GOME Observations of Stratospheric Trace Gas Distributions during the Splitting Vortex Event in the Antarctic Winter of 2002. Part I: Measurements

Andreas Richter, Folkard Wittrock, and Mark Weber

Institute for Environmental Physics, University of Bremen, Bremen, Germany

Steffen Beirle, Sven Kühl, Ulrich Platt, Thomas Wagner, and Walburga Wilms-Grabe

Institute for Environmental Physics, University of Heidelberg, Heidelberg, Germany

John P. Burrows

Institute for Environmental Physics, University of Bremen, Bremen, Germany

ABSTRACT

Measurements from the Global Ozone Monitoring Experiment (GOME) are used to study the chemical evolution of the stratosphere during the unusual 2002 winter in the Southern Hemisphere. The results show that chlorine activation as indicated by OClO columns was similar to previous years in the vortex until the major warming on 26 September 2002 after which it decreased rapidly. Similarly, NO_2 columns were only slightly larger than in previous years before the warming, indicating strong denoxification and probably also denitrification. After the warming, very large NO_2 columns were observed for a few days, which then decreased again as the vortex reestablished itself until the final warming. Ozone columns were much larger than in any previous year from September onward, mainly as a result of the unusual dynamical situation. Analysis of the global long-term time series of GOME measurements since 1996 provides a unique opportunity to set the austral winter 2002 into perspective. The GOME data reveal the large difference in variability of chlorine activation between the two hemispheres, whereas denoxification shows surprisingly little variation from year to year in both hemispheres. However, NO_2 depletion in the Southern Hemisphere is usually sustained for about one month longer in the Antarctic stratosphere as a result of the stable vortex. Compared to the observations in the Northern Hemisphere, the austral winter 2002 was still stable and cold and had a high potential for chemical ozone destruction.

1. Introduction

Since the beginning of systematic measurements in 1957, the polar vortex formed each winter in the Southern Hemisphere (SH) has been very stable and reproducible in terms of area covered, temperatures reached, and time of final breakup. Unlike in the vortex in the Northern Hemisphere, major warmings before the final warmings have not been observed (Roscoe et al. 2005), which probably is the result of less planetary wave activity resulting from the much smaller sea–land contrast in the SH. The darkness and low temperatures in the vortex are the prerequisite for the strong activation of chlorine species and the subsequent massive ozone loss in polar spring that leads to the well-known Antarctic ozone hole. Although stratospheric chlorine levels in the Northern Hemisphere are comparable to those in the south, and locally chemical ozone loss rates are of the same magnitude in both hemispheres, the polar vortex shows much larger variability in size, shape, and intensity due to the stronger planetary wave activity. Consequently, stratospheric temperatures are higher, probabilities for polar stratospheric clouds (PSCs) are lower (Pawson et al. 1995), and the vortex breaks up earlier, which until now has prevented the formation of a similar ozone hole in the Northern Hemisphere.

In the austral winter 2002, the polar vortex in the Southern Hemisphere was less stable than usual and, for the first time, a major warming and a split vortex were observed in September for altitudes above 500-K potential temperature. Although the vortex later reestablished itself, total ozone values were much larger than usual, raising the question if this is the first sign of a change in winter patterns or if it is just an interesting, but unusual, singular event.

Corresponding author address: Dr. Andreas Richter, Institute of Environmental Physics, University of Bremen, P.O. Box 33 04 40, D-28334 Bremen, Germany.
E-mail: andreas.richter@iup.physik.uni-bremen.de

Satellites provide unique information due to the temporal and spatial coverage of the observations. In this paper, measurements from the Global Ozone Monitoring Experiment (GOME) are used to characterize the chemical evolution of the air masses during the split vortex event in the SH spring 2002. By comparing O_3, NO_2, and OClO columns from that year to results from the seven years of GOME measurements in both hemispheres, the SH winter 2002 is put into perspective and similarities and differences between the two hemispheres are discussed.

Throughout the paper, NO_2 is used as an indicator of denoxification. Whether or not NO_y has been removed permanently from the stratosphere (denitrification) cannot be determined from the NO_2 columns directly. However, the time required for the recovery of the NO_2 columns in spring is related to the degree of denitrification in the air masses. The second quantity discussed in detail is OClO, a quantitative indicator for chlorine activation albeit not a direct measurement. In the stratosphere, formation of OClO is almost exclusively through thereaction of BrO and ClO (Solomon et al. 1987; Toumi 1994). In the sunlit stratosphere, BrO is the main form of inorganic bromine, both inside and outside the polar vortex, and concentrations vary much less than those of ClO. Thus, OClO concentrations are determined by ClO concentrations and can serve as a quantitative indicator of chlorine activation. Only at low sun [solar zenith angle (SZA) $>92°$] does the relationship between ClO and OClO become nonlinear as a result of twilight photochemistry [see the discussion in Sessler et al. (1995), Schiller and Wahner (1996), and Tørnqvist et al. (2002)], and a quantitative relationship can no longer be established.

