Continuous monitoring of the high and persistent chlorine activation during the Arctic winter 1999/2000 by the GOME instrument on ERS-2

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[1]   Measurements of OClO total column amounts during the cold Arctic winter 1999/2000 retrieved from observations by the satellite instrument Global Ozone Monitoring Experiment (GOME) are presented. OClO is formed as a minor product of the reaction of BrO + ClO and thus serves as an indicator for a stratospheric chlorine activation. As a result of the good spatial and temporal coverage of GOME, it is possible to follow the temporal development of the stratospheric chlorine activation during the winter and spring on a daily basis. An initial weak chlorine activation was observed during mid November, shortly after stratospheric temperatures were sufficiently low that formation of polar stratospheric clouds resulted. Strong chlorine activation started around 22 December, when PSC formation was possible over a large altitude range. Chlorine activation was significant and large until the beginning of March, peaking mid February. In the middle of March the chlorine activation steeply decreased and ended around 20 March, when the polar vortex broke up. The duration and the magnitude of the chlorine activation in the Arctic winter 1999/2000 were higher than during all previous Arctic winters since the launch of the GOME instrument in April 1995.   *INDEX TERMS:* 0340 Atmospheric Composition and Structure: Middle atmosphere-composition and chemistry; 0341 Atmospheric Compostion and Structure: Middle atmosphere-constituent transport and chemistry (3334); 0305 Atmospheric Compostion and Structure: Aerosols and particles (0345, 4801)   *KEYWORDS:* ozone hole, polar stratospheric ozone, satellite observations, GOME, chlorine activation, polar stratospheric clouds

1.   Introduction

[2]   *Solomon et al.* [1986] first proposed that activation of stratospheric inorganic chlorine, namely, the conversion of the ozone-inert reservoir species HCl and ClONO$_2$ into ozone-destroying chlorine oxides (ClO, Cl$_2$O$_2$), is responsible for the dramatic ozone destruction observed since the beginning of the 1980s over Antarctica during late winter and spring [*Farman et al.*, 1985, see also *Chubachi*, 1984] (reviews of the historical research progress are given by *Solomon* [1988,1990,1999]). In the cold stratosphere of the polar night, inorganic reservoir species are converted into activated chlorine species by heterogeneous reactions, which occur on polar stratospheric clouds (PSCs) formed at the low stratospheric temperatures prevailing during polar night [*Solomon et al.*, 1986; *Toon et al.*, 1986, 1990; *Carslaw et al.*, 1998a, 1998b; *Schreiner et al.*, 1999; *Voigt et al.*, 2000a, 2000b].

[3]   One commonly used remote sensing technique for the

monitoring of the stratospheric chlorine activation is the spectroscopic measurement of OClO [see, e.g., *Solomon et al.*, 1987a; *Fiedler et al.*, 1993; *Pommereau and Piquard*, 1994; *Renard et al.*, 1997; *Otten et al.*, 1998; *Frieβ et al.*, 1998; *Wagner et al.*, 1999; *Hild et al.*, 1999; *Wittrock et al.*, 1999; *Miller et al.*, 1999; *Hendrick et al.*, 2000]. The most important stratospheric source of OClO is the reaction of BrO and ClO. In the sunlit stratosphere, inorganic bromine primarily resides in BrO. BrO concentrations vary only slightly, and the availability of ClO often limits the formation of OClO. It has been shown by *Schiller and Wahner* [1996] that OClO (at SZA < 92°) can serve as an indicator for the amount of stratospheric ClO, i.e., the chlorine activation [see also *Solomon et al.*, 1987a].

[4]   In this study we present OClO data derived from spectra measured by the Global Ozone Monitoring Experiment (GOME) on board the European research satellite ERS-2, which was launched in April 1995. These data allow to monitor the stratospheric chlorine activation on a daily basis. This work complements the OClO data sets of earlier Arctic and Antarctic winters presented in a previous study [*Wagner et al.*, 2001].

2.   Instrument and Data Analysis

[5]   In this section we give a short overview over the instrument and data analysis; more detailed description is found elsewhere [*Wagner et al.*, 2001; *Leue et al.*, 2001; *Richter and*

