UV-Visible Measurements of Stratospheric Trace Constituents at Harestua, Norway During SESAME

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INTRODUCTION

One of the objectives of the Second European Stratospheric Arctic and Mid-latitude Experiment (SESAME) was to study the connection between the high and the middle latitudes. To this aim, it was decided to increase the number of available ground-based instruments for stratospheric monitoring around 60°N. This would fill the geographical gap between the observing sites situated beyond the Arctic circle and the mid-latitude Alpine stations of the Network for the Detection of Stratospheric Changes (NDSC) located around 45°N. In total, ground-based instruments were operated at about 35 sites, mainly in Europe.

The present poster reports on ground-based UV-visible measurements of stratospheric OClO, NO_2 and O_3 performed during SESAME by the Belgian Institute for Space Aeronomy (BISA) and the "Université Libre de Bruxelles" (ULB) at Harestua, Norway (60°N, 10°E). Observations of the zenith scattered light were carried out from the middle of January until the end of March 1994 (SESAME phase I) and from the beginning of November 1994 until the end of March 1995 (SESAME phase III). During January-February 1995, the NO_2 and O_3 measurements were complemented by direct Sun observations of the same constituents using a Fourier transform UV-visible spectrometer. During both winter periods, significant OClO signatures were observed in January and March. As expected, the abundance of OClO is correlated to the potential vorticity at 475 K, the largest OClO columns being seen inside the polar vortex. There is a general anti-correlation between OClO and NO_2, large OClO contents appearing to be mainly driven by the stratospheric temperature.

MEASUREMENTS

Most of the measurements were carried out using two grating spectrographs looking at the zenith-scattered sunlight. In addition, some direct Sun measurements were performed between January and March 1995 using a Fourier transform spectrometer operated in the UV-visible range. During the third phase of SESAME, the UV-visible observations were complemented by FTIR measurements performed by the Sweedish Environmental Institute (Galle et al., 1995; Arlander et al., 1995).

Details regarding the grating instrument were given in Van Roozendael et al. (1995). It is based on two commercial spectrographs (ORIEL, MultiSpec) mounted together in a waterproof protection case thermally regulated. One of these spectrometers is equipped with an EG&G CCD detector (256x1024 pixels) cooled to -56°C and covers the spectral range from 340 to 395 nm. It was dedicated to the OClO measurements. The second spectrometer uses, as a detector, an EG&G Reticon photodiode array (1024 pixels) cooled to -40°C. Covering the region from 400 to 560 nm, it allows the measurement of NO_2 as well as O_3 in the Chappuis bands. Both detectors and spectrometers were carefully characterized in the laboratory before being operated in the field with the aim to identify and quantify possible sources of instrumental artefacts (Van Roozendael et al., in preparation). The data analysis is

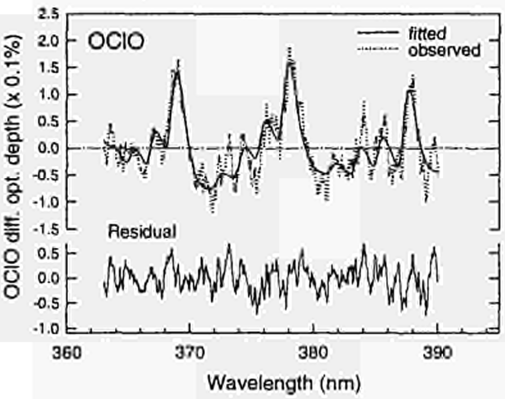

Fig. 2 Typical example of fitted and observed optical depths for OClO shown together with residuals, as measured with the BISA instrument (91° SZA).

