Ozone depletion observed by the Airborne Submillimeter Radiometer (ASUR) during the Arctic winter 1999/2000

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Received 15 February 2001; revised 6 August 2001; accepted 6 August 2001; published 21 September 2002.

[1] In the winter 1999/2000 the Airborne Submillimeter Radiometer (ASUR) participated in the Stratospheric Aerosol and Gas Experiment III Ozone Loss and Validation Experiment/Third European Stratospheric Experiment on Ozone project on board the NASA research aircraft DC-8. During three deployments in early December 1999, late January, and early March 2000, the ASUR instrument took various measurements of ozone and key species related to stratospheric ozone chemistry. After the sunlight reached the vortex region in January 2000 peak values of about 1.8 ppb ClO were measured by ASUR. There was nearly no ozone destruction observed during the period between mid December 1999 and late January 2000. As expected from ASUR observation of high chlorine activation and continuously low temperatures until mid March, significant ozone depletion was observed between late January and mid March 2000. In order to determine ozone loss it is important to separate dynamical and chemical effects. Since N_2O is a good tracer due to its chemical stability in the lower stratosphere for determining ozone changes due to descent of air, ozone loss can be estimated from simultaneous measurements of ozone and N_2O by ASUR. Between mid December 1999 and mid March 2000 a chemical ozone loss of about 30% (eq 1.1 ppm) in the altitude range between 19.0 and 22.2 km and of about 40% (eq 1.15 ppm) between 16.0 and 18.1 km was observed. The air masses subsided 2.1–3.2 km in the lower stratosphere due to diabatic descent in the period from mid December 1999 to mid March 2000 as derived from ASUR N_2O measurements. Vortex-averaged ASUR measurements of ozone are systematical greater than results from the Global Ozone Monitoring Experiment (GOME) which has a similar vertical resolution than ASUR. This, however, has little impact on the determination of delta ozone and chemical loss estimates. *INDEX TERMS:* 0341 Atmospheric Composition and Structure: Middle atmosphere—constituent transport and chemistry (3334); 0340 Atmospheric Composition and Structure: Middle atmosphere—composition and chemistry; 0933 Exploration Geophysics: Remote sensing; 0394 Atmospheric Composition and Structure: Instruments and techniques; *KEYWORDS:* Ozone depletion, diabatic descent, Arctic stratosphere, microwave radiometry, descent rates, ASUR

Citation: Bremer, H., M. von König, A. Kleinböhl, H. Küllmann, K. Künzi, K. Bramstedt, J. P. Burrows, K.-U. Eichmann, M. Weber, and A. P. H. Goede, Ozone depletion observed by the Airborne Submillimeter Radiometer (ASUR) during the Arctic winter 1999/2000, *J. Geophys. Res.*, *107*(D20), 8277, doi:10.1029/2001JD000546, 2002.

1. Introduction

[2] The Arctic winter stratosphere of 1999/2000 was one of the coldest in the last 10 years. Unusually low temperatures in the lower stratosphere sufficiently low for nitric acid trihydrate (NAT) formation were observed in the northern polar region from December 1999 until early March 2000. This was a longer period than in any of the previous 22 years [*Manney and Sabutis*, 2000]. Previous campaigns showed the strong correlation between chlorine activation on polar stratospheric clouds at low temperatures and catalytic ozone destruction in the polar stratosphere [*World Meteorological Organization* (*WMO*), 1999].

[3] In winter 1999/2000 one of the biggest field campaigns ever in the Northern Hemisphere, the joint European and United States Stratospheric Aerosol and Gas Experiment (SAGE) III Ozone Loss and Validation Experiment/Third European Stratospheric Experiment on Ozone (SOLVE/THESEO-2000) campaign, took place. Many instruments on ground-based, balloon-borne, and airborne platforms participated within this campaign, taking various measurements of trace gases, aerosols, and ozone itself. First calculations performed by the three-dimensional (3-D)

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chemical transport model SLIMCAT show in good agreement with sonde measurements an ozone loss of about 70% in a narrow layer at the 450 K isentrope until end of March 2000 [*Sinnhuber et al.*, 2000].

[4] The Airborne Submillimeter Radiometer (ASUR) participated on board the NASA DC-8 measuring ozone, HCl, ClO, and HNO_3 as well as the dynamical tracer N_2O during all three deployments. This data set, covering the period from just after the formation of the vortex in early December 1999 until the beginning of the breakdown in mid March 2000, allows estimation of the accumulated ozone loss over the course of the entire winter. To distinguish between dynamical and chemical effects, the diabatic descent within the polar vortex was estimated from ASUR N_2O data. Furthermore, ASUR ozone measurements were also compared to satellite observations by the Global Ozone Monitoring Experiment (GOME) [*Burrows et al.*, 1999] during February and March 2000.