2. Instrument and data analysis

The GOME instrument is a UV/visible grating spectrometer on board the European Remote Sensing (ERS) satellite, *ERS-2*, covering the wavelength range of 280–800 nm with a spectral resolution of 0.2–0.4 nm (Bednarz 1995). GOME observes the radiance scattered from the atmosphere and reflected on the surface in near-nadir viewing geometry and also takes a measurement of the extraterrestrial solar irradiance once per day. *ERS-2* was launched in April 1995, and GOME data are available from July 1995 onward. As a result of the failure of the last tape recorder on *ERS-2* on 22 June 2003 only very limited data is available after that date.

From the radiance and irradiance measurements, atmospheric columns of a number of trace species can be determined using the well-known differential optical absorption spectroscopy (DOAS) technique (Platt 1994; Burrows et al. 1999). Briefly, the analysis consists of two steps: 1) the determination of the integrated absorber content in the optical depth derived from the measured radiance and solar irradiance [slant column (SC)] and 2) the correction for the effective light path and conversion to the total amount of the absorber integrated vertically through the atmosphere [vertical column (VC)]. The latter is derived through division by an enhancement or airmass factor (AMF), computed by a radiative transfer model for a standard atmosphere: AMF = SC / VC. For OClO, conversion to vertical columns is complicated by rapid photolysis, the change in SZA over one GOME pixel, and also the uncertainty in the vertical profile. Therefore, slant columns at 90° SZA are used throughout the paper as in previous studies (Wagner et al. 2001; Wittrock et al. 1999), which allows meaningful comparison of OClO columns measured at different times but does not give the vertical column.

In this study, GOME measurements of O_3, NO_2, and OClO are used. While the total ozone columns are taken from the operational level-2 data product (DLR2002), both NO_2 and OClO are retrieved from the raw spectra using a DOAS retrieval. Details on the spectral analysis of GOME measurements for NO_2 and OClO can be found elsewhere (Burrows et al. 1999; Richter and Burrows 2002; Leue et al. 2001; Wittrock et al. 1999; Wagner et al. 2001, 2002). However, there is one GOME-specific problem that needs to be discussed in more detail, as it affects the interpretation of long time series such as those used in this work. The diffuser used for the GOME solar irradiance measurements introduces spectral artifacts that lead to offsets in the measured slant columns that vary over time. Therefore, a correction must be applied to the slant columns for each day as described below. A detailed discussion of the GOME diffuser problem can be found in Richter et al. (2002) and Martin et al. (2002).

As no OClO is expected in the sunlit tropical stratosphere, an offset was subtracted from each orbit of OClO data to force the measurements to 0 over the equator. This is equivalent to the approach used in previous studies where OClO was retrieved using an earthshine measurement at high solar zenith angle as background spectrum (Wagner et al. 2001).

For NO_2 the situation is less simple as significant stratospheric column amounts are to be expected over the Tropics. To avoid the impact of polluted air masses, a clean region in the Pacific (5°S–5°N, 130°–170°E) was selected and for each day the averaged GOME measurement determined. Assuming that for this area a vertical column of 2×10^{15} molecules (mol) cm^{-2} is appropriate independent of season, the difference between the measured value and the assumed slant column was subtracted from all measured GOME NO_2 slant columns of that day to compensate the diffuser offset. The value chosen is representative of NO_2 columns measured in tropical regions from the ground (Noxon 1979) and from ships (Senne et al. 1996). Some seasonal and interannual deviations are to be expected, but several years of ground-based measurements in

Tarawa Kiribati, indicate that the amplitude of such variations is below 1×10^{15} mol cm^{-2} (P. Johnston 2003, personal communication).

Clearly, the assumption of a constant value over the reference area introduces an uncertainty into the analysis of the slant columns by adding an absolute error SC_err to the slant column. However, the impact is small for measurements at high latitudes where the solar zenith angle and therefore the light path enhancement (AMF) with respect to the VC is larger:

$$VC\_err = SC\_err/AMF.$$

Any changes in tropical NO$_2$ columns with season or from year to year will have an impact on the NO$_2$ columns derived at high latitudes. Assuming an uncertainty in the tropical vertical column of 1×10^{15} mol cm^{-2} and considering the airmass factors in the Tropics and in high latitudes, the error introduced at 70°–80° latitude is about 5×10^{14} mol cm^{-2} in summer and smaller than 1×10^{14} in winter.

3. GOME results for the SH winter of 2002

In early September, the polar vortex was stable and relatively well centered over the South Pole (Fig. 1). According to European Centre for Medium-Range Forecasts (ECMWF) and U.K. Met Office (UKMO) analyses, temperatures were low enough for PSC formation, which is confirmed by Polar Ozone and Aerosol Measurement (POAM) and Scanning Imaging Absorption Spectrometer for Atmospheric Chartography (SCIAMACHY) observations (Nedoluha et al. 2003; von Savigny et al. 2005). Figure 2 shows the evolution of the potential daily Antarctic PSC volume for the winter 2002, confirming that stratospheric temperatures were below PSC existence threshold until around the time of the sudden warming event. Compared to earlier winters, the daily PSC volume remained high throughout the winter but was at or slightly below the lower limit of the observed range during the last decade. As in recent years, daily minimum ozone values observed by GOME south of 50°S decreased rapidly by early August (Fig. 3). This sharp decrease is most likely related to the combination of rapid catalytic chemical depletion setting in after the sun returns into the polar region and inhibited transport of ozone rich air from midlatitudes (Sinnhuber et al. 2003). GOME NO$_2$ columns within the vortex were also low (Fig. 1) as a result of denoxification and probably denitrification. At the same time, large OClO columns were observed throughout the vortex (Fig. 1), indicating that chlorine was well activated at that time.