based on the well known method of differential absorption spectroscopy (DOAS). A twilight spectrum, a control spectrum, absorption cross sections of NO_2, O_3, O_4, H_2O and OClO as well as a pseudo cross section for the Ring effect are fitted together using a coupled linear/non-linear least-squares algorithm written at the BISA. The conversion from slant to vertical columns is obtained using air-mass factors (AMFs) calculated according to *Solomon et al.* (1987). For NO_2 and O_3 retrievals, a single reference spectrum is used. On the contrary, OClO data are analysed by reference to a control spectrum taken at 82° SZA each twilight. This procedure minimises the instrumental noise that might be introduced by slight mechanical or thermal instabilities. Fig. 1 presents a typical OClO fits showing the fitted and observed differential optical depth of each constituent, after removal of all other absorption signatures. Residual levels displayed on the bottom illustrate the quality of the measurements. The BISA instrument has been qualified for the NDSC (NO_2 measurements) after participation to the Intercomparison of UV-visible spectrometers for measurements of stratospheric NO_2 for the Network for the Detection of Stratospheric Change held in Lauder during the period May 12-23, 1992 (Hofmann et al., 1995). In addition, it took part to the UV-visible instrument intercomparison organised by the Aberystwyth University at Camborne (Wales) during September 1995 as part of SESAME activities.

The mobile Fourier Transform Spectrometer (FTS) *BRUKER IFS120M* was operated at the Harestua observatory from 20 January to 3 March 1995. The optical design of the instrument is given in Fig. 2. A Fourier transform spectrometer combines the advantages of a large spectral interval investigated in one scan and a built-in wavenumber calibration. The detector used was a GaP diode, which allowed to record the 20000 to 30000 cm^{-1} spectral region suitable to measure O_3 and NO_2 column amounts. Although interferograms were recorded at various resolutions during the campaign, the results shown here were obtained from spectra calculated for a resolution of 64

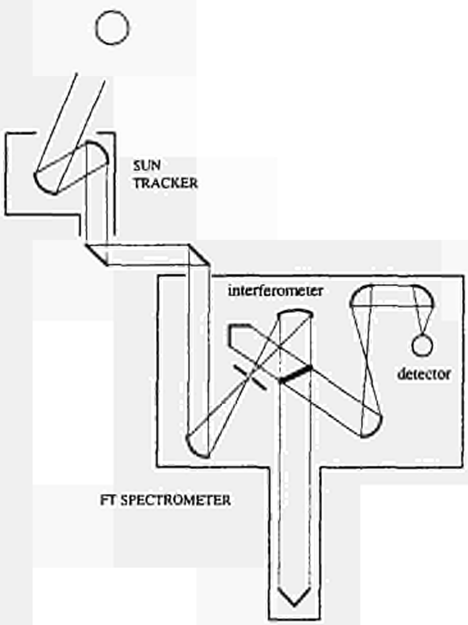

Fig. 1 Schematic description of the Fourier transform spectrometer *BRUKER IFS120M*.

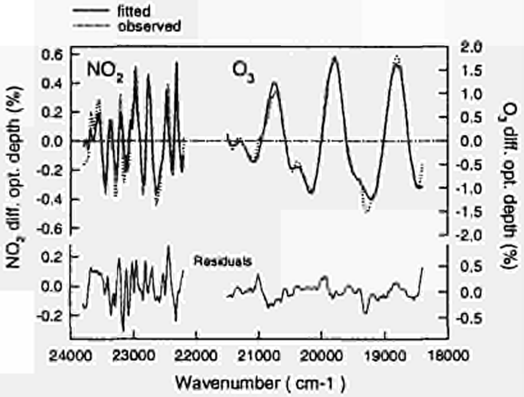

Fig. 3 Example of NO_2 and O_3 fits obtained with the FTS *BRUKER IFS120M* in direct-sun geometry.

cm^{-1} (0.6 nm at 300 nm). The spectral regions used to derive the O_3 and NO_2 total columns are respectively 18400-21500 cm^{-1} and 22200-23800 cm^{-1}. Fig. 3 shows an example of fitted and observed differential optical depths together with residuals for NO_2 and O_3, as obtained with the analysis programme. Although residuals are still larger than with the grating spectrometers, these first reported FTS measurements of stratospheric species in the UV-visible range are in good agreement with co-located instrument results.

