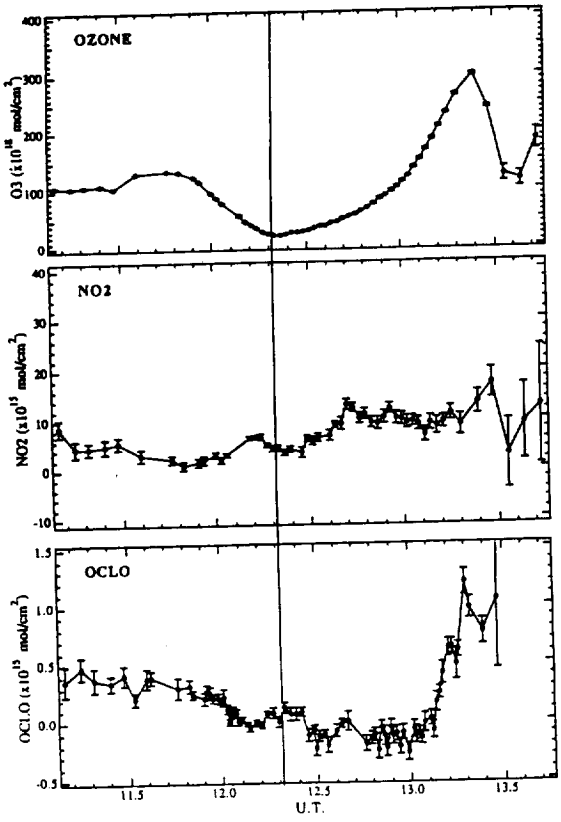

Fig. 3. Data from flight #6. Part 2. Ozone, NO_2 and OClO column densities along the line of sight (see text). Error bars correspond to one standard deviation error in the correlation between the signal and absorption cross-sections of the constituents. Vertical bars indicate the time of arrival of balloon at float.

incoming fluxes are decreasing progressively because of the increasing attenuation along the line of sight. Suddenly after 93° SZA, the sunlight reduces abruptly. The CI which was a maximum, shifts rapidly toward the blue. The sun is then not visible and measurements are no longer useful. The optical thickness of the aerosols cloud is too large to allow further observations.

During the last third of the ascent as well as during sunset, fluctuations of 20 to 30% appear in the fluxes. These, which are present only during cold temperature flights, are due to PSCs located around 25 km. Minima correspond to additional attenuation in the sun direction, while maxima are thought to be caused by enhanced forward scattering, known to be important for large particles, when the cloud is just below or above the sun disk. In the spectra, they correspond to large (10-15%) and narrow Mie scattering differential features identified as resulting from monodispersed particles of 3.3 μm radius. This is thought to be the explanation of the nacreous aspect of these clouds when sighted from the ground.

Ozone, NO_2 and OClO column densities along the line of sight, with error bars indicating one standard deviation error in the correlation between spectra and absorption cross sections, are displayed in figure 3 at the same time scale as in figure 2. The I_0 spectrum used as reference and to which actual spectra are compared, is the one obtained during flight #7 at 88°SZA on the morning and at an altitude of 30 km. As explained earlier the first 5 points and last 7 ones are not usable because the sun is not observed directly. The time when the balloon reaches float is indicated on each curve by a vertical bar. In such plot, the altitude of the maximum concentration of the constituent corresponds to the maximum slope of the curve. Ozone is the lowest at float and its maximum slopes are observed around 18 km during ascent and at the largest SZA during sunset. Indeed, the ozone maximum is located at 18 km. The behaviour of NO_2 is totally different. Its minimum column is observed at the smallest SZA during ascent and not at float. The maximum slopes are at or near float. The maximum concentration is then at or above float level. Column densities are more than 20 times smaller than that observed in the same conditions at mid