As a result of unusually strong planetary wave activity, a major stratospheric warming started on 20 September, rapidly extending down to the lower stratosphere and essentially splitting the upper part of the polar vortex into two parts. On 27 September, the vortex above 500 K had separated, and one of the parts began to dissipate by moving equatorward and mixing with low-latitude air. At that time, only marginal OClO could be detected by GOME close to the South Pole, indicating that chlorine activation was unusually low there. This rapid disappearance of OClO is surprising as ClO and OClO should not disappear instantaneously, even in the absence of PSCs, but rather decay over a period of several days. However, as discussed above, OClO rapidly photolyzes during daytime and therefore can only be observed at twilight. Given the orbital characteristic of *ERS-2*, this implies that chlorine activation in the strongly displaced parts of the vortex might not have been picked up, as the solar elevation is too large at the time of overpass. Nevertheless, large OClO concentrations can be observed also at higher sun, as demonstrated in the measurements from 1 September, and therefore chlorine activation must have been small by the end of September. This is in qualitative agreement with the model results of Grooß et al. (2005), who report almost complete deactivation within the split vortex on 24 September and also ground-based measurements by Frieß et al. (2003). Although the vortex reestablished itself over the South Pole by mid October, no OClO could be detected, indicating that significant chlorine activation indeed ended with the major warming.

GOME NO$_2$ columns in the vortex air increased but were still much lower than in the surrounding midlatitude air. As is to be expected from the different vertical profiles of the two species, the ozone and NO$_2$ minima are somewhat displaced, in agreement with the vertical tilt of the polar vortex. To give an impression of the vertical vortex displacement, the 990-K vortex edge [defined as -36 modified potential vorticity units (MPVU), where 1 MPVU = 1.0×10^{-6} km^2 s^{-1} K kg^{-1}] is overlaid over the NO$_2$ columns in Fig. 1. Inbetween the two parts of the vortex, in a region with unusually high temperatures, the NO$_2$ columns increased rapidly to the largest values observed in the whole hemisphere (Fig. 1). Similar patches of high NO$_2$ were also observed in other regions at the vortex edge and persisted for several days. Episodes of very large NO$_2$ columns were also observed from the ground as the air masses moved over the stations (Frieß et al. 2003). Although the bulk of these high NO$_2$ concentrations is probably found at a higher altitude than the ozone maximum, they still may contribute to rapid ozone destruction in the affected air masses. This is illustrated by the model simulations of Grooß et al. (2005), where high ozone depletion rates are found in midlatitude air masses displaced over the South Pole, mostly due to NO$_x$ catalytic cycles.