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Burrows, 2001]. The GOME instrument consists of a set of four spectrometers that simultaneously measure sunlight reflected from Earth's atmosphere and the ground in four spectral windows covering the wavelength range between 240 and 790 nm with moderate spectral resolutions [*European Space Agency*, 1995; *Burrows et al.*, 1999, and references therein]. From the raw spectra monitored by GOME, the slant column density (SCD, the integrated trace gas concentration along the light path) is determined using differential optical absorption spectroscopy (DOAS) [*Platt*, 1994]. In brief, the measured spectra are modeled with a nonlinear fitting routine [*Stutz and Platt*, 1996] that suitably weights the absorption spectra of atmospheric trace gases (for the GOME OClO analysis the spectra for OClO, NO_2, and O_4 were included) and a solar background spectrum. To correct the "filling-in" of the solar Fraunhofer lines [*Grainger and Ring*, 1962] also a Ring spectrum is included in the analysis [*Bussemer*, 1993]. For the spectral analysis of OClO the wavelength range from 363 to 393 nm was used. From the inferred absorption, and the knowledge of the differential (narrow band) absorption cross section [*Wahner et al.*, 1987], the OClO SCD is calculated. The precision of the GOME OClO SCD measurements is estimated to be about ± 10% for large OClO SCDs (or to smaller than 4×10^{13} molecules cm^{-2}); the detection limit being about 5×10^{13} molecules/ cm^2 per spectra for this analysis [*Wagner*, 1999]. These uncertainties are mainly caused by an imperfect correction of the Ring effect, by uncertainties in the wavelength calibration of the GOME spectra and the included cross sections, as well as by spectral interferences between different cross sections [*Wagner*, 1999]. Additional systematic errors (≤8%) might arise from errors in the absolute calibration of the OClO cross section [*Wahner et al.*, 1987]. The GOME spectra were analyzed with respect to NO_2 in a similar way; details of this analysis can be found in *Wagner* [1999] and *Richter and Burrows* [2001].

[6] The light reaching the instrument is either reflected from the Earth's surface or scattered back from the atmosphere. Thus the measured stratospheric OClO SCD depends strongly on SZA. This has to be taken into account for the interpretation of daily maps of GOME OClO observations, in particular since the local SZA varies during one GOME orbit. For example, GOME observations at higher latitudes take place at larger SZA and thus show larger OClO SCDs compared to those at lower latitudes. The dependence of the OClO SCD from SZA is further increased by the photolysis of OClO, which results in OClO concentrations being strongly enhanced at large SZA. As a consequence, daily maps of GOME OClO SCDs are an excellent qualitative indicator of chlorine activation. For a quantitative interpretation with respect to stratospheric chlorine activation, however, the OClO SCDs measured at a fixed SZA are compared. In this study the daily maximum OClO SCDs measured at 90° SZA are determined [see also *Wagner et al.*, 2001]. From the OClO SCD also the vertically integrated concentration (vertical column density, VCD) can be determined by diving by the so-called air mass factor (AMF), which is derived from the modeling of the atmospheric radiative transport [*Solomon et al.*, 1987b; *Marquard et al.*, 2000]. For an assumed OClO concentration maximum at an altitude of about 18 km an AMF of about 11 can be applied (for SZA = 90°) [*Wagner*, 1999]. However, because of the strong photochemically induced change of the OClO concentration along the absorption path, in this study we only present the SCDs of OClO.

[7] It should be noted that for a quantitative interpretation of the OClO SCDs (at SZA = 90°) several factors have to be taken into consideration. These include the temperature dependence of the OClO formation reaction and of the thermal decomposition of Cl_2O_2, the dependence of the AMF on the OClO profile, and the dependence of the OClO formation on the BrO concentration. As discussed by *Wagner et al.* [2001], only the temperature dependence of the chemical reactions is expected to cause systematically higher OClO values at higher temperatures (for a given chlorine activation, i.e., for a given amount of ClO + $2Cl_2O_2$). While the formation reaction of OClO gets faster for decreasing temperatures, the thermal decomposition of Cl_2O_2 gets slower for decreasing temperature. Overall, we expect that these temperature dependencies cause slightly increasing OClO concentrations for decreasing temperatures. In particular, about 10% of the interhemispheric differences of the OClO SCDs measured by GOME in both hemispheres can be attributed to the different stratospheric temperatures during polar winter [*Wagner et al.*, 2001].

[8] Also, NO_2 concentration changes significantly along the absorption path, especially for SZA around 90°. However, the dependence of the AMF on the NO_2 profile is generally smaller than for OClO, since the concentration maximum of NO_2 is located at higher altitudes. In addition, DOAS measurements of NO_2 are conventionally expressed as VCDs. Thus we present the GOME NO_2 observations as VCDs in this study [*Leue et al.*, 2001; *Richter and Burrows*, 2001]. It is important to note that during the course of the year the locations (latitudes) of the GOME overpass at SZA = 90° change systematically (see Figure 1).

3. Results

[9] In Figure 1 a number of important results are presented. The evolution of the daily maximum OClO SCD at 90° SZA and the altitude range where PSC existence (T_{NAT}) was possible [*European Center for Medium-Range Weather Forecasts (ECMWF)*, 2000] as well as the altitude range where PSCs were actually measured, are shown [see also *Manney and Sabutis*, 2000]. PSC data observed by POAM (taken from http://opt.nrl.navy.mil/solve/solve_data.html [see also *Fromm et al.*, 1999; *Bevilacqua et al.*, 2000; *Fromm et al.*, 2000]) are also shown. They are in good agreement with several other PSC observations [see, e.g., *Blum et al.*, 2000; *Browell et al.*, 2000; *Kivi et al.*, 2000; *Voigt et al.*, 2000c]. In order to quantify the denoxification of the stratosphere, GOME observations of NO_2 have been used [see also *Leue et al.*, 2001; *Richter and Burrows*, 2001]: the daily minimum NO_2 VCDs at a SZA of 90° being plotted in Figure 1. Please note that the minimum NO_2 columns do not necessarily occur at exact the same location as the maximum OClO columns. Nevertheless, enhanced OClO columns were always observed over areas where also low NO_2 columns occurred. It should be also noted that the interpretation of total column measurements is usually not straight forward. For example, changes of the NO_2 and OClO concentrations can occur at different altitudes and are not necessarily related to each other. Concerning the potential influence of tropospheric NO_2 on the GOME observations [*Leue et al.*, 2001; *Richter and Burrows*, 2001] this effect can be neglected for the large SZA (90°) considered here.

