Stratospheric chlorine activation in the austral and boreal winters 1995 through 2000 as derived from GOME OClO measurements

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Abstract. Measurements of OClO total column amounts by the GOME instrument (Global Ozone Monitoring Experiment) in the austral and boreal winter stratospheres from 1995 through 2000 are presented. GOME is a four channel UV/visible spectrometer (240 nm – 790 nm) deployed on the polar orbiting European ERS-2 satellite since April 1995. Previous studies have shown that the observations of OClO can serve as an indicator for a stratospheric chlorine activation. The GOME observations allows us to infer the first global data set of OClO, and to study continuous time series of its occurrence in both winter stratospheres. It is found that, while OClO regularly occurs over Antarctica in similar amounts and seasonal timing during the different winters, its occurrence is much more variable in the Arctic winter stratosphere. About 40% larger OClO column amounts are found in the Antarctic polar stratosphere than in its northern counterpart.

Introduction

Until today important improvements in the understanding of the stratospheric ozone chemistry have been made by applying spectroscopic techniques like UV/vis absorption measurements of stratospheric OClO [e.g., *Solomon et al.,* 1987]. Under perturbed (i.e., chlorine activated) stratospheric conditions column amounts of OClO were monitored utilizing the characteristic absorption features of the OClO molecule in the near UV spectral range. OClO is primarily formed via the reaction of BrO and ClO:

$$\begin{aligned} BrO + ClO &\rightarrow Br + OClO \quad (\approx 30\%) \quad (1)\\ &\rightarrow Br + ClOO \quad (\approx 60\%) \quad (2)\\ &\rightarrow BrCl + O_2 \quad (\approx 10\%) \quad (3) \end{aligned}$$

Since stratospheric BrO concentrations vary only slightly, the availability of ClO is limiting the formation of OClO. In consequence, OClO can serve as an indicator for the amount of stratospheric ClO, i.e., the chlorine activation [*Solomon et al.,* 1987].

Experimental set-up and data analysis

The GOME instrument measures sun light scattered by the atmosphere and/or reflected back from the earth's surface from 240 to 790 nm with moderate spectral resolution (full width at half maximum: 0.2 to 0.4 nm). GOME's OClO observations provide many advantages compared to previous OClO measurements, mainly because OClO is monitored with a global coverage at a moderately good spatial (ground pixel size 320 km East/West by 40 km North/South), and temporal resolution (see also [ESA 1995]). The raw spectra, are analyzed using the differential optical absorption spectroscopy (DOAS) [*Platt,* 1994]. In brief, the measured spectra (wavelength range: 363 nm to 393 nm) are modeled with a non-linear fitting routine that suitably weights the absorption spectra of atmospheric trace gases [*Stutz and Platt,* 1996]. From the inferred absorption the along the light path integrated trace gas concentration (slant column density, SCD) is calculated (details are described in Wagner [1999] and Wagner et al. [2000].