Localized events of very large NO$_2$ columns can often be observed during the final warming in the SH and are related to thermal decay and rapid photolysis of NO$_2$ reservoirs of warm midlatitude air when transported into polar day. However, as the warming in September was unusually early, photolysis rates in the polar region were not significantly larger than in midlati-
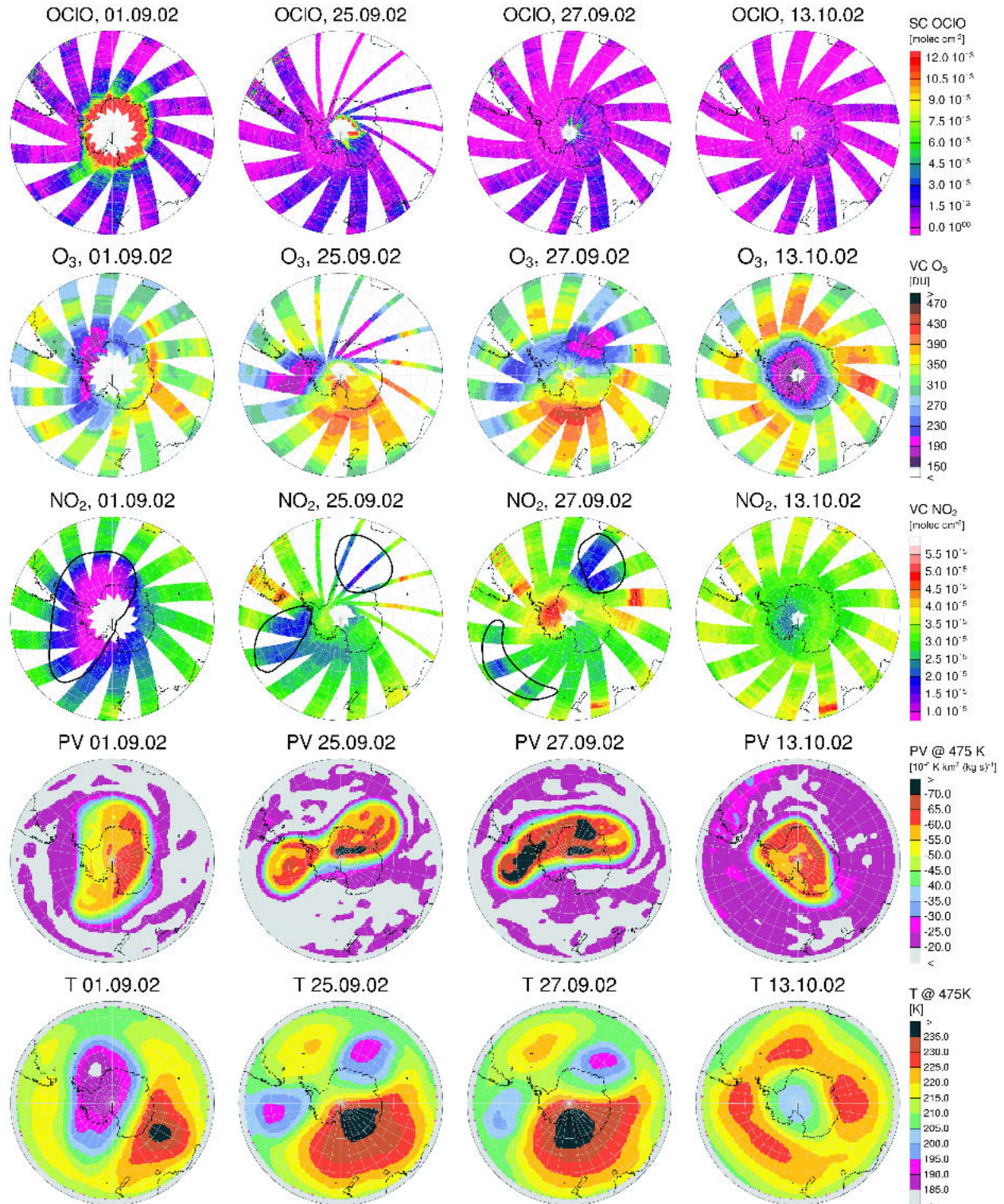
FIG. 1. Selected daily images of GOME trace gas measurements compared to ECMWF temperature and potential vorticity at 475 K. OClO columns are slant columns in mol cm^{-2} and not corrected for light path enhancement. NO$_2$ is given as vertical columns in mol cm^{-2}, and ozone is given as vertical column in Dobson units (1 DU = 2.89 × 10^{16} molecules cm^{-2}). The heavy line in the NO$_2$ plots is the 36-MPVU isoline at 990 K.
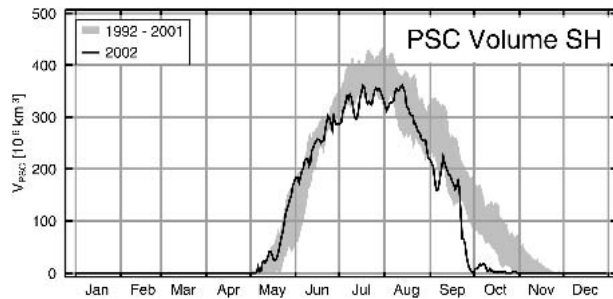
FIG. 2. Daily PSC volume at extratropical latitudes as approximated by the temperature region between 100 and 4 hPa, where stratospheric temperature according to UKMO meteorological analyses was below T_{NAT}, a threshold temperature for possible existence of polar stratospheric clouds. For the determination of T_{NAT}, the formula of Hanson and Mauersberger (1988) was used, and typical mixing ratios of 5–9 ppb for HNO_3 and 5 ppm for H_2O have been assumed. Shading indicates the range observed during the last decade excluding 2002. The solid line represents data from 2002.

tudes; therefore photolysis cannot fully explain the observations. Also, comparison with ECMWF and UKMO analysis data shows that not all of the maxima in NO_2 coincide with the maxima in temperature or regions with strong descent as one might expect. In addition, many of the events are located in the region of the vortex edge (see Fig. 1), and from the column measurement it is not clear if they belong to vortex or nonvortex air masses. A fully quantitative understanding of these events will therefore have to rely on model simulations and the availability of vertical profiles of NO_2 and other members of the NO_y family from SCIAMACHY and Michelson Interferometer for Passive Atmospheric Sounding (MIPAS).

