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The northern hemisphere stratospheric vortex during the 2002–03 winter: Subsidence, chlorine activation and ozone loss observed by the Odin Sub-Millimetre Radiometer

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[1] We present profile measurements of key constituents relevant to stratospheric chemistry and dynamics such as ozone (O_3), nitrous oxide (N_2O), and chlorine monoxide (ClO) taken during the 2002–03 northern hemisphere winter by the Odin Sub-Millimetre Radiometer (SMR), a limb-sounding satellite sensor launched in February 2001. The observations of the chemically passive tracer N_2O show a subsidence of lower stratospheric air masses inside the Arctic vortex in the range of 3–5 km, or 60–100 K in terms of potential temperature, for the period November 2002 to March 2003. Activated chlorine in the form of ClO was observed inside the vortex from the beginning of December until mid-February. The accumulated chemical ozone loss over this period, derived from the correlation of ozone with N_2O, is estimated to be 28 ± 9% on the 50 ppbv level of N_2O, i.e., for lower stratospheric air masses subsiding from ~23 down to 19 km, the lower limit of the Odin/SMR ozone measurement in the 501.8 GHz band. *INDEX TERMS:* 0340 Atmospheric Composition and Structure: Middle atmosphere—composition and chemistry; 0341 Atmospheric Composition and Structure: Middle atmosphere—constituent transport and chemistry (3334); 0394 Atmospheric Composition and Structure: Instruments and techniques. **Citation:** Urban, J., et al. (2004), The northern hemisphere stratospheric vortex during the 2002–03 winter: Subsidence, chlorine activation and ozone loss observed by the Odin Sub-Millimetre Radiometer, *Geophys. Res. Lett.*, *31*, L07103, doi:10.1029/2003GL019089.

1. Introduction

[2] The Sub-Millimetre Radiometer (SMR) on board the Odin satellite, launched in February 2001 into a polar sun-synchronous orbit, employs 4 tunable single-sideband Schottky-diode heterodyne receivers in the 485–580 GHz spectral range. The receiver noise temperature is in the order of 3000 K. A 1.1 m telescope is used for passive observations of thermal emission originating from the Earth's limb. Spectra are recorded using two 800 MHz wide auto-correlator spectrometers with effective resolution of 2 MHz. Measurements are performed in a time sharing mode with astronomical observations [see, e.g., *Frisk et al.*, 2003; *Olberg et al.*, 2003].

[3] In aeronomy mode, various target bands are dedicated to observations of trace constituents relevant to stratospheric/mesospheric chemistry and dynamics such as O_3, ClO, N_2O, HNO_3, H_2O, CO, and isotopes of H_2O and O_3 [e.g., *Murtagh et al.*, 2002]. Profile information is retrieved from the spectral measurements of a limb-scan by inverting the radiative transfer equation for a non-scattering atmosphere. A retrieval algorithm based on the "Optimal Estimation Method" [*Rodgers*, 1976] has been adopted for the ground segment of Odin/SMR [*Merino et al.*, 2001; *Eriksson et al.*, 2002; *Baron et al.*, 2002; *Urban et al.*, 2004].

2. Stratospheric Mode Measurements

[4] In stratospheric mode, the Odin/SMR instrument scans the limb of the atmosphere between roughly 7 and 70 km. Thermal emission-lines of the chemically active radical chlorine monoxide (ClO) and the chemically inert long-lived tracer for transport processes nitrous oxide (N_2O) are measured along with a weak ozone (O_3) transition in a band centered at 501.8 GHz.

[5] We present early results obtained with the Bordeaux level 2 processing system CTSO (Chaîne de Traitement Scientifique Odin) employing the forward and inversion model MOLIERE-5 [*Urban et al.*, 2004] to retrieve profile information from the calibrated spectral measurements of the limb-scans (level 1b). Throughout this work we use CTSO-version 222 data for the 501.8 GHz band (ClO, N_2O, O_3). At polar latitudes, measured ClO profiles cover the altitude range 17 to 50 km with a vertical resolution of typically 2–2.5 km in the lower stratosphere and a corresponding single-scan retrieval precision of around 0.15 ppbv. For comparison, the single-profile precision for ClO (version 5) data measured by the Microwave Limb Sounder on the UARS satellite is about 0.3 ppbv in the lower stratosphere with corresponding altitude resolution of about 4 km [*Livesey et al.*, 2003]. N_2O is retrieved above