RESULTS AND DISCUSSION

Fig. 4 and Fig. 5 show a summary of the observations achieved during SESAME phase I and phase III. Time series of NO_2, OClO and O_3 column amounts are presented together with 50 hPa temperatures and potential vorticities (PV) produced by the European Centre for Medium-term Weather Forecast (ECMWF) at the 475 and 550 K isentropic levels. The limits of the vortex at both isentropic levels are indicated by horizontal lines (PV contours at 36 and 102×10^{-6} K m^2/kg s).

The difference in the 50 hPa temperature evolution between SESAME phases I and III is striking. Much lower stratospheric temperatures were experienced during phase III, especially from mid-December until the end of January when the northern hemisphere minimum temperature (NH min) remained close to or below the limit of PSC type II (ICE) formation. Temperature and PV at 475 K and 550 K are anti-correlated, episodes of low temperature at Harestua corresponding to excursions of the polar vortex above the site. Two significant inside-vortex episodes were monitored during phase I and phase III, both years at the end of January and mid-March. Note, in Fig. 5, the good agreement obtained between the direct Sun and zenith-sky measurements (both for NO_2 and O_3 column amounts).

NO_2 column amounts are significantly lower in January-February 1995 than during the same period in 1994. This behaviour can be explained by the coldness of the stratosphere during this period which is believed to favour NOx conversion to their reservoirs as well as possible denitrification of the polar stratosphere by condensation of nitric acid on polar stratospheric clouds (PSCs). Corollary, the OClO contents inside the polar vortex during January 1995 are quite large as compared to January 1994 indicating larger chlorine activation in January 1995. Most OClO observations are obtained inside or at the edge of the polar vortex which indicates that the activation is essentially limited to the vortex region. However significant OClO amounts were detected outside the vortex as well, for example at the beginning of January 1995.

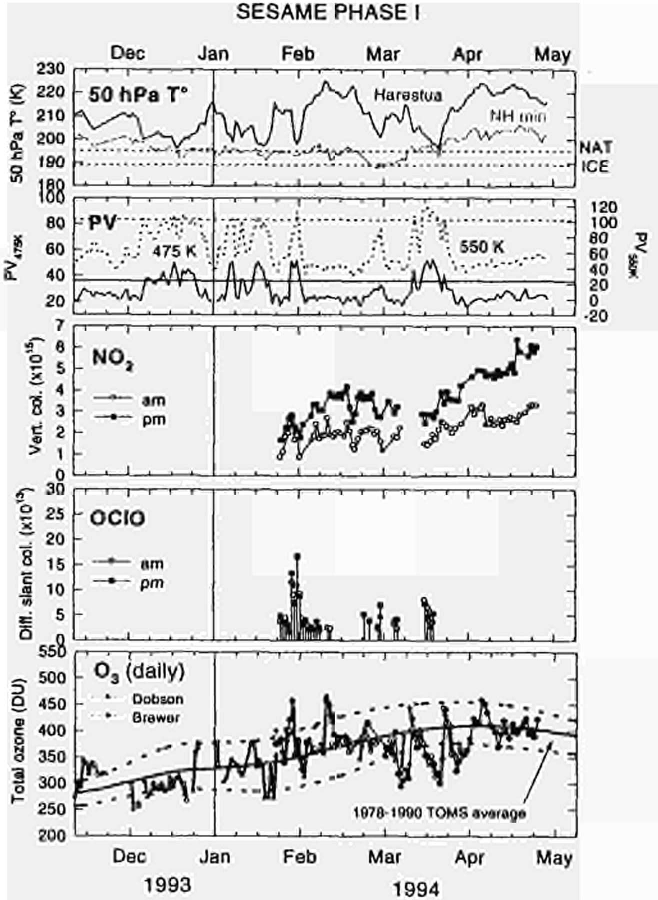

Fig. 4 Time series of NO_2, OClO and O_3 observations performed at the Harestua station during SESAME phase I, together with stratospheric temperatures and potential vorticities.