[10] Enhanced values of OClO are closely related to the evolution of stratospheric temperatures, PSC appearance and stratospheric NO_2 columns [*Manney and Sabutis*, 2000]. Six
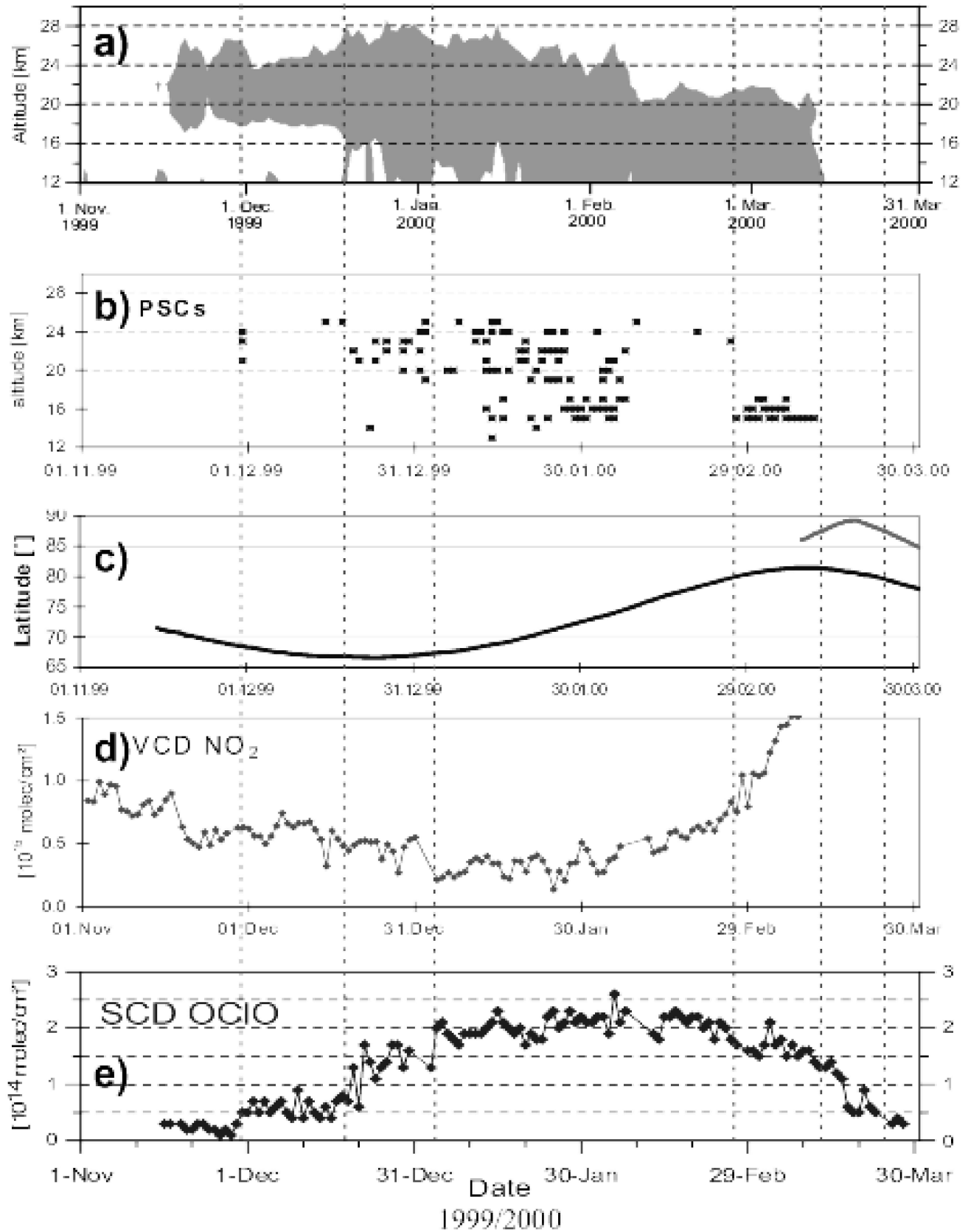
Figure 1.   Time series during the winter of 1999/2000 of (a) the stratospheric altitudes where PSC existence was possible, (b) PSC sightings by POAM, (c) the latitude of the GOME observations for SZA = 90°, (d) minimum GOME NO$_2$ VCDs measured at 90° SZA, and (d) maximum GOME OClO SCDs measured at 90° SZA. After 10 March GOME observations for SZA = 90° are possible at two latitude bands.

periods can be characterised and are discussed below. For each period, selected maps of GOME OClO together with the stratospheric temperature and potential vorticity are presented [*European Center for Medium-Range Weather Forecasts (ECMWF)*, 2000].