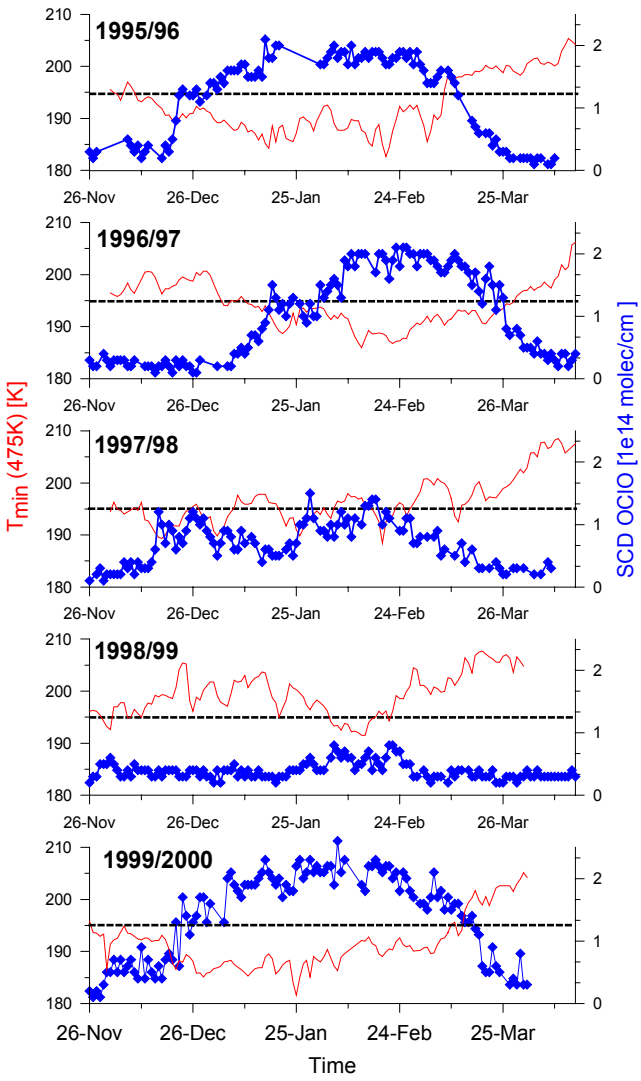

Figure 1. Time series of the daily maximum OClO-SCDs (at a SZA of 90°) observed by GOME (thick lines with diamonds) together with the minimum stratospheric temperatures at the 475 K level [ECMWF, 2000] (thin lines) for the five monitored Arctic winters, since the ERS-2 satellite has been launched in 1995. The dashed line indicates the formation threshold for Type I-PSCs (NAT) at the 475 K level.

Since both the absorption path through the atmosphere and the OClO photolysis depend strongly on the solar zenith angle (SZA), for a quantitative interpretation of the GOME OClO measurements we selected GOME measurements at a fixed SZA (90°).

Observations

In Figure 1 time series of the daily maximum OClO-SCD values for SZA=90° together with minimum stratospheric temperatures around 19 km (at the 475 K level taken from the ECMWF analysis [*ECMWF,* 2000]) are shown for the Arctic winters 1995/96 through 1999/2000. The OClO observations indicate that chlorine activation occurred in each of the boreal winters, but show a large year to year variability related to the meteorological conditions. In the 'cold' winters 1995/96, 1996/97, and 1999/2000 enhanced OClO SCDs continuously prevailed over periods of several months. In contrast in the 'warm' winters 1997/98 and 1998/99 lower OClO-SCDs and more sporadic chlorine activation were observed. In winter 1997/98, however, the chlorine activation took place until early March.

In contrast, the temporal evolution of the stratospheric temperatures over Antarctica is very regular in individual winters [*WMO,* 1989*; Solomon,* 1999]. Accordingly, the seasonal timing for the daily maximum OClO-SCDs does not show much year to year variability (Figure 2). A strong chlorine activation in the austral winter and spring vortex occurs typically for a period of about 4 months, a finding being in agreement with *Waters et al.* [1993] and *Santee et al.* [1995].

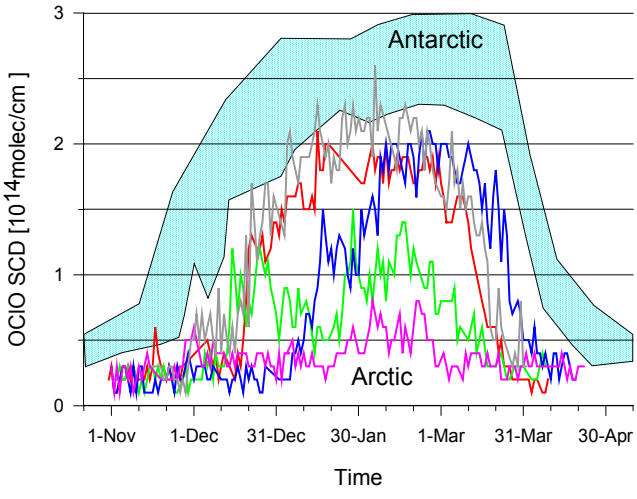

Figure 2. Comparison of the maximum daily OClO-SCD (SZA = 90°), measured over the Arctic and the Antarctic in all consecutive winters 1995 through 2000. The Antarctic values are represented by an envelope of all winters; for comparison with the measurements over the Arctic, they are shifted by 6 months.

Conclusions

From GOME OClO observations in both hemispheres (1995 to 2000) we draw the following conclusions:

- The maximum OClO values in the southern hemisphere are about 30% larger than those in the northern hemisphere (see also *Miller et al.* [1999]) - besides a temperature effect (≈15%) of OClO formation - most probably due to a larger altitude range of chlorine activation in the Antarctic compared to the Arctic winter stratosphere.
- Large differences in the OClO SCDs were found between Arctic winters reflecting the high variability of the meteorological conditions. In the 'cold' winters 1995/96, 1996/97, and 1999/2000 enhanced OClO SCDs were continuously observed during several months (with the highest values in 1999/2000); in the 'warm' winters 1997/98 and 1998/99 enhanced OClO SCDs occurred only sporadically and were significantly smaller.
- The duration of the chlorine activation in the Arctic is in the range from a few days to about 3 months and thus at least 1 month shorter than in the southern hemisphere.
- In the southern hemisphere the temporal evolution and the magnitude of the OClO SCDs are very similar for all winters.

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