2. Instrument

[5] The Airborne Submillimeter Radiometer is a passive heterodyne radiometer developed in 1991 by the University of Bremen [*Crewell et al.*, 1994; *Wehr et al.*, 1995]. By using a liquid helium-cooled superconductor-insulator-superconductor (SIS) detector developed by the Space Research Organization of the Netherlands [*Mees et al.*, 1995; *de Valk et al.*, 1997], system noise temperatures of 550–1450 K (depending on the exact frequency) in single sideband mode are achieved. In 1998 the frequency range was extended and now allows continuous tuning of the instrument over the band from 604.3 to 662.3 GHz. Within this range various key species of ozone chemistry like HCl, ClO, HNO_3, and HO_2, the dynamical tracers N_2O and CH_3Cl as well as water vapor and ozone can be detected. For detection an acousto-optical spectrometer with a spectral resolution of 1.5 MHz and a total bandwidth of 1.5 GHz is used [*Rosolen et al.*, 1994].

[6] ASUR is designed to operate on board an aircraft flying near the tropopause in order to avoid strong absorption due to tropospheric water vapor. In the recent winter 1999/2000, ASUR was installed on board the NASA research aircraft DC-8. Observing to the right side of the aircraft and with an antenna beam angle of 1.3 degrees (full width at half maximum), the instrument takes measurements at a constant zenith angle of 78 degrees.

3. Data Analysis

[7] Before the retrieval of vertical profiles of volume mixing ratio, the individual measured spectra are calibrated, corrected for window transmission, and averaged over a time span of about 100 seconds for ozone and up to 150 seconds for N_2O in order to achieve a sufficient signal-to-noise ratio. The shape of the pressure-broadened lines contain information on the vertical distribution of the emitting molecules. Height-resolved volume mixing ratios are retrieved by using the optimal estimation method, a nonlinear least squares technique described by *Rodgers* [1976, 1990]. To stabilize the solution of the inverse problem, a priori information is used. Multiple line inversion is applied since the observed line of N_2O (652.833 GHz) is located on the wings of strong ozone lines and the observed line for ozone (625.372 GHz) retrieval is adjacent to HCl lines.

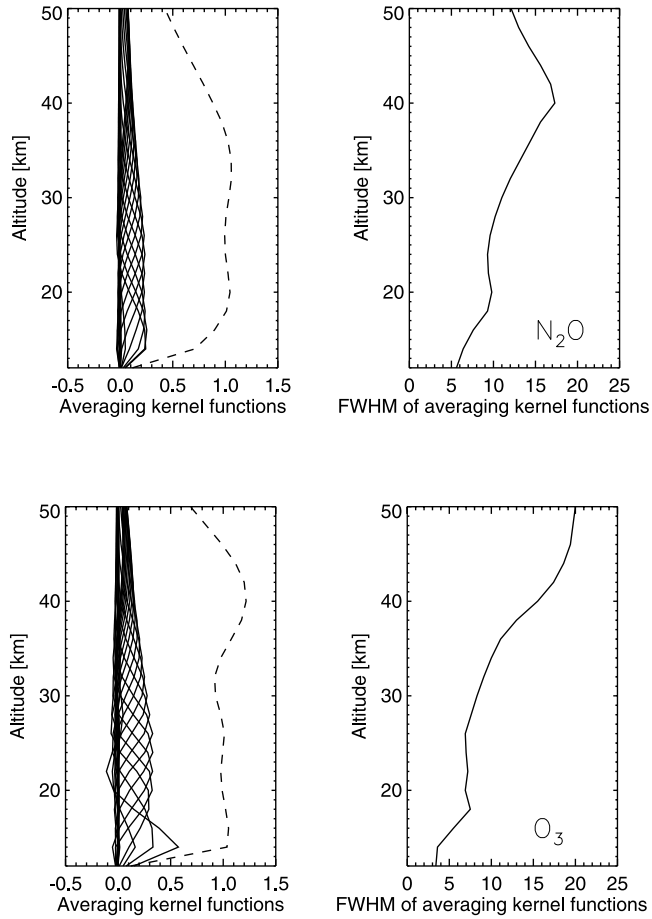

Figure 1. (left) Averaging kernel functions and (right) the full width of half maximum (FWHM) of the averaging kernel functions for (top) a typical N_2O and (bottom) ozone measurement. The dashed line is the sum of the averaging kernels.

[8] The microwave measurement technique results in a typical vertical resolution of 6–10 km in the lower stratosphere, derived from the full width at half maximum of the averaging kernels (Figure 1). The averaging kernel function of a certain altitude level shows the contribution of other altitude levels to the retrieved result at the considered altitude. Values around 1 in the envelope curve (the sum of averaging kernels) indicate that the major contribution on the solution comes from the measurement. It can be concluded that between 16 and 50 km (40 km for N_2O) the solution is not dominated by a priori information. The horizontal resolution depends on aircraft speed and the required integration time to achieve a sufficient signal-to-noise ratio. Typical values are 20–30 km for ozone and N_2O.