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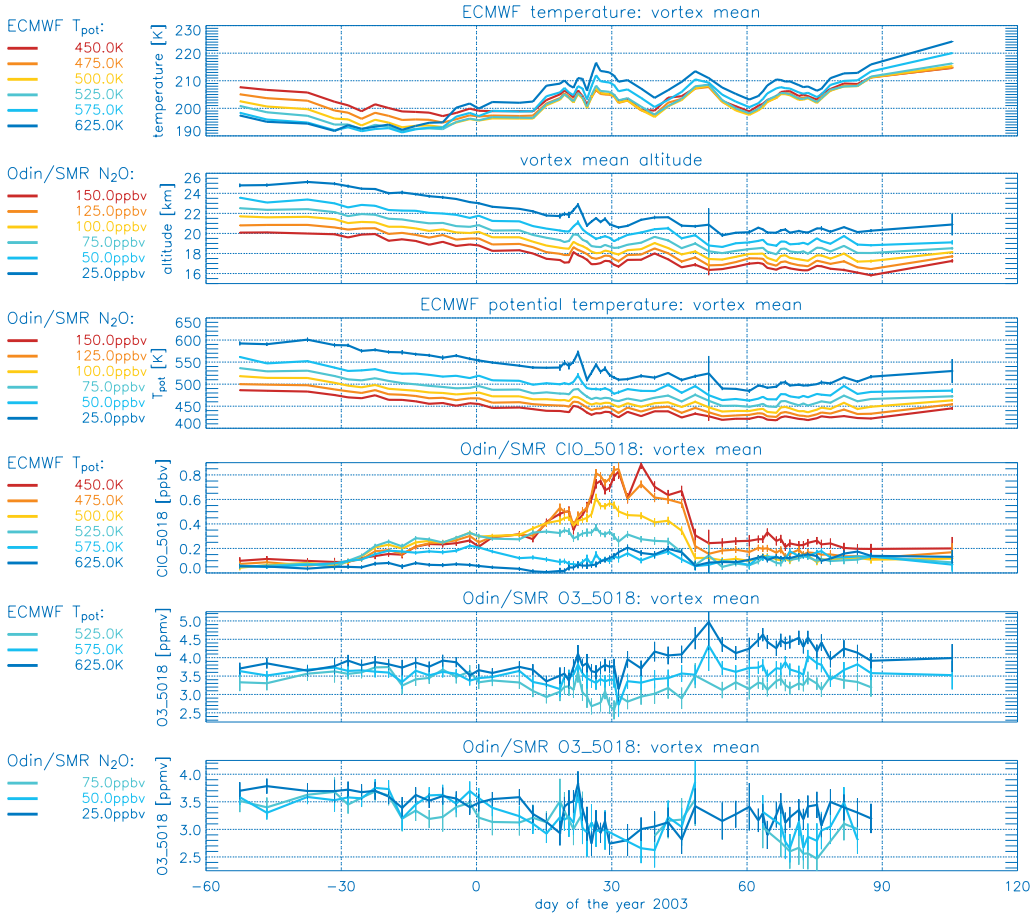

Figure 1. Evolution of the northern hemisphere vortex during winter 2002–03. Top: vortex mean temperature (ECMWF) for different levels of potential temperature representative of the lower stratosphere. Second and third plot: vortex mean altitude and corresponding ECMWF potential temperatures for selected Odin/SMR N_2O levels. 4th and 5th plot: vortex mean ClO and O_3 as observed by Odin/SMR for selected levels of potential temperature. Bottom: average chemical ozone loss inside the Arctic vortex during winter 2002–03 as observed by Odin/SMR for selected levels of N_2O.

14 km with a vertical resolution of 2 km and a precision in the order of 5% (10–20 ppbv). The measurement range for ozone in the 501.8 GHz band is ~20–50 km with a resolution of around 2.5 km and a single-scan precision in the order of 25% (0.5–1.5 ppmv). From the preliminary validation analysis, we estimate upper limits for the systematic errors of the measurements in the lower stratosphere to be about 0.2 ppbv for ClO, 20–40 ppbv for N_2O, and 1 ppmv for O_3 in the 501.8 GHz band.

[6] Note that the horizontal resolution of a limb measurement depends mainly on the altitude resolution: a resolution of 2 km corresponds to a limb path of ~320 km in the layer around the tangent point. Also note that due to the satellite motion of ~7 km/s and the time required for vertically scanning the limb of the atmosphere (~90 s), the retrieved vertical profile corresponds to a range of ±315 km around the average scan position along the satellite track.