GOME minimum polar ozone was found to have rapidly increased from around 140 DU (20 September) to about 180 DU at the time of the observed vortex splitting (28 September). This represents a strong and unusual departure from prior years when minimum ozone bottoms out near 100–110 DU during that time of the year.
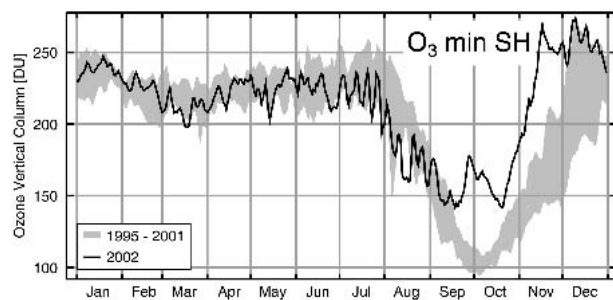

FIG. 3. Daily minimum GOME total ozone observed poleward of 50°S. Shading indicates the range observed during the period Jul 1995–Dec 2001. The solid line represents data from 2002.

In October, the surviving part of the vortex reestablished itself over the South Pole, temperatures dropped, and ozone columns remained small. Minimum ozone decreased again by 40 DU, which is mainly explained by the effect of the vertical realignment of the vortex as discussed in Hoppel et al. (2003), and started to increase after the final warming at the end of October. It remained above the range observed in prior years from October until early December. No OClO could be detected in October, in agreement with model results (Grooß et al. 2005) showing that chlorine was no longer in its active forms at that time. NO_2 columns over the South Pole were found to be lower than outside the vortex, but the differences were small and no strong denoxification was apparent.

4. Comparison with previous winters and the Northern Hemisphere

In a sense, the Southern Hemisphere winter of 2002 was much closer to winters in the Northern Hemisphere and, in fact, in some respects falls nicely between the two extremes (Weber et al. 2003). A series of minor warmings in the upper stratosphere was observed throughout the winter starting in May (Allen et al. 2003). This is in agreement with the high variability of the midlatitude eddy heat flux, which has been responsible for the increase in mid to high latitude ozone (Weber et al. 2003). However, the minimum temperature at 46 hPa (lowermost stratosphere) remained low as in prior years until the major warming event (Sinnhuber et al. 2003). This explains why the vortex was still relatively stable and much colder than in the Northern Hemisphere, and PSC covered large volumes for most of the winter period. It is therefore interesting to compare the GOME measurements of this particular winter with those of other years and also in the Northern Hemisphere.

a. OClO

As has been discussed in detail in Wagner et al. (2001, 2002) and Wittrock et al. (1999), OClO columns observed in the Antarctic winter and spring are larger than those observed in the Northern Hemisphere. Also, the variability of OClO is much larger in the Northern Hemisphere. This is illustrated in Fig. 4, where the range of 90° SZA OClO measurements from both hemispheres is shown for the last three winter months. It is important to note that, as a result of increasing day length, the latitude of 90° SZA measurements moves equatorward during the time period shown. In Fig. 4, the average of all 90° SZA measurements within the vortex (defined by a 36 PVU threshold on the 495-K level) are shown for each day. This differs slightly from the data presented in Wagner et al. (2002) where daily maximum OClO columns were discussed. Measurements from the year 2002 that are not included in the
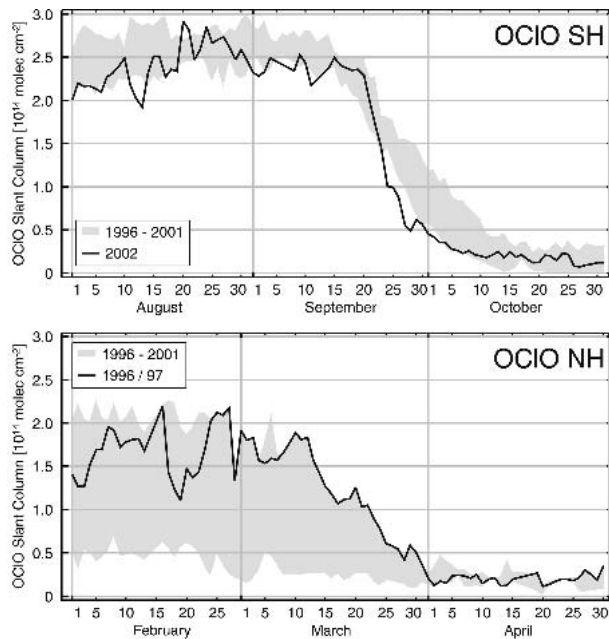
FIG. 4. OClO slant columns observed by GOME in (top) the Southern Hemisphere and (bottom) the Northern Hemisphere. The values given are the average of all measurements taken between 89° and 91° SZA where the PV on the 495-K level was above 36 PVU (see text for details). The shaded areas give the range of values observed from 1996 to 2002, the latter being excluded for the SH plot. Also shown are the results for individual winters: (top) 2002 and (bottom) 1996/97.

shaded area in Fig. 4a indicate that chlorine activation in that year ended approximately one week earlier than in any other year of the GOME time series. OClO columns before the warming were similar to those observed in previous years but slightly more variable, probably related to the observed anomalies in heat flux (Weber et al. 2003), which may have had an impact on temperature and PSC formation. In summary, GOME measurements show that chlorine activation in the vortex was comparable to previous years throughout the winter with the exception of the last week of September when activation ended rapidly. This should result in vortex total ozone loss typical of SH winters with some reduction at the end of September, in agreement with the results of the POAM measurements (Hoppel et al. 2003) and modeling studies of Sinnhuber et al. (2003) and Grooß et al. (2005).