[9] The accuracy for ozone is 0.3 ppm or 15%, whichever is greater, and 30 ppb or 15% for N_2O, respectively. Errors are mainly determined by the signal-to-noise ratio, the line strength, and the spectral resolution. Typical values for the precision, which is limited by the measurement noise, are 0.1 ppm for ozone and 15 ppb for N_2O. For more detailed

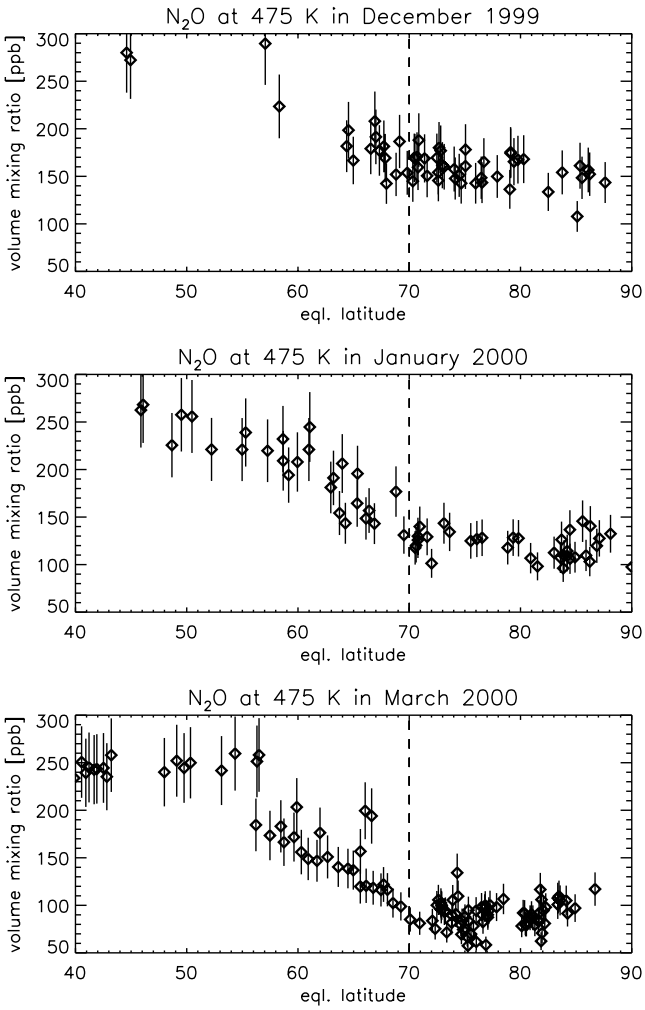

Figure 2. All ASUR N_2O measurements during winter 1999/2000 as a function of equivalent latitude. N_2O volume mixing ratios in ppb at 475 K of potential temperature are plotted. (top) December 1999, (middle) January 2000, and (bottom) March 2000. Error bars indicate precision of the single measurement.

information on data analysis the reader is referred to *Wehr et al.* [1995].

4. Diabatic Descent Inside the Polar Vortex

[10] To estimate the vertical motion of the air masses inside the polar vortex, tracer measurements are used. N_2O is well suited in the lower stratosphere as it has only tropospheric sources and is mainly removed by photolysis at high altitudes. This results in a lifetime of more than 1 year below 33 km altitude [*WMO*, 1986].

[11] During three deployments of the DC-8, ASUR took numerous measurements of N_2O within the polar vortex. To distinguish between inside and outside vortex measurements, the data are sorted by equivalent latitude. The inner and outer edge of the vortex are estimated by finding the minimum and maximum of the second derivative of potential vorticity as a function of equivalent latitude [*Nash et al.*, 1996]. During winter 1999/2000 the inner edge varied between 65 and 70 degrees equivalent latitude on the 475 K surface of potential temperature. The region with an equivalent latitude larger than 70 degrees is for all three deployments of the NASA DC-8 well inside the polar vortex. This is also illustrated by Figure 2, showing all ASUR N_2O measurements at 475 K versus equivalent latitude for the three deployments (top, December 1999; middle, January 2000; bottom, March 2000). In December, N_2O measurements reached a constant level at 64 degrees equivalent latitude and above while in January and March constant values were reached at 70 degrees.