3. Evolution of the Northern Hemisphere Stratosphere During Winter 2002–03

3.1. Methodology

[7] While altitude range and measurement accuracy are fixed quantities inherent to the measurement system, data averaging allows us to reduce further the statistical error due to measurement noise. For the analysis of the temporal evolution of the measured quantities we keep the best vertical resolution and calculate vortex mean profiles using all Odin/SMR measurements inside the vortex at selected observation days. We define the vortex edge by using the observations of nitrous oxide: limb-scans resulting in retrieved N_2O volume mixing ratios (VMR) smaller than 70 ppbv at the potential temperature surface of 550 K (and at higher levels), according to analyses from the European Centre for Medium-Range Weather Forecasts (ECMWF), are considered to be vortex measurements. Typically, we average between 30 to 60 measurements in the lower stratosphere resulting from about 15 orbits per day.

[8] The evolution of vortex averages of ECMWF temperature as well as Odin/SMR N_2O, ClO, and O_3 during winter 2002–2003 is presented in Figure 1. In addition, Figure 2 shows the geographical distribution of these quantities for selected days, interpolated on the 525 K potential temperature surface. Results are discussed in detail in the following sections.

3.2. Meteorological Situation

[9] According to meteorological analyses [e.g., *EORCU*, 2003], the 2002–03 Arctic winter is dominated by a very cold vortex during November and December. A minor

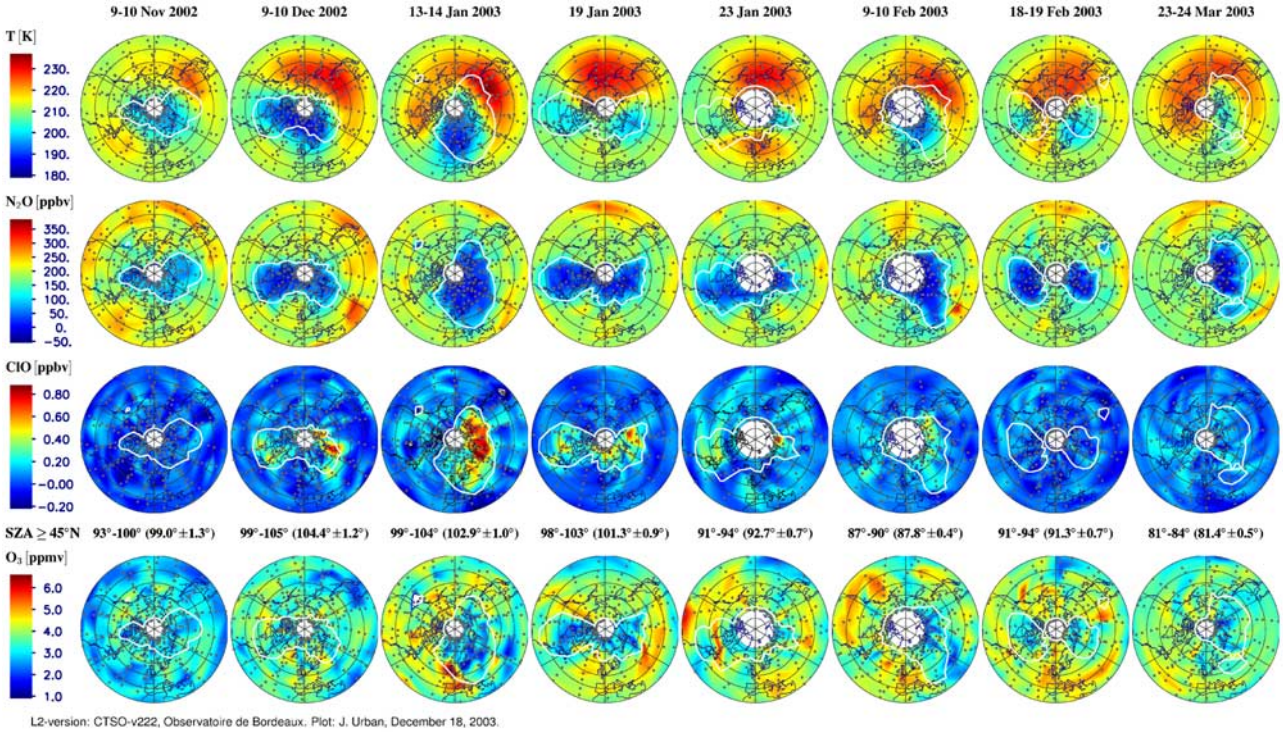

Figure 2. The northern hemisphere lower stratosphere during winter 2002–03 at the 525 K level of potential temperature. Top row: ECMWF temperature fields. Below: Odin/SMR measurements of N_2O (2nd row), ClO (third row) and O_3 (bottom row). The minimum latitude shown is 30°N. Average positions of the individual limb-scans are indicated (crosses). The white line is the 125 ppbv contour of N_2O which roughly indicates the edge of the vortex. The solar zenith angle at the time of the measurements is given below the ClO graphs, numbers in parentheses are the vortex mean value and its standard deviation.