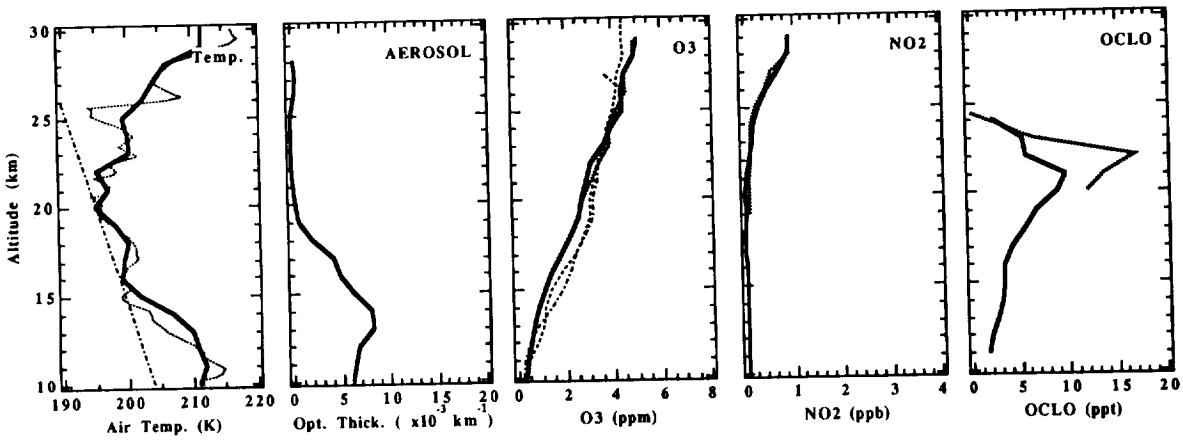

Fig. 4. Atmospheric temperature, aerosol optical thickness and ozone, NO_2 and OClO mixing ratios retrieved from the results shown in fig. 2 and 3. Measurements made during ascent are shown in full line while those obtained during sunset are indicated in dotted line. The additional dotted lines in the case of temperature and ozone show the results of soundings performed 2 hours after the main balloon launch at Kiruna and later in the evening at Sodankyla. The large variability of stratospheric temperatures measured 2 hours apart (i. e. around 26 km), is consistent with the presence of orographic PSC observed from the balloon and sighted also from the ground.

latitudes (Pommereau et al., 1987). From the location of the maximum slopes of OClO during both ascent and sunset, one can immediately see that there is a dense layer of this constituent around 18 km. The negative amounts of OClO indicate only that there was a little less OClO at high altitude during flight #6 than during flight #7. Note that the minimum is not observed when reaching float, but a little later. This could be just related to the presence of PSCs in the field of view and not necessarily to a diurnal variation. Jumps in size of error bars are due to the use of 2 sets of data in the same plot: alternate measurements made on "visible spectra" for which the exposure is adjusted at 500 nm and on "UV" spectra adjusted at 390 nm. The exposure and therefore the increase in counts number in the UV case, makes the noise to be reduced by a factor of 2 approximately.

Assuming concentrations remain constant during twilight, vertical distributions of the above constituents can be retrieved. They are displayed in mixing ratio units in figure 4 together with aerosol optical thickness and temperature recorded during the ascent of the balloon and 2 hours later by an ozone sonde when the main balloon was already at float. The large variability of stratospheric temperatures measured 2 hours apart (i. e. around 26 km), is consistent with the presence of orographic PSC observed from the balloon and sighted from the ground. It is clear from the plots, that NO_2 is considerably reduced at all levels below 24 km, that is where aerosols are present, but that the OClO layer is limited to those levels where the temperature approaches that of the condensation of NAT. The NO_2 integrated column is 0.47 10^{15} mol./cm^2 that is in agreement with ground-based measurements (Figure 1.). The one of OClO is 1.5 10^{13} mol./cm^2, that is of the order of the largest recorded in Arctic or Antarctica (Solomon et al., 1990).

The results of the 11 flights are available, from which we anticipate being able to investigate the evolution of the volcanic aerosol layer, conditions of formation of PSCs, size and number densities of their particles and their consequences on NO_2 and OClO as well as to check the influence of the aerosol layer on total column measurements of the same constituents by ground-based spectrometry.

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ACKNOWLEDGMENTS

The design of the balloon and its equipment as well as operations, were conducted by the CNES Balloon Division in cooperation with the personnel of ESRANGE. Meteorological data were provided by the Norwegian Institute for Air Research.

This work is supported by the Programme Atmosphere Moyenne of CNRS in France and by the Commission of European Communities (Grant STEP CT91 0141).

They are all gratefully acknowledged.