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[1] We present ozone loss estimated from airborne measurements taken during January–February and March in the Arctic winter 2002/2003. The first half of the winter was characterized by unusually cold temperatures and the second half by a major stratospheric sudden warming around 15–18 January 2003. The potential vorticity maps show a vortex split in the lower stratosphere during the major warming (MW) in late January and during the minor warming in mid-February due to wave 1 amplification. However, the warming can be termed as a vortex displacement event as there was no vortex split during the MW period at 10 hPa. Very low temperatures, large areas of polar stratospheric clouds (PSCs), and high chlorine activation triggered significant ozone loss in the early winter, as the vortex moved to the midlatitude regions. The ozone depletion derived from the ASUR measurements sampled inside the vortex, in conjunction with the Mimosa-Chim model tracer, shows a maximum of 1.3 ± 0.2 ppmv at 450–500 K by late March. The partial column loss derived from the ASUR ozone profiles reaches up to 61 ± 4 DU in 400–550 K in the same period. The evolution of ozone and ozone loss assessed from the ASUR measurements is in very good agreement with POAM observations. The reduction in ozone estimated from the POAM measurements shows a similar maximum of 1.3 ± 0.2 ppmv at 400–500 K or 63 ± 4 DU in 400–550 K in late March. Our study reveals that the Arctic winter 2002/2003 was unique as it had three minor warmings and a MW, yet showed large loss in ozone. No such feature was observed in any other Arctic winter in the 1989–2010 period. In addition, an unusually large ozone loss in December, around 0.5 ± 0.2 ppmv at 450–500 K or 12 ± 1 DU in 400–550 K, was estimated for the first time in the Arctic. A careful and detailed diagnosis with all available published results for this winter exhibits an average ozone loss of 1.5 ± 0.3 ppmv at 450–500 K or 65 ± 5 DU in 400–550 K by the end of March, which exactly matches the ozone depletion derived from the ASUR, POAM and model data. The early ozone loss together with considerable loss afterwards put the warm Arctic winter 2002/2003 amongst the moderately cold winters in terms of the significance of the ozone loss.

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1. Introduction

[2] Polar stratospheric ozone depletion in the Arctic was first identified in 1989 [*Hofmann et al.*, 1989] and significant reduction in ozone has been measured since then in each cold winter [*World Meteorological Organization* (*WMO*), 2007]. The difference in ozone loss from one winter to the other is found to be extremely large and is highly controlled by temperature history of the winters. The meteorology of Arctic winters is characterized by intermittent stratospheric sudden warmings. Therefore, the extent of ozone loss in an Arctic winter is determined by the dynamics of the region. This is clearly manifested with the range of ozone depletion

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Table 1. The Flight Route of the Falcon-20 Research Aircraft During the EuPLEx 2003 and SCIA-VALUE 2003 Campaigns in the Arctic Winter 2002/2003

Flight Date[a]	Flight Track
EuPLEx	
14-01-2003	Munich - Kiruna
15-01-2003	Kiruna local flight
19-01-2003	Kiruna local flight
23-01-2003	Kiruna local flight
26-01-2003	Kiruna local flight
07-02-2003	Kiruna local flight
08-02-2003	Kiruna local flight
09-02-2003	Kiruna local flight
12-02-2003	Kiruna - Munich
SCIA–VALUE	
10-03-2003	Munich - Kiruna
12-03-2003	Kiruna - Ny Ålesund - Kiruna
13-03-2003	Kiruna - Keflavik
14-03-2003	Keflavik - Kangerlussuaq
15-03-2003	Kangerlussuaq - Keflavik
17-03-2003	Keflavik - Munich
19-03-2003	Munich local flight

[a]Dates given as dd-mm-yyyy.