warming occurred at the end of December and a major warming in mid-January, followed by two minor warmings in February and March. The final warming started at the end of March and the vortex disappeared by the end of April. Temperatures were below 192 K, the 30 hPa threshold for polar stratospheric cloud (PSC) formation, mainly from November 20, 2002 up to January 15, 2003 before the major warming, and then again for a short time in the middle of February. Calculations indicate consistently that for the middle of February and at the beginning of March PSC formation was possible only in the potential temperature range of 475 K and below [*EORCU*, 2003]. Figure 1 (top) shows the evolution of the mean temperature of the vortex (from ECMWF) during the winter. Temperatures were averaged corresponding to the Odin/SMR measurements, i.e., using ECMWF profiles interpolated on the average positions and times of the limb-scans, and are presented for selected levels of potential temperature representative of the lower stratosphere between 425 and 625 K. The geographical distribution of ECMWF temperatures on the 525 K surface of potential temperature is presented in Figure 2 (top row). The dynamical situation of the polar vortex is, in particular, reflected in the Odin/SMR measurement of the chemically inert and therefore long-lived species nitrous oxide (Figure 2, 2nd row), showing at this level low volume mixing ratios inside the vortex (<125 ppbv) due to the subsidence of vortex air and the negative vertical gradient of the N_2O mixing ratio profile. After the formation of the vortex in early November and a very cold and stable period until mid-January, the N_2O field shows a very elongated vortex during the strong warming period at the end of January with two less intense cold poles according to ECMWF temperatures (see plots for 19–23/1/2003). Colder temperatures are again established in the first half of February (see plot for 9–10/2/2003), but an unstable vortex, split into two parts, is observed during the February warming (see plot for February 18–19, 2003). Temperatures remain relatively high until the beginning of the final warming at the end of March, except for a short period at the beginning of March.

3.3. Subsidence

[10] Air inside the polar vortex is affected by diabatic cooling and subsidence during winter [e.g., *Müller et al.*, 1997; *Bremer et al.*, 2002]. We use Odin/SMR N_2O measurements to quantify this effect for the 2002–03 winter. Figure 1 (second and third row) shows the subsidence of lower stratospheric air masses, characterized by constant mixing ratios of N_2O, in terms of altitude and potential temperature. Vortex air characterized by an average N_2O volume mixing ratio of 25 ppbv (150 ppbv) sinks from an altitude of about 25 km (20 km) in November 2002 down to about 20–20.5 km (16–17 km) at the end of February/beginning of March 2003. In terms of potential temperature, we measure subsidence from 590–600 K (480–490 K) down to 490–500 K (420–430 K), for the same period and levels of N_2O. Some variability is observed in the period after mid-January, reflecting the unstable

vortex and the possibility of mixing with air masses from outside the vortex.

3.4. Chlorine Activation

[11] Odin/SMR measurements indicate enhanced vortex-averaged values of lower stratospheric ClO, characteristic of ongoing chemical ozone destruction, starting at the beginning of December up to mid-February (Figure 1, 4th row). The maximum of activation is found in the 450–475 K range of potential temperature during the first half of February, somewhat lower values of activation are observed at the 500–525 K levels. Absolute quantities have to be interpreted with care due to the strong diurnal variation of ClO, since Odin's terminator orbit with the ascending node at 18:00 hours leads to observations at high solar zenith angles (>90°) in the winter hemisphere, i.e., during most of the observation period discussed here. A detailed analysis of chlorine activation using Odin/SMR ClO measurements and 3-d model data will therefore be the subject of a separate publication (P. Ricaud et al., manuscript in preparation). The geographical distribution of chlorine activation on the 525 K surface of potential temperature is shown in Figure 2 (third row). The area of chlorine activation is well confined to the vortex limits (N_2O @ 525 K < 125 ppbv) and to the area of low temperatures up to the middle of February. Afterwards, only insignificant chlorine activation is observed at this level, keeping the SMR single-profile error due to statistical measurement noise of ~0.15 ppbv for ClO in mind. Vortex-averaged values of ClO at 475 K and particularly at 450 K (Figure 1) indicate slightly enhanced ClO up to the middle of March compared to the November value, with values slightly above our rather conservative upper limit of 0.2 ppbv for the systematic error of the ClO measurement.