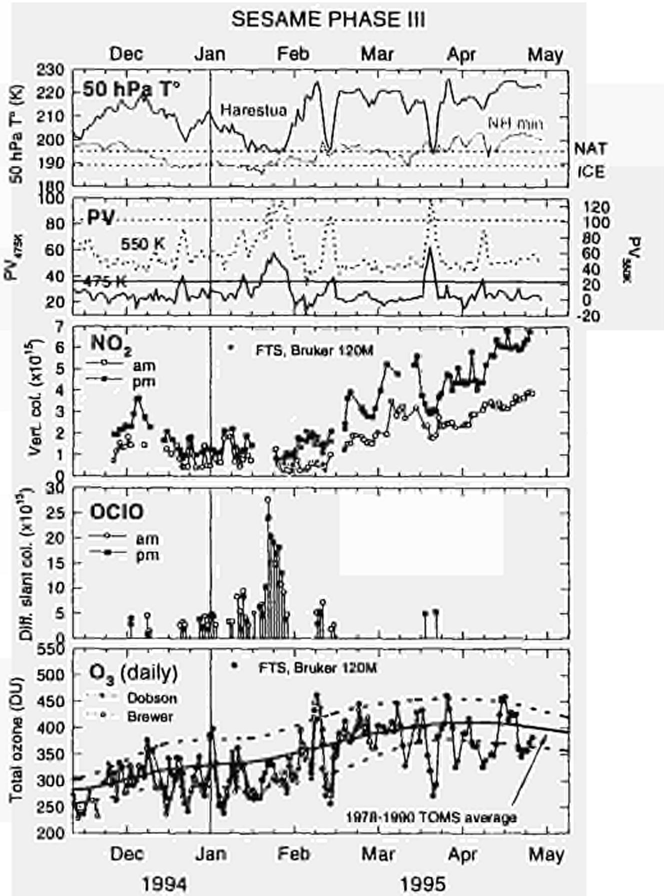

Fig. 5 Same as Fig. 4, but for SESAME phase III.

The O_3 time series are shown on the bottom of Fig. 4 and 5. Note the good agreement between DOAS results and Dobson or Brewer measurements taken in Oslo (60 km away from Harestua). The SESAME results are compared with the long term (1978-1990) TOMS climatology above Oslo (average of monthly means in solid line, minimum and maximum values in dotted lines). It is interesting to point out that O_3 values were rather low in March 1994 and 1995 when the polar vortex was moving above Harestua (as compared to the 12-year TOMS climatology. However tropopause heights calculated from O_3 soundings recorded at the nearby station of Gardermon, show that the O_3 was closely related to the tropopause pressure during this period. Moreover, 3-D model calculations by Chipperfield et al. (this issue) strongly suggest a very poor contribution from chemical origin to the low O_3 values measured above Harestua during SESAME phase III.

More details on the interpretation of the ground-based UV-visible and FTIR measurements performed at Harestua during the third phase of SESAME can be found in (Arlander et al., 1995).

CONCLUSION

UV-visible measurements of OClO, NO_2 and O_3 column amount were performed at Harestua, 60° N during SESAME phases I and III. The good agreement of the FTS measurements with the other instrument results demonstrates the feasibility of the stratospheric UV-visible measurements as a new application of the Fourier Transform Spectroscopy. Higher resolution spectra (8 and 0.5 cm^{-1}) were recorded and are still to be investigated. From these spectra, more accurate results for NO_2 and O_3 are expected as well as measurements of additional molecules (possibly OClO and BrO).

A first comparison of the results obtained during the two SESAME winter periods shows large differences in the NO_2 and OClO column amounts reflecting large differences in the stratospheric meteorology. Low O_3 column amounts (as compared to the long term climatology derived from TOMS data) are measured in March both during SESAME phase I and phase III.

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