[12] *Manney and Sabutis* [2000] pointed out that the vortex was not well defined below 500 K in November 1999. Maps of potential vorticity calculated with a reverse domain filling method show that some mixing into the vortex occurred during late November and early December but ceased by mid December. Afterward the vortex remained stable in the lower stratosphere until breakdown started in mid March 2000. The minor warming event in February 2000 weakened the vortex and detached parts of the boundary region, but the inner vortex region ($\geq$70 degrees of equivalent latitude) was relatively unaffected by extravortex intrusion. From the top panel in Figure 2 it can be seen that in a region between 64 and 70 degrees of equivalent latitude, ASUR N_2O measurement show slightly greater values of volume mixing ratio (approximately 20 ppb) than in the vortex core. It has to be noted that the measurements shown were recorded on eight flights between 30 November and 15 December. We assume that noncontinuous horizontal mixing from regions outside the vortex leads to a significant variation of N_2O within the vortex. Figure 2 clearly shows constant N_2O levels above 70 degrees equivalent latitude supporting our assumption.

[13] Figure 3 shows mean vertical profiles of N_2O averaged for each deployment (40–75 single measure-

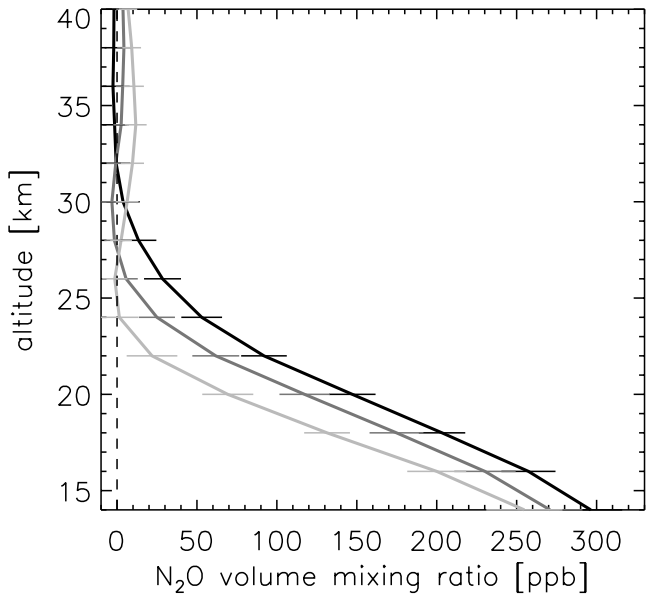

Figure 3. Evolution of ASUR N_2O measurements during winter 1999/2000. The profiles are an average of all measurements with an equivalent latitude between 70 and 90 degrees for each deployment. Black; December 1999; grey; January 2000; light grey; March 2000. Error bars indicate standard deviation about the mean.

ments), all taken well inside the polar vortex at an equivalent latitude between 70 and 90 degrees. The error bars correspond to the standard deviation about the mean.

[14] The error of a single N_2O measurement due to noise is smaller than ±15 ppb for all altitudes. Analysis has shown that a vertical shift of the profile of less than 1 km can easily be detected.

[15] From mid December 1999 until late January 2000, diabatic descent of about 1.6 km at the 100 ppb level was derived (dropping from 22.2 to 20.6 km). Between late January and mid March 2000 the descent was 1.6 km (from 20.6 to 19.0 km). At the 100 ppb N_2O level the total diabatic descent from early December 1999 until mid March 2000 was approximately 3.2 km and at the 200 ppb level (18.1 km down to 16.0 km) approximately 2.1 km. This is equivalent to 52 ± 16 K or 0.58 ± 0.18 K d^{-1} in units of potential temperature at the 475 K isentrope in March representing approximately the 100 ppb level. Other measurements from various instruments have shown that from 26 November until 5 March the vortex subsided 65 ± 12 K averaged over 40–280 ppb N_2O (J. B. Greenblatt et al., Experimental determination of vortex subsidence for the 1999–2000 Arctic Winter and comparison with models, submitted to *Journal of Geophysical Research*, 2001) which is in good agreement with ASUR.

5. Ozone Depletion

5.1. ASUR Measurements

[16] Figure 4 gives an overview of all ASUR ozone measurements inside the polar vortex (70–90 degrees equivalent latitude). Vertical profiles are shown as a function of equivalent latitude for all three DC-8 deployments. Individual measurements are binned every 0.5 degrees of equivalent latitude. The crosses in the bottom of each panel indicate individual measurements.

[17] Between 24 and 30 km altitude the ozone measurements show an increase of up to 2.25 ± 0.1 ppm from December 1999 until January 2000 and up to 0.5 ± 0.1 ppm from January 2000 until March 2000. This increase is mainly caused by meridional transport of air from the tropical source region at high altitudes into the vortex and subsequent descent into lower altitudes at polar latitudes.

[18] Between 18 and 24 km altitude the ozone volume mixing ratio mainly remained constant between the first (December) and second (January) deployment. During the third deployment in March the ozone measurements show a decrease of up to 0.6 ± 0.1 ppm compared to January. This decrease is caused by photochemical destruction of ozone which is partly compensated by descending air due to diabatic cooling.