3.5. Ozone Loss

[12] Odin/SMR vortex average ozone derived from the 501.8 GHz band is presented in Figure 1 (5th row) for selected levels of potential temperature. Note that the lower limit of Odin/SMR ozone measurements from this band is usually in the order of 500–525 K. We observe the strongest depletion of ozone at the lowest levels measured, i.e., at the 525 K surface of potential temperature. From the average value at the end of November of ~3.7 ± 0.2 ppmv, ozone is reduced by 27 ± 7% to 2.7 ± 0.2 ppmv at the end of January. In February, ozone values at this level increase up to 3.5 ppmv, which can mainly be attributed to mixing with ozone rich air from outside the vortex, as no strong subsidence is observed during that period. At higher levels (575–625 K), the same behavior is observed, but ozone losses in January are less pronounced. Note that an anti-correlation of ozone and chlorine monoxide vortex averages is observed for the 525 K level, in particular visible during the January warming event (e.g., January 23, 2003) and in February. The geographical distribution of ozone on the 525 K surface is shown in Figure 2 (bottom row). Low values of ozone, obviously correlated with the position of the vortex and the activation of chlorine, can be observed in January 2003. For the other periods, losses are within the single-scan error due to measurement noise of 0.4–0.6 ppmv at this level.

[13] The correlation with N_2O can be used to remove the effect of subsidence which masks the chemical losses in the lower stratosphere of the Arctic vortex. For a given air parcel which is transported vertically or horizontally through the lower stratosphere, the ratio of the usually long-lived species N_2O and O_3 to each other should be constant, unless chemical conversion or irreversible mixing occurs. This technique has been widely used in the past to identify small chemical losses from a large variability due to transport processes [e.g., *Proffit et al.*, 1990; *Plumb and Ko*, 1992; *Müller et al.*, 1997; *Bremer et al.*, 2002]. Figure 1 (bottom) shows the vortex-averaged ozone mixing ratios corresponding to selected N_2O values (25, 50, and 75 ppbv). On the 25 ppbv level of N_2O, ozone is depleted by 27 ± 7% by the end of January (~2.7 ± 0.2 ppmv) with respect to its average mixing ratio at the end of November of 3.7 ± 0.2 ppmv. Air masses on this level subsided from about 25 km (or 600 K) down to about 21 km (~525 K) during this period (see Figure 1, second row). Accumulated losses of 28 ± 9% are observed at the 50 ppbv level of N_2O (subsiding from ~23.5 to ~19.5 km), where a minimum ozone value of 2.6 ± 0.3 ppmv is observed in mid-February compared to its average value at the end of November of about 3.6 ± 0.2 ppmv. Ozone values for mid-March indicate slightly larger accumulated chemical losses at the 75 ppbv level of N_2O, but data are too sparse to study the temporal evolution of ozone after mid-February. At the end of the winter data for this correlation analysis are quite limited, since ozone is retrieved above 500–525 K only, whilst N_2O values at this lower limit start being very small (<25 ppbv) by the end of January due to continued subsidence (Figure 1, third row). An improved correlation analysis will be possible in the future when Odin/SMR ozone retrievals from the simultaneously measured 544.6 GHz band, which extend to lower altitudes, become available. Data retrieved from this line were not used in this study because of an uncertainty in the pressure broadening parameter. Ozone measurements of the Odin/OSIRIS (Optical Spectrograph and InfraRed Imaging System) instrument, available under sunlit conditions only, might also be used at the end of the winter season. Nevertheless, our results appear to be in reasonable agreement with preliminary results based on other observation techniques reported so far for the same altitude range [e.g., *EORCU*, 2003].

4. Summary and Conclusions

[14] We present Odin/SMR observations of key constituents for chemistry and dynamics in the lower stratosphere of the Arctic polar vortex during the 2002–03 northern hemisphere winter. Diabatic cooling leads to subsidence of lower stratospheric air masses inside the vortex of 3–5 km, or 60–100 K in terms of potential temperature, during the period mid-November 2002 to the beginning of March 2003, derived from N_2O measurements within the range 25–150 ppbv. Enhanced activated chlorine is observed during the particularly cold early phase of the winter starting at the beginning of December and up to mid-February, with slightly enhanced values persisting until mid-March. Maximum accumulated ozone losses of 27 ± 7% are observed at the end of January on the 525 K potential temperature surface, the lower limit of ozone measurements for this band. A correlation analysis of O_3 versus N_2O yields chemical ozone loss of similar amounts for the period from the end of November 2002 up to the first half of February 2003. Future analyses will also make use of O_3 and HNO_3 measurements

from the 544.6 GHz band in order to study ozone losses at lower altitudes as well as denitrification.

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