b. NO_2

GOME measurements of total NO_2 in both hemispheres are summarized in Fig. 5. As in Fig. 4, the range of values observed from 1996 to 2002 is shown as the shaded area, and some specific years are plotted on top. For the Southern Hemisphere, both 2000 and 2002 data have been not been included in the shaded area. In contrast to OClO, NO_2 columns can be converted to vertical columns by applying an appropriate airmass factor, and

vertical columns averaged over all measurements between 70° and 80° latitude are given in the plots. The diurnal and seasonal variation of NO_2 is governed by photolysis of NO_2 itself and its main reservoir species (N_2O_5, HNO_3, and to a lesser extent $ClONO_2$). In polar spring, NO_2 concentrations can be further reduced if substantial denitrification has taken place and the reservoirs needed for NO_2 reformation are depleted, as is often the case in the SH.

As can be seen from Fig. 5, the year to year variability in NO_2 columns between 70° and 80°S is small. In particular, in autumn, the NO_2 columns from all seven years of GOME observations are nearly identical, emphasizing the fact that they are mainly controlled by photolysis, which is only a function of season. In spring, there is more variability, related to high NO_2 events similar to the one observed on 27 September 2002 and also to the time of final warming and increasing NO_2 columns, which varies from year to year. In summer, values are less variable than in spring but not as reproducible as in autumn, which might be related to some residual effect of denitrification in the vortex even after mixing with air from lower latitudes. From the beginning of August onward, NO_2 in the split vortex winter was larger than in previous years. At the end of September, values increased rapidly to about twice typical values, but came back to normal as the vortex reestab-
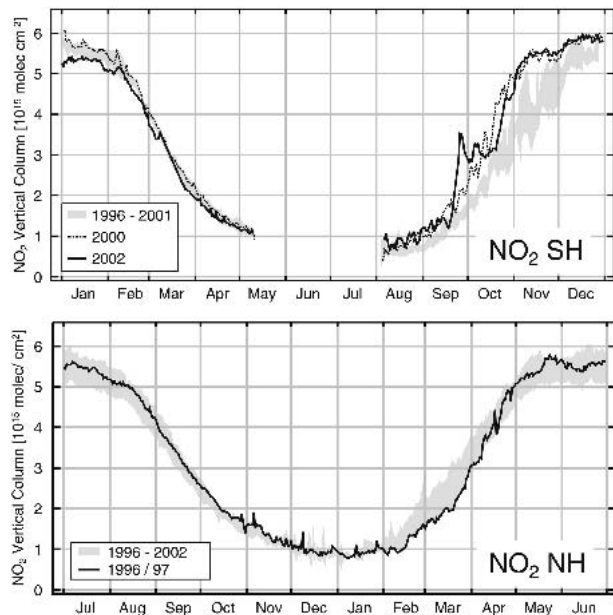

FIG. 5. Annual cycle of GOME NO_2 vertical columns averaged over 70°–80° (top) south and (bottom) north. The shaded areas give the range of values observed from 1996 to 2002; for the Southern Hemisphere the years 2000 and 2002 have been excluded. Also indicated are the values measured in some individual years: 2000 and 2002 for the southern Hemisphere and 1997 for the Northern Hemisphere. In the SH, no data are available in this latitude region for May, Jun, and Jul as the solar zenith angle at the time of GOME overpass is larger than the threshold value of 92°.

lished itself over the South Pole in mid October. They then increased in the final warming and remained clearly higher than usual for the rest of the year. Interestingly, the year 2000 also behaved quite unusually with respect to NO_2. With the exception of late September/early October, NO_2 columns were as large or even larger than those measured in 2002. Why NO_2 was high in 2000 is not fully understood—while it was relatively warm from mid September onward, which would explain part of the observations, the first part of the 2000 winter was rather cold and showed one of the largest PSC probabilities ever.

According to the multiyear GOME dataset, NO_2 columns in the NH behave quite similarly to those in the SH: high values in summer and low values in winter, small variability in autumn and somewhat larger variations in spring. Compared to the SH, year to year variations are larger in autumn and, in particular, in summer but not in spring. Total columns in summer are similar in both hemispheres, but spring recovery of NO_2 is delayed by more than one month in the SH as a result of strong denoxification and denitrification in the persistent polar vortex. Only the very large NO_2 values from September 2002 would be above the range of observed columns in March in the NH. In general, low NO_2 is correlated to low temperatures in spring, and in cold winters, such as 1996/97, NO_2 columns are small in the NH (see Fig. 5). The reason for the larger variability of NO_2 in the summer NH is not clear; it could be related to varying degrees of denitrification in the winter or to variations in transport patterns. Also, some impact of anthropogenic pollution cannot be excluded in the GOME data even at these latitudes, but its effects should be small.

For ozone, it has recently been shown by Fioletov and Shepherd (2003) that anomalies established in wintertime can persist throughout the year, and some of the observed differences in summer NO_2 values may also have their origin in wintertime chemistry. However, for NO_2 the situation is different from that for ozone, as its concentrations depend mainly on photochemistry and much less on transport processes.