[19] Below 18 km an increase of 0.25 ± 0.1 ppm between December and January was observed due to diabatic descent of air with greater volume mixing ratios. For the time period between the second and third deployment, ozone values decreased by 0.6 ± 0.1 ppm due to chemical destruction.

[20] Chemical changes within ozone measurements shown in Figure 4 are masked by dynamical effects due to diabatic cooling of air. An approach to separate chemistry and dynamics with the use of ASUR N_2O measurements will be given in section 5.3.

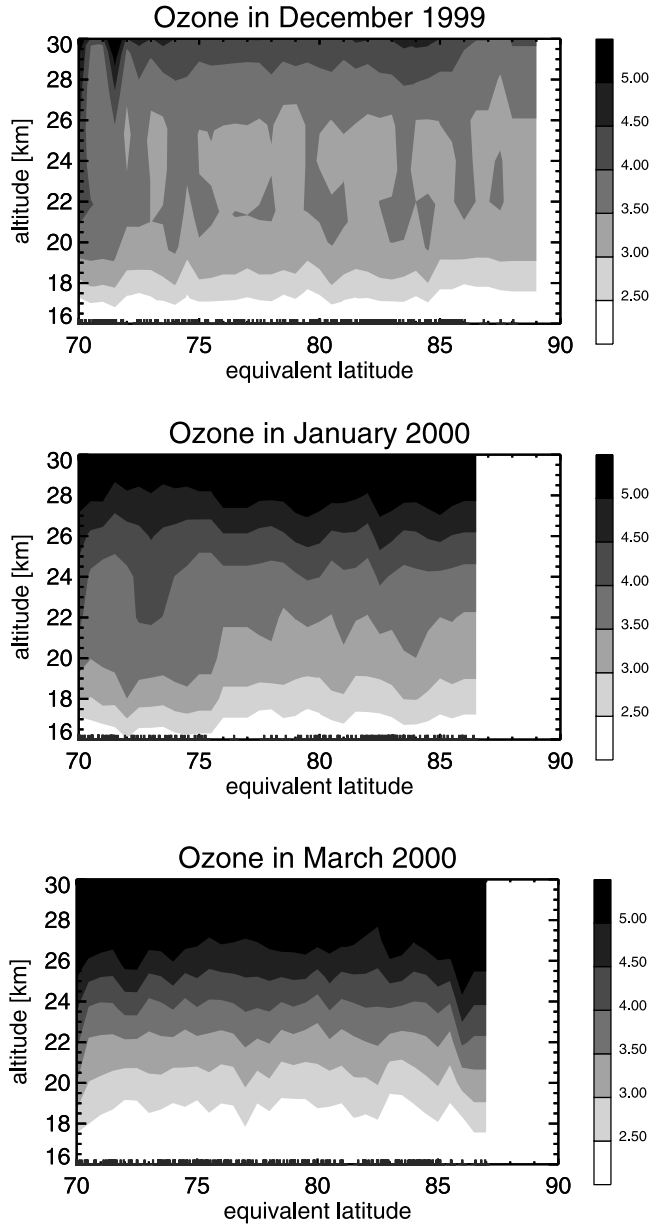

Figure 4. Evolution of ozone volume mixing ratio (ppm) of all ASUR measurements within the polar vortex. Individual measurements are binned every 0.5 degrees. Crosses in the bottom indicate individual measurements.

5.2. Comparison ASUR-GOME

[21] The Global Ozone Monitoring Experiment (GOME) aboard the ERS-2 satellite was launched in April 1995 [*Burrows et al.*, 1999]. The ERS-2 satellite is in a Sun-synchronous orbit allowing GOME a complete coverage at the equator within 3 days and less for higher latitudes. GOME detects earthshine radiance and solar irradiance in the UV/visible (UV/VIS) spectral range. Ozone vertical profiles can be retrieved by inversion of the radiances with an algorithm similar to the one used for NASA's solar backscatter UV (SBUV) instrument [*Bhartia et al.*, 1996] and optimized for GOME [*de Beek et al.*, 1997; *Hoogen et al.*, 1999]. The profiles have a vertical resolution of about 6–8 km in the lower stratosphere and more below and above.