3.1. From Mid November Until 29 November 1999: First Appearance of Enhanced OClO SCDs

[11] During this period the stratospheric temperatures strongly decreased, and around 15 November they reached values which allowed the existence of PSCs (T_{NAT}) for an altitude range between about 17 and 25 km. After the middle of November, the minimum of the total NO_2 VCD measured by GOME over the Arctic fell to values around 5×10^{14} molecules cm^{-2} indicating a strong denoxification of the stratosphere. On 19 November the first evidence for chlorine activation was found in the GOME OClO data (see Figure 2a). Although from POAM (http://opt.nrl.navy.mil/solve/solve_data.html) and balloon-borne measurements [*Kivi et al.*, 2000] the first PSC sightings were reported on 28 November 1999, the occurrence of enhanced OClO data as well as the evolution of stratospheric temperature and NO_2 VCD indicate that their first appearance was probably well before 19 November. In Figure 2a it can be seen that enhanced OClO SCDs on 19 November appeared over regions which clearly lie outside the polar vortex (between Greenland and Scandinavia). However, they coincide with the area of the minimum stratospheric temperature ($<T_{NAT}$ at the 475 K level). It should be noted that during November the OClO SCDs are still relatively small compared to those found in later periods. In particular, at 90° SZA (corresponding to a latitude of about 70°N) the maximum OClO SCDs during that period are still below the detection limit (see Figure 1). Nevertheless, for several cases OClO SCDs significantly above the detection limit were found for larger SZA (at higher latitudes) and even for smaller latitudes (19 November; see Figure 2a).

[12] It should be noted that during the polar night a large central part of the Arctic stratosphere can not be observed by GOME; thus especially early events of chlorine activation with possibly restricted spatial extension might be overlooked by our observations.

3.2. From 30 November Until 21 December: First Period of Continuously Enhanced OClO SCDs

[13] During this period the altitude range where temperatures were low enough for PSC existence was similar to the previous period (Figure 1). PSCs were actually measured between about 20 and 25 km altitude, e.g., by POAM. Although during this period the minimum stratospheric NO_2 VCDs were somewhat higher than at the end of November, slightly enhanced OClO SCDs were present. Figure 2a shows a map of GOME OClO for November 30 when activated air masses were found over northern Europe.

3.3 From 22 December Until 1 January: First Period of Strong Chlorine Activation

[14] Around December 22 the stratospheric temperatures again strongly decreased leading to PSC formation over a wider altitude range (from about 15 to 28 km) compared to the previous periods. Indeed, enhanced PSC loading was actually observed by POAM during this period (Figure 1). The stratospheric NO_2 VCDs were again as low as the end of November (around 5×10^{14} molecules cm^{-2}). Correspondingly, the

OClO SCDs steeply increased after 22 December. In Figure 2a it can be seen (for 22 December) that these high OClO SCDs were present over an extended area of cold air. This region essentially coincided with the location of the polar vortex. Its center was located between the North Pole and Scandinavia. It should be noted that enhanced OClO SCDs were also observed over northeastern Siberia, where a slightly deformed part of the polar vortex was situated. Stratospheric temperatures over that region were far too high for PSC formation, which indicates that activated air masses can be rapidly transported within the polar vortex.

3.4. From 2 January Until 26 February: Period of the Strongest Chlorine Activation

[15] During this period, very high OClO SCDs were continuously observed by GOME. These high values occurred during a period where low temperatures over an extended altitude range (about 12 to 25 km) favored PSCs formation as demonstrated by actual PSCs detections (Figure 1). During the first half of the period, PSCs were nearly continuously detected between about 13 and 24 km. At this time the areas having small NO_2 VCDs (with minimum values down to below 2×10^{14} molecules cm^{-2}) became very large. During the whole period the OClO SCDs stayed high (around 2×10^{14} molecules cm^{-2}) with the maximum values (2.6×10^{14} molecules cm^{-2}) occuring on February 5. During most of this period the polar vortex was stable and coincided well with the area of minimum temperatures (see Figure 2, where maps for 7 and 22 January and for 5 February are shown).

3.5. From 27 February Until 13 March: Last Period During Which PSCs Were Observed

[16] After 28 February, PSCs were only detected at low altitudes (<17 km). At higher altitudes the stratospheric temperatures increased to values above the PSC formation temperature. During this period also the stratospheric NO_2 VCD strongly increased (see Figure 1); after 28 February the minimum values were above 1×10^{15} molecules cm^{-2}, and after 8 March they were above 1.5×10^{15} molecules cm^{-2}. However, while NO_x recovery took place at altitudes above about 20 km (leading to the relatively high NO_2 VCDs measured by GOME), low NO_2 concentrations were still observed at altitudes below according to balloon-borne observations of NO_2 [*THESEO*, 2000]. The relatively high OClO SCDs ($\approx 1.6 \times 10^{14}$ molecules cm^{-2}) very probably belong to this altitude range.