observed over the years, with 5–7% or <40 DU in warm winters and 25–30% or >60 DU in cold winters [*Andersen and Knudsen*, 2002; *Harris et al.*, 2002; *Rex et al.*, 2002, 2004; *Goutail et al.*, 2005; *Tilmes et al.*, 2006; *WMO*, 2007; *Müller et al.*, 2007; *Blumenstock et al.*, 2009]. Another important feature observed is the spread in the ozone loss derived by different measurement techniques in each year. For instance, *Newman et al.* [2002] list a deviation from 0.7 to 2.1 ppmv for the Arctic winter 2000 and *Kuttippurath et al.* [2010] find a similar range in the ozone loss for the Arctic winter 2005 by various methods. However, this high spread to a large extent is due to differences in sampling and estimation method. Therefore, a reasonable agreement among various ozone loss estimates can be reached by selecting a common criterion for the loss estimations, such as similar vortex sampling, vortex edge criterion, time of the estimation, and the same method of loss computation, as demonstrated by *Harris et al.* [2002]. The large interannual variability and the differences in estimated ozone losses still attest the necessity to assess ozone loss in each Arctic winter by different methods. It is also essential to diagnose the evolution of ozone in each winter to assist the interpretation and prediction of its future development in an ozone recovery perspective. Therefore, in this study we present the ozone loss determined using airborne measurements, in combination with satellite observations, for the Arctic winter 2002/2003 and compare the inferred loss with other available results for this winter and other Arctic winters.

[3] The Arctic winter 2002/2003 was exceptional as it was unusually cold in the first half and was subjected to a major warming (MW) in the second half. Two major field campaigns were conducted to probe the evolution of ozone, polar stratospheric clouds (PSCs), and ozone loss by various instruments during the winter. The campaigns were executed in the framework of the European Polar Lee-wave Experiment (EuPLEx) in January–February 2003 [*Kleinböhl et al.*, 2005] and the SCIAMACHY validation and utilization experiment (SCIA–VALUE) in March 2003 [*Kuttippurath et al.*, 2007]. A suite of instruments participated in the campaign and performed ground-based, airborne and in situ measurements [*Christensen et al.*, 2005; *Goutail et al.*, 2005; *Raffalski et al.*, 2005; *Streibel et al.*, 2006; *Kuttippurath et al.*, 2007]. The airborne sub-millimeter radiometer (ASUR) was aboard the German Falcon-20 aircraft to perform measurements as a part of these campaigns. Both of the surveys provided a good set of ASUR trace gas measurements, including ozone and Chlorine Monoxide (ClO). We investigate the ozone loss features of the winter using the ASUR observations and compare with the results drawn from other works [*Tilmes et al.*, 2003; *Urban et al.*, 2004; *Christensen et al.*, 2005; *Feng et al.*, 2005; *Goutail et al.*, 2005; *Grooß et al.*, 2005; *Raffalski et al.*, 2005; *Singleton et al.*, 2005; *Streibel et al.*, 2006; *Tripathi et al.*, 2006; *Konopka et al.*, 2007; *Müller et al.*, 2007; *El Amraoui et al.*, 2008; *Ryskin and Kulikov*, 2008; *Sonkaew et al.*, 2011]. In this study we analyze the meteorological situation, evolution of ozone and chemical ozone loss with the ASUR measurements, which have hitherto not been used for the study of Arctic ozone loss in 2002/2003. To get the complete evolution of ozone and ozone loss during the winter, we complement the ASUR measurements with POAM observations. The passive tracer method is applied to compute ozone loss from the measurements [e.g., *Kuttippurath et al.*, 2010], for which the passive tracer needed is simulated by the chemical transport model (CTM) Mimosa-Chim [e.g., *Kuttippurath et al.*, 2009]. Therefore, the ozone loss determined from the measurements is compared to the modeled loss too.