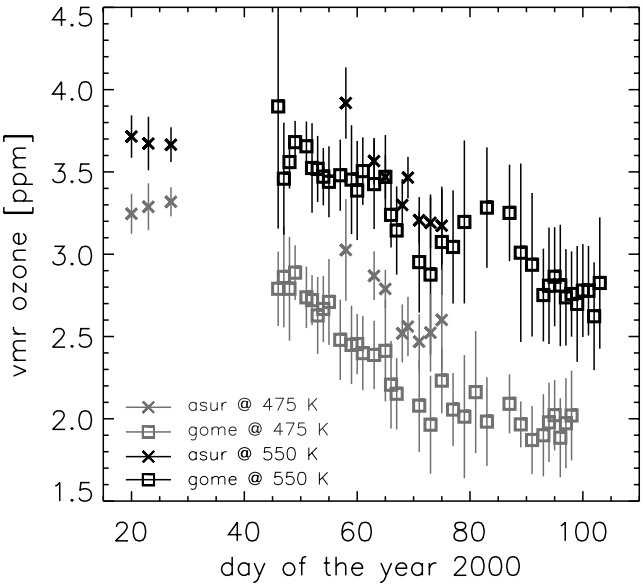

Figure 5. Comparison between ASUR and GOME ozone measurements at 475 K (ASUR, grey crosses; GOME, grey squares) and 550 K (ASUR, black crosses; GOME, black squares) isentropic level inside the vortex (vmr, volume mixing ratio). Error bars are the sum of standard deviation of the single measurements and uncertainties for the given potential temperature level.

Measuring in UV/VIS, GOME needs sunlight and reaches sufficient coverage of the polar vortex at day 45 in winter 1999/2000 [*Weber et al.*, 2000; *Eichmann et al.*, 2002].

[22] The comparison of ASUR and GOME daily vortex averaged ozone values as a function of time is shown in Figure 5. Error bars illustrate the sum of standard deviation and uncertainty within the potential temperature. Because of similar vertical resolution of both instruments, volume mixing ratios on isentropic surfaces are comparable. While ASUR ozone measurements show up to 20% greater values at the 475 K isentrope of potential temperature than GOME, the mixing ratios at the 550 K isentrope agree to within 10%. ASUR ozone data seem to overestimate ozone at the peak altitude, but this has no impact on comparisons of trends and delta ozone. The observed trends are similar for GOME and ASUR. The decrease of ozone was not completed by mid March at the end of the ASUR measurements but was still ongoing until end of March 2000. During the ASUR measurement period GOME measured a decrease of 0.6 ± 0.1 ppm ozone at 475 K and 0.7 ± 0.1 ppm at 550 K, respectively. From ASUR data an ozone loss of 0.75 ± 0.1 ppm at 475 K and 0.6 ± 0.1 ppm at 550 K can be derived. For the entire period from day 45 until day 90 an ozone reduction of 1.05 ± 0.1 ppm for both levels can be estimated from GOME data at 475 K and 550 K in good agreement with ASUR. However, mid winter measurements have been covered by ASUR only, since GOME measurements were not possible because of lack of sunlight.

5.3. Estimation of Ozone Loss

[23] Using N_2O as a tracer to distinguish between dynamical and chemical effects, chemical ozone loss can be estimated from a correlation of simultaneous ozone and N_2O measurements (see also *Proffitt et al.* [1989, 1990]). The surface of constant N_2O mixing ratio follows air motion more precisely than isentropic surfaces do since the lifetime of N_2O is much longer than that of potential temperature [*Plumb and Ko*, 1992]. The N_2O tracer method was employed earlier on ASUR data for correlative purposes [*de Valk et al.*, 1997].

[24] Figure 6 (top) shows daily mean ozone values inside the polar vortex (70–90 degrees equivalent latitude) on two different N_2O levels for all three deployments. Given in Figure 6 (bottom) are potential temperature versus day of the year 2000. Error bars in both panels are derived from the sum of standard deviation and the uncertainty in the given