3.6. From 14 March Until 24 March: Strong Decline of Stratospheric Chlorine Activation

[17] After 13 March, stratospheric temperatures were too warm for PSC formation, and PSCs were no longer detected by POAM. Approximately 5 days after the final PSC was observed, the OClO SCDs dropped steeply to values around the detection limit (about 0.5×10^{14} molecules cm^{-2}) indicating a very rapid deactivation of the remaining activated air masses (at altitudes below about 20 km). This rapid deactivation is in particular due to the NO_x recovery at altitudes below about 20 km which took place during early March (see Figure 2a, where maps for 15 and 19 March are shown). After 13 March the NO_2 VCD exceeded 2×10^{15} molecules cm^{-2}) throughout the Arctic, a value significantly higher than during previous years. During the winter 1996/1997, when stratospheric chlorine activation continued into March, the NO_2 VCDs were still
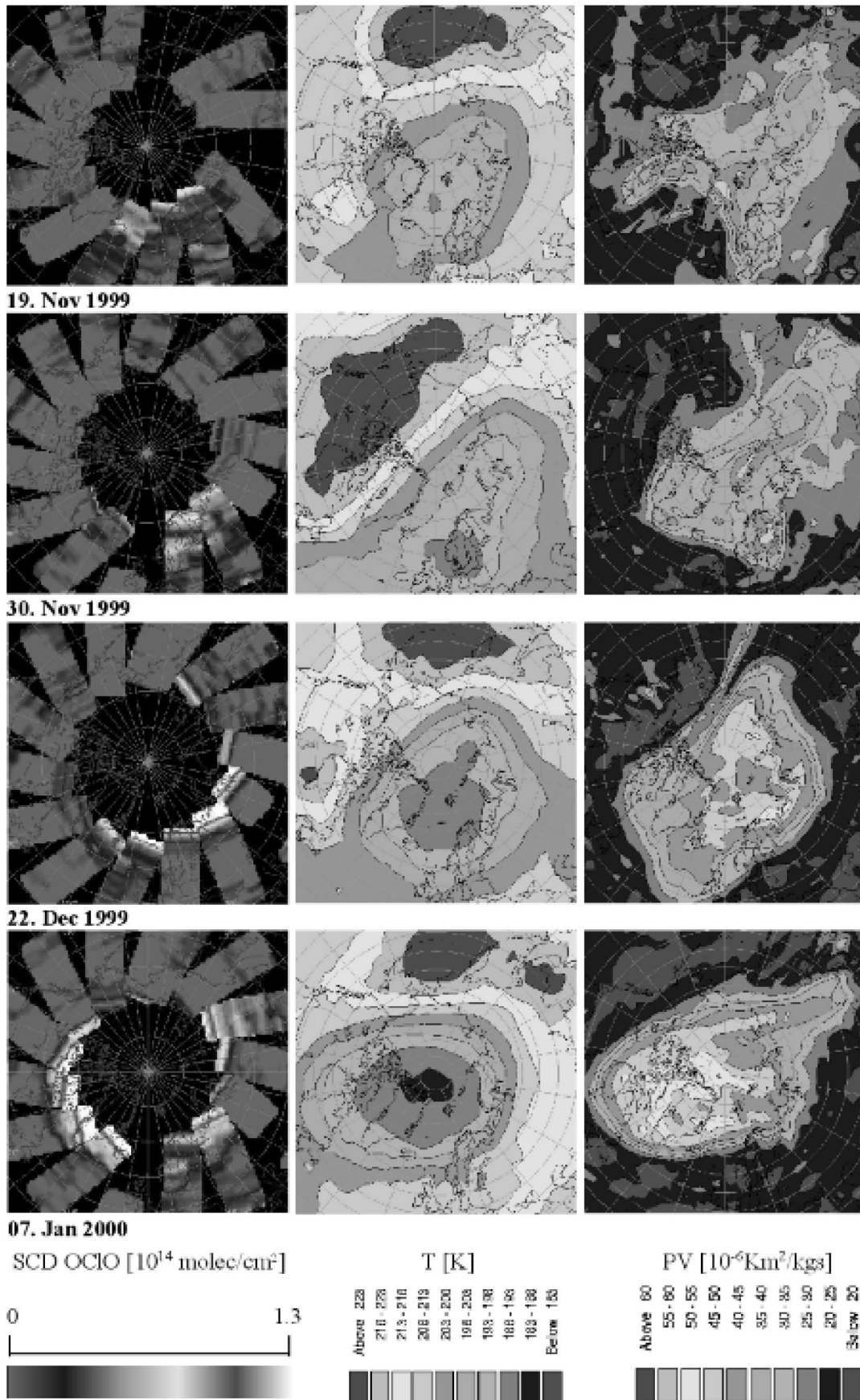
Figure 2. Daily maps of GOME OClO, stratospheric temperatures, and potential vorticity at the 475 K level (from ECMWF analysis) for selected days during the Arctic winter 1999/2000: (a) 19 and 31 November, 22 December, and 7 January. (b) January 22, 5 February, and 15 and 19 March.
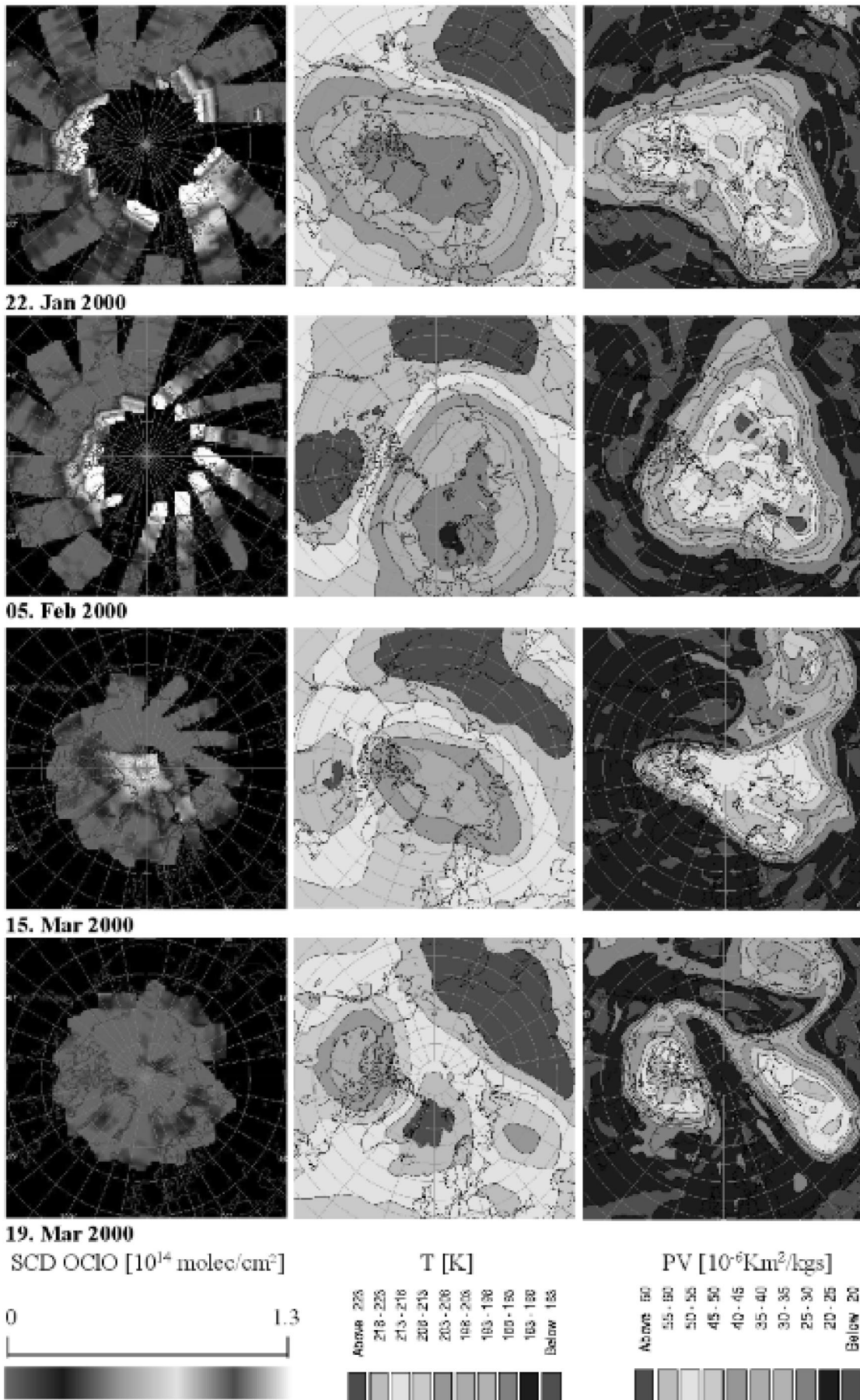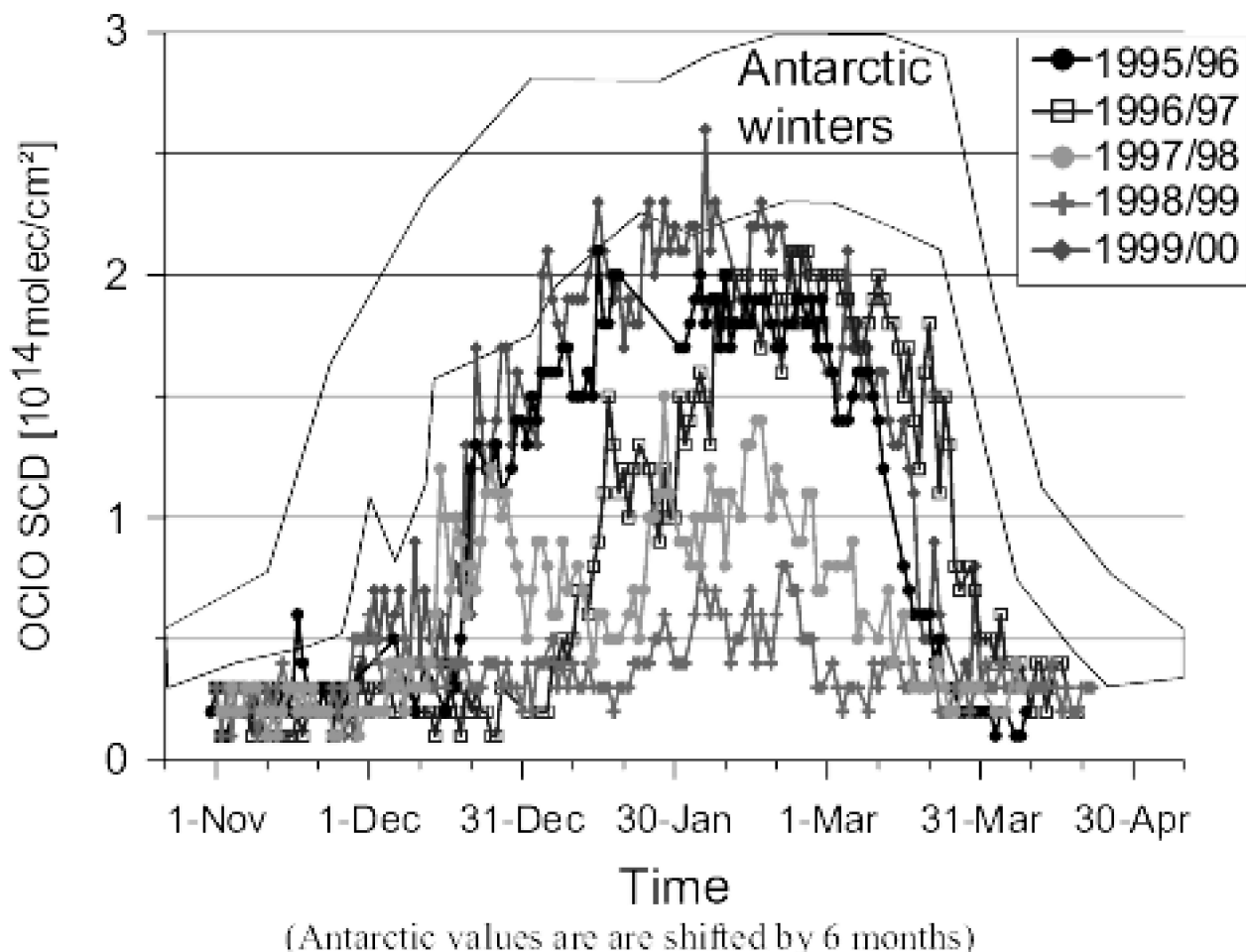
22. Jan 2000