5. Summary and conclusions

In this paper, GOME satellite measurements of ozone, NO_2, and OClO were used to characterize the unusual austral winter/spring of 2002, and the results were compared to the seven years of data from the instrument in both hemispheres. This long and homogeneous global dataset provides a unique opportunity to study to what extent the austral winter of 2002 was special and if it approached the situation commonly observed in the NH.

An early and rapid reduction in OClO columns was observed in the vortex after the major warming in September 2002, indicating unprecedented early deactiva-

tion of chlorine. This should effectively have stopped the chlorine-catalyzed ozone depletion after the warming. Compared to all other years of the GOME SH record, this was the earliest deactivation ever observed; however, compared to even the coldest winters in the Northern Hemisphere, activation was still large and persistent.

GOME data also show that the NO_2 column rose rapidly after the warming, and very large columns were observed by GOME over the South Pole for several days with values more than twice those observed in previous years. While high NO_2 columns are often observed during the final warming in the SH, the mechanism behind this is not yet fully understood for the early warming in 2002. As the vortex became reestablished, NO_2 columns decreased again and, while they never were as low as usual in the vortex, similar values were also observed in October 2000. The overall enhancement of NO_2 observed after the major warming indicates a potential for increased ozone destruction via the NO_x catalytic cycle, which in part could offset the reduction in chlorine activation.

In general, polar NO_2 columns observed by GOME from 1996 to 2002 show surprisingly little year to year variability in autumn and summer, in particular in the Southern Hemisphere. The values observed in both hemispheres are very similar in summer but systematically larger in the NH in the other seasons. The winter depletion in NO_2 is longer in the Southern Hemisphere than in the Northern Hemisphere by about one month, probably as a result of both lower temperatures and denitrification. Related to this time shift, a much larger variability in NO_2 is observed in the south during recovery when midlatitude air is mixed into the polar region.

From early September onward, GOME minimum polar ozone south of 50° was much larger than in previous years and never reached the low values typical for the "ozone hole." Most of this effect is reproduced by models treating ozone as a tracer, implying that the reason for the high ozone is dynamical rather than chemical (Hoppel et al. 2003; Sinnhuber et al. 2003). As the anomalies in PSC volume, chlorine activation, and NO_2 were most apparent after the warming, both denitrification and chemical ozone loss had probably been comparable to previous years up to that point.

A quantitative comparison of the results discussed in this paper with model simulations will be presented in W. Wilms-Grabe et al. (2003, unpublished manuscript). From the measurements alone, it can be concluded that although this winter was very unusual for the Southern Hemisphere, it still was much more stable, colder, and favorable to chemical ozone destruction than any Northern Hemispheric winter observed so far. The impact of 2002 on ozone trend studies is large in the polar region but also for midaltitudes; whether this is the first indication of a change in trend or just an isolated event will have to be decided in coming years.

REFERENCES

Allen, D. R., R. M. Bevilaqua, G. E. Nedoluha, C. E. Randall, and G. L. Manney, 2003: Unusual stratospheric transport and mixing during the 2002 Antarctic winter. *Geophys. Res. Lett.*, **30,** 1599, doi:10.1029/2003GL017117.

Bednarz, F., Ed., 1995: *The GOME Users Manual.* ESTEC Publishing Division, 123 pp.

Burrows, J. P., and Coauthors, 1999: The Global Ozone Monitoring Experiment (GOME): Mission concept and first scientific results. *J. Atmos. Sci.,* **56,** 151–175.

DLR, 2002: GOME Level 1 to 2 algorithms description. Iss./Rev. 3/A, Tech. Note ER-TN-DLR-GO-0025, DLR, Oberpfaffenhofen,Wessling, Germany, 58 pp.

Fioletov, V. E., and T. G. Shepherd, 2003: Seasonal persistence of midlatitude total ozone anomalies. *Geophys. Res. Lett.,* **30,** 1417, doi:10.1029/2002GL016739.

Frieβ, U., K. Kreher, P. V. Johnston, and U. Platt, 2005: Ground-based DOAS measurements of stratospheric trace gases at two Antarctic stations during the 2002 ozone hole period. *J. Atmos. Sci.,* **62,** 765–777.

Grooß, J.-U., P. Konopka, and R. Mueller, 2005: Ozone chemistry during the 2002 Antarctic vortex split. *J. Atmos. Sci.,* **62,** 860–870.

Hanson, D. R., and K. Mauersberger, 1988: Laboratory studies of the nitric acid trihydrate: Implications for the south polar stratosphere. *Geophys. Res. Lett.,* **15,** 855–858.

Hoppel, K., R. Bevilaqua, D. Allen, G. Nedoluha, and C. Randall, 2003: POAM III observations of the anomalous 2002 Antarctic ozone hole. *Geophys. Res. Lett.,* **30,** 1394, doi:10.1029/2003GL016899.