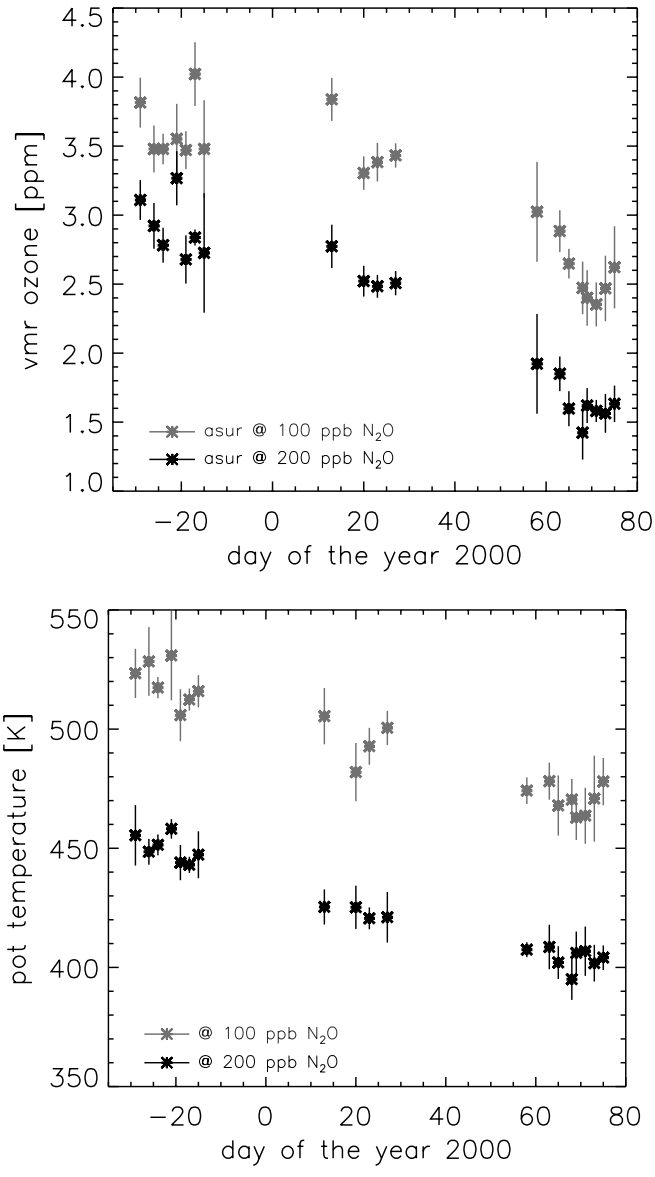

Figure 6. (top) Daily mean ASUR ozone volume mixing ratios (vmr, ppm) and (bottom) potential temperature derived from DAO (Data Assimilation Office) analysis at two different N_2O levels within the polar vortex: black stars, 200 ppb N_2O; grey stars, 100 ppb N_2O. Error bars are the sum of standard deviation of the individual measurements and uncertainties for the given N_2O level.

N_2O level. The 100 ppb N_2O level represents an altitude of 22.2 km (525 K) in December 1999 which subsided down to 19 km (470 K) until March 2000. The second level at 200 ppb N_2O corresponds to an altitude of 18.1 km (450 K) in early December and 16.0 km (405 K) in March.

[25] In December 1999 the ozone single measurements show a greater variation than in later months. *Manney and Sabutis* [2000] pointed out that the November vortex was not well defined but strengthened rapidly during December. This may have resulted in a vortex that was not well mixed in early December. In this period some mixing between the inner vortex and the boundary region may have occurred. ASUR ozone measurements taken within the boundary region showed 0.3 ± 0.1 ppm greater values of volume mixing ratio at 20 km altitude than inside the vortex. The next to the last flight during the first deployment shows 0.5 ppm greater ozone values at the 100 ppb N_2O level than adjacent flights. This flight was mainly conducted near the edge of the vortex (over southern Sweden) and only a few measurements were taken inside the vortex. Ozone rich air from the vortex boundary region may have been transported into the vortex which could explain the greater ozone values. For further discussion of ozone loss, only measurements of the four last flights of the December deployment will be used as initial values. The single measurements show similar (within the error bars) volume mixing ratios, and it is assumed that the main mixing events had occurred before mid December. The assumption is supported by calculation of potential vorticity maps with a reversed domain filling method showing that mixing into the vortex was negligible after mid December.

[26] In general, ASUR ozone measurements show no significant ozone loss (0.1 ± 0.1 ppm) from December to January 2000 at 100 ppb N_2O (approximately 525 K in December and 490 K in January, respectively). In the lower stratosphere (at 200 ppb N_2O, approximately 450 to 420 K) there is 0.25 ± 0.1 ppm loss from mid December until January 2000. The main loss occurred between the second and the third deployment of the DC-8 in January and March 2000 at both N_2O levels.

[27] During December 1999, no chlorine measurements were taken within the polar vortex and with a solar zenith angle smaller than 86 degrees by ASUR (Figure 7). In January, with decreasing solar zenith angle in the vortex region, high chlorine activation was observed with peak values up to 1.8 ppb ClO anticorrelated with very low HCl mixing ratios. While in the beginning of March mean values of 0.9 ppb ClO were still observed by ASUR, the values decreased rapidly until mid March indicating deactivation into $ClONO_2$ [*von König*, 2001]. The HCl volume mixing ratio remained very low until mid March. The ozone depletion anticorrelates well with the observed chlorine activation with only little losses during the dark period (December 1999 until January 2000).