05. Feb 2000

15. Mar 2000

19. Mar 2000

SCD OClO [10^{14} molec/cm²]

0   1.3

T [K]

Above 225
218 - 225
213 - 218
208 - 213
203 - 208
198 - 203
193 - 198
188 - 193
183 - 188
Below 183

PV [10^{-6}Km²/kgs]

Above 60
55 - 60
50 - 55
45 - 50
40 - 45
35 - 40
30 - 35
25 - 30
20 - 25
Below 20

Figure 2. (continued)

Comparison of the OClO SCDs in Arctic and Antarctic winters
(daily maximum OClO-SCD at 90° SZA)

Figure 3. Comparison of the daily maximum OClO SCD (at 90° SZA), measured over the Arctic and the Antarctic in the winters 1995–2000. For comparison of both data sets, the *x* axis are shifted against each other by 6 months.

around 10^{15} molecules cm^{-2} in mid March. In that winter the chlorine activation lasted until about 3 weeks after the last PSCs had appeared [*Wagner et al.*, 2001]. It should be noted that some remaining air masses having chlorine activated air might not be detected by GOME at this time of year. This is because of the limited areas observed having large SZAs during this part of the year. Nevertheless, after the small peak on 21 March, no significant indication of chlorine activation was identified in the GOME OClO data. The rapid decline of the chlorine activation during the middle of March (see Figure 2b) was probably also favored by the split up of the polar vortex and the mixing of vortex air with air from outside (see Figure 2b). In this way, even air masses for which denitrification due to sedimentation of PSCs had occurred [*Santee et al.*, 2000] can be enriched with NO$_y$ again.