Leue, C., M. Wenig, T. Wagner, U. Platt, and B. Jähne, 2001: Quantitative analysis of NO_x emissions from GOME satellite image sequences. *J. Geophys. Res.,* **106,** 5493–5505.

Martin, R. V., and Coauthors, 2002: An improved retrieval of tropospheric nitrogen dioxide from GOME. *J. Geophys. Res.,* **107,** 4437, doi:10.1029/2001JD001027.

Nedoluha, G. E., R. M. Bevilacqua, M. D. Fromm, K. W. Hoppel, and D. R. Allen, 2003: POAM measurements of PSCs and water vapor in the 2002 Antarctic vortex. *Geophys. Res. Lett.,* **30,** 1796, doi:10.1029/2003GL017577.

Noxon, J. F., 1979: Stratospheric NO_2. 2. Global behaviour. *J. Geophys. Res.,* **84,** 5067–5076.

Pawson, S., B. Naujokat, and K. Labitzke, 1995: On the polar stratospheric cloud formation potential of the northern stratosphere. *J. Geophys. Res.,* **100,** 23 215–23 225.

Platt, U., 1994: Differential optical absorption spectroscopy (DOAS). *Air Monitoring by Spectroscopic Techniques*, M. W. Sigrist, Ed., Chemical Analysis Series, Vol. 127, Wiley, 27–84.

Richter, A., and J. P. Burrows, 2002: Retrieval of tropospheric NO_2 from GOME measurements. *Adv. Space Res.,* **29,** 1673–1683.

——, F. Wittrock, A. Ladstätter-Weißenmayer, and J. P. Burrows, 2002: GOME measurements of stratospheric and tropospheric BrO. *Adv. Space Res.,* **29,** 1667–1672.

Roscoe, H. K., J. D. Shanklin, and S. R. Colwell, 2005: Has the Antarctic vortex split before 2002? *J. Atmos. Sci.,* **62,** 581–588.

Schiller, C., and A. Wahner, 1996: Comment on 'Stratospheric OClO measurements as a poor quantitative indicator of chlorine activation' by J. Sessler, M. P. Chipperfield, J. A. Pyle and R. Toumi. *Geophys. Res. Lett.,* **23,** 1053–1054.

Senne, T., J. Stutz, and U. Platt, 1996: Measurement of the latitudinal distribution of NO_2 column density and layer height in Oct/Nov 1993. *Geophys. Res. Lett.,* **23,** 805–808.

Sessler, J., M. P. Chipperfield, J. A. Pyle, and R. Toumi, 1995: Stratospheric OClO measurements as a poor quantitative indicator of chlorine activation. *Geophys. Res. Lett.,* **22,** 687–690.

Sinnhuber, B.-M., M. Weber, A. Amankwah, and J. P. Burrows, 2003: Total ozone during the unusual Antarctic winter of 2002. *Geophys. Res. Lett.,* **30,** 1580, doi:10.1029/2002GL016798.

Solomon, S., G. H. Mount, R. W. Sanders, and A. L. Schmeltekopf, 1987: Visible spectrospcopy at McMurdo Station, Antarctica, 2, Observations of OC1O. *J. Geophys. Res.,* **92,** 8329–8338.

Tørnkvist, K. K., D. W. Arlander, and B.-M. Sinnhuber, 2002: Ground-based UV measurements of BrO and OClO over Ny-Ålesund during winter 1996 and 1997 and Andøya during winter 1998/99. *J. Atmos. Chem.,* **43,** 75–106.

Toumi, R., 1994: Reaction of ClO with NO_3: OClO formation and night-time O_3 loss. *Geophys. Res. Lett.,* **21,** 1487–1490.

von Savigny, C., and Coauthors, 2005: The ozone hole breakup in September 2002 as seen by SCIAMACHY on *ENVISAT. J. Atmos. Sci.,* **62,** 721–734.

Wagner, T., C. Leue, K. Pfeilsticker, and U. Platt, 2001: Monitoring of the stratospheric chlorine activation by Global Ozone Monitoring Experiment (GOME) OClO measurements in the austral and boreal winters 1995 through 1999. *J. Geophys. Res.,* **106,** 4971–4986.

——, F. Wittrock, A. Richter, M. Wenig, J. P. Burrows, and U. Platt, 2002: Continuous monitoring of the high and persistent chlorine activation during the Arctic winter 1999/2000 by the GOME instrument on ERS-2. *J. Geophys. Res.,* **107,** 8276, doi:10.1029/2001JD000466.

Weber, M., S. Dhomse, F. Wittrock, A. Richter, B.-M. Sinnhuber, and J. P. Burrows, 2003: Dynamical control of NH and SH winter/spring total ozone from GOME observations in 1995–2002. *Geophys. Res. Lett.,* **30,** 1583, doi:10.1029/2002GL016799.

Wittrock, F., A. Richter, and J. P. Burrows, 1999: Validation of GOME BrO and OClO observations in the northern hemisphere. *Proc. of the European Symp. on Atmospheric Measurements from Space (ESAMS 99),* WPP-161, Noordwijk, Netherlands, European Space Agency, 735–738.