[28] Consistent with high active chlorine values observed in January and March 2000, large ozone loss was derived from ASUR measurements. At 100 ppb N_2O level (approximately 470 K in March) a decrease of 1.0 ± 0.1 ppm and 0.9 ± 0.1 ppm at 200 ppb (approximately 405 K in March) was derived. For the entire time period (early December 1999 until mid March 2000) a total chemical loss of about 40% at the 200 ppb N_2O level and about 30% at 100 ppb

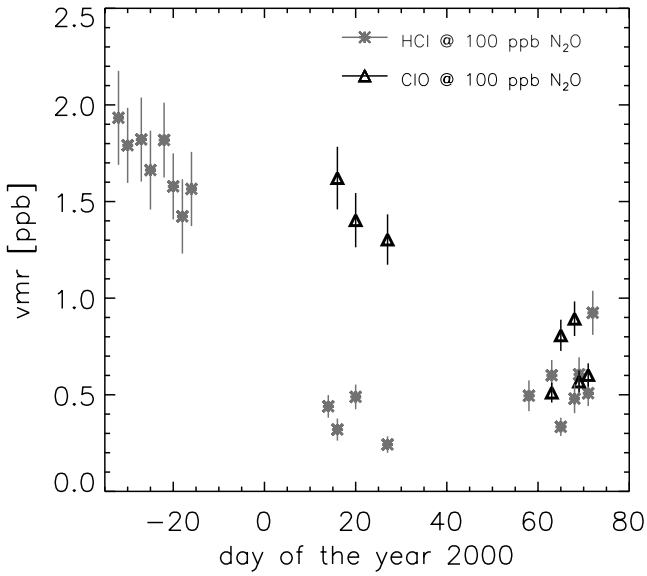

Figure 7. ASUR daily mean of HCl (grey stars) and ClO (black triangles) volume mixing ratios (vmr) within the polar vortex at 100 ppb N_2O. Only ClO measurements with solar zenith angles smaller than 86 degrees are shown. Error bars are the sum of standard deviation of the individual measurements and uncertainties for the given N_2O level. In December 1999, no ClO measurements were taken with an equivalent latitude greater 70 degrees and a solar zenith angle smaller than 86 degrees.

N_2O were estimated (see Figure 6). This is equivalent to a mean ozone loss rate of 19 ± 2 ppb per day between mid January and mid March 2000.

[29] As mentioned before, GOME measurements reached sufficient coverage of the polar vortex by day 45. At this time, chlorine-catalyzed chemical ozone depletion was already in progress. On the other hand, the ASUR measurements were completed by mid March, while the ozone loss was still ongoing (as shown in Figure 5). To account for vertical motion of air masses in the GOME ozone measurements, diabatic heating rates are calculated using a radiative transfer model [*Eichmann et al.*, 1999]. Taking diabatic descent into account, GOME measurements show a total loss of ozone at the 475 K isentropic surface (eq ≈ 100 ppb N_2O in March 2000) of about 40% during winter 1999/2000. This is in agreement with the loss observed by ASUR.

6. Conclusions

[30] From ASUR N_2O measurements a mean diabatic descent rate of 0.58 ± 0.18 K per day at the 475 K isentrope (in March 2000) was derived for polar vortex air masses. This is equivalent to a subsidence of 2.1–3.2 km in the lower stratosphere from mid-December 1999 until mid March 2000.

[31] ASUR vortex ozone measurements show a slight increase of 0.25 ± 0.1 ppm below 18 km altitude between early December and mid January due to descent of ozone rich air into the lower stratosphere. Between January and March an ozone decrease of 0.6 ± 0.1 ppm was observed in the lower stratosphere.

[32] During the period with sufficient low solar zenith angle within the vortex region, GOME measured an ozone decrease of 1.05 ± 0.1 ppm between day 45 and 90 of the year 2000. This is equivalent to an ozone loss of 40% at the 475 K isentropic surface. The comparison with ASUR shows a good agreement for this ozone trend in both data sets.

[33] Taking the diabatic descent into account, an accumulated loss of up to 1.10 ± 0.1 ppm, 30% (at 470 K, 100 ppb N_2O) and 1.15 ± 0.1 ppm, 40% (at 405 K, 200 ppb N_2O) from mid December 1999 until mid March 2000 was estimated from ASUR. Considering diabatic descent, a mean ozone chemical loss rate of 19 ± 2 ppb per day at 475 K of potential temperature could be derived between January and March.

[34] Measurements of ozone sondes showed a reduction of more than 70% in the initial ozone values over a 1-km-thick layer near 18 km altitude and more than 50% ozone loss over a 3-km thick layer [*Rex et al.*, 2002]. This is in good agreement with calculations by SLIMCAT which showed a total loss of 70% in a narrow layer at the 450 K isentropic surface [*Sinnhuber et al.*, 2000]. At this altitude the vertical resolution of the ASUR instrument is approximately 8 km (see Figure 1). Taking this into account, the sondes measurements agree well with the loss estimated by ASUR.

[35] **Acknowledgments.** We would like to acknowledge G. Näveke for technical support before and during the campaign. We would also like to thank the European Union for funding our campaign participation (contract EVK2-CT-1999-00047), NASA for the opportunity to participate on board the DC-8, and the aircraft crew for their excellent support.

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