[18] In Figure 3 the OClO SCDs of the Arctic winter 1999/2000 are compared to those of previous Arctic and Antarctic winters (1995-1999 [see *Wagner et al.*, 2001]). While the OClO

SCDs of the winter 1999/2000 are still systematically smaller than those generally observed in the Antarctic winters, they are, however, the strongest observed so far by GOME during Arctic winters. Furthermore, the duration of strongly enhanced OClO SCDs (about 3 months) is also longer than in previous Arctic winters. In conclusion, the Arctic winter 1999/2000 shows the strongest stratospheric chlorine activation since observation began with GOME after the launch of ERS-2 in April 1995.

[19] The GOME OClO observations are qualitatively consistent with several other observations of the stratospheric halogen activation made during the SOLVE/THESEO campaign [see, e.g., *Hendrick et al.*, 2000; *Stimpfle et al.*, 2000; *Stroh et al.*, 2000; *Toohey et al.*, 2000; *Von König et al.*, 2000; *Wilmouth et al.*, 2000; *F. Wittrock et al.*, manuscript in preparation, 2001]. A detailed comparison of GOME observations with modeling studies for the Arctic winter 1999/2000 will be presented by *F. Wittrock et al.* (manuscript in preparation, 2001).

4. Conclusions

[20] Retrievals of OClO amounts from the GOME measurements have enabled the stratospheric chlorine activation to be studied on a daily basis during the Arctic winter 1999/2000. These data allow us to characterise the evolution of stratospheric chemistry with respect to ozone destruction. It was possible to investigate the different phases of chlorine activation and deactivation and to relate them to the stratospheric temperatures, PSC appearance, denoxification, and renoxification. Compared to previous Arctic winters since the launch of ERS-2 (April 1995), the winter 1999/2000 showed the strongest and most persistent chlorine activation. Continuously high levels of OClO were observed over 3 months, from mid December to mid March. The first sporadic events of chlorine activation appeared in mid November, after the stratospheric temperatures dropped below the PSC formation temperature. During that period, also small NO_2 VCDs (around 5×10^{14} molecules cm^{-2}) indicated the beginning of a strong stratospheric denoxification. During the period with the highest OClO SCDs, PSCs were present over a large altitude range (between about 13 and 24 km) indicating that chlorine activation took part over a large altitude range. The large vertical extension of activated air masses is the most probable reason for the higher OClO SCDs compared to previous Arctic winters [see, e.g., *Von der Gathen et al.*, 2000]. However, during the later part of the winter (late February to early March), PSCs were only present within a smaller altitude range (Figure 1 [see also *Manney and Sabutis*, 2000]).

[21] Shortly after the last PSC sighting (by POAM) on 13 March the stratospheric chlorine activation sharply decreased; the OClO SCDs dropped to levels around or below the detection limit on 20 March. This rapid conversion into the reservoir species was much faster than in previous winters (since 1995) [*Wagner et al.*, 2001] when the drop of the GOME OClO SCDs down to the detection limit occurred typically about 2 to 3 weeks after the stratospheric temperatures increased above the PSC formation threshold. The most likely explanation for this sudden ending of the chlorine activation during the winter 1999/2000 is that the conversion of ClO into the reservoir species $ClONO_2$ is favored by the relatively high values of NO_2 ($\approx 2 \times 10^{15}$ molecules cm^{-2}) measured by GOME during that period. NO_x-rich air masses could have transported from outside the vortex after 15 March, when the polar vortex split up into separate pieces. In comparison, during the winter 1996/1996 the NO_2 VCDs were around 10^{15} molecules cm^{-2} during mid March, and the chlorine activation lasted until about 3 weeks after the latest PSC occurrence. The rapid decline during mid March is probably one important reason for the fact that the ozone destruction during the winter 1999/2000 was not substantially higher than those in previous Arctic winters in spite of the significantly stronger chlorine activation [*Goutail et al.*, 2000]. Another, probably even more important reason is that during the later period of chlorine activation (late February to early March) when photolytic ozone destruction is most effective, the chlorine-activated air masses were restricted to an altitude range below about 20 km [*THESEO*, 2000; *Manney and Sabutis*, 2000]. In contrast, during previous winters ozone destruction took place over a more extensive altitude range [*Goutail et al.*, 2000].

[22] Although the OClO SCDs during the Arctic winter 1999/2000 are higher than those during previous Arctic winters, they still are systematically smaller (by about 25%) compared to those appearing during the Antarctic winters. This difference might be only partly be explained by the temperature dependence of the OClO formation reaction [*Wagner et al.*, 2001]. Since in both hemispheres similar maximum values of the stratospheric ClO concentration were measured by the MLS instrument [*Waters et al.*, 1993a, 1993b; *Santee et al.*, 1995, 1996, 1997], the difference in the column is most probably caused by a larger altitude range of activated air masses in the Antarctic [*Wagner et al.*, 2001].

[23] Also the duration of the strong chlorine activation in 1999/2000 (about 3 months) was longer than those of previous Arctic winters but was still shorter than in the Antarctic (where it typically lasts about 4 months). The strong chlorine activation during the Arctic winter 1999/2000 as measured by GOME is in good agreement with several other studies related to stratospheric chlorine activation.

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