[4] We organize the article in the following way. First we introduce the ASUR and POAM measurements in Section 2 and then the model simulations in Section 3. The results are presented in Section 4, in which the meteorology of the winter and temporal evolution of the vortex are discussed. This section also compares the ozone measurements and ozone loss estimated from ASUR to that of POAM, both in mixing ratio and partial column. Section 5 discusses the ozone loss found from ASUR, POAM and the model with other published results for the studied winter and Section 6 compares ozone loss inferred in this winter to those of other Arctic winters. Section 7 concludes the study.

2. Measurements

2.1. ASUR

[5] The ASUR measurements taken during the EuPLEx and SCIA–VALUE 2003 campaigns are used here. The campaigns and observations are described in detail by *Kuttippurath et al.* [2007]. A total of 12 flights, between 13 January 2003 and 20 March 2003, was carried out with more than 70 flight hours of measurements for various stratospheric constituents. Both campaigns surveyed similar latitudes between 50° and 80°N with a focus on the polar vortex. Therefore, a large number of measurements were taken inside the vortex to allow a reasonable analysis of the polar processing and ozone loss. Further details of these measurements are given in the aforesaid references and in Table 1 and Section 4.2.

[6] ASUR is a passive heterodyne receiver operating in a tuning frequency range at 604.3–662.3 GHz. The receiver has two spectrometers; an acousto-optical spectrometer (AOS) and a chirp transform spectrometer. We use the stratospheric measurements performed with the AOS, which

has a bandwidth of 1.5 GHz and a resolution of 1.27 MHz. The observations are performed on board a research aircraft to avoid signal absorption by tropospheric water vapor. The sensor observes upward at a constant zenith angle of 78° and measures thermal emissions from the rotational states of the observed species. Vertical profiles of ozone and ClO are retrieved in an equidistant altitude grid of 2 km spacing using the method of *Rodgers* [1976]. An in-house radiative transfer model is applied to invert the measurement spectra for a non-scattering atmosphere, for which the a priori profile was taken from *Bremer et al.* [2002]. The altitude range of ASUR ozone and ClO is 15–50 km and the vertical resolution of both measurements is about 6–25 km, where the resolution decreases with altitude from the lower to the upper stratosphere. Horizontal resolution of the measurements is 18 km and 40 km for ozone and ClO, respectively, and the accuracy of measurements is about 12–15% [*Kuttippurath et al.*, 2007].

2.2. POAM

[7] The Polar Ozone and Aerosol Measurement (POAM)–III, a United States Naval Research Laboratory space experiment, was launched on the French Système Probatoire d'Observation de la Terre–4 satellite in March 1998 into a polar, sun-synchronous orbit. In this orbit 14 occultations were obtained per day around a circle of latitude in each hemisphere, with consecutive observations separated by ~25° longitude. The latitude range is 63°–88° in the southern hemisphere, and 55°–71° in the northern hemisphere (NH). The NH measurements were carried out during the spacecraft sunrise that corresponds to local sunset. The instrument operated in its nominal auto mode and measured atmospheric slant path transmission in 9 channels at 354–1018 nm. Inversion of the optical depth data yields vertical profiles of ozone in the altitude range of 13–60 km with a vertical resolution of about 1 km and an accuracy of ±5% [*Randall et al.*, 2003].

3. Ozone and Tracer Simulations

[8] We have used the Mimosa-Chim CTM for the ozone and passive tracer simulations. This model has been used successfully in polar ozone loss studies [e.g., *Kuttippurath et al.*, 2009, 2010; *Tripathi et al.*, 2006, 2007]. The spatial domain of the model is 30°–90°N with 1° × 1° horizontal resolution. There are 16 isentropic vertical levels between 350 K and 950 K with a resolution of 1.5–2 km. The model is forced by European Centre for Medium-Range Weather Forecasts (ECMWF) analyses. The chemical fields are initialized from the 3-D CTM REPROBUS output [*Lefèvre et al.*, 1998] and it uses the MIDRAD radiation scheme [*Shine*, 1987]. Climatological H_2O, CO_2 and interactive O_3 fields are used for the calculation of heating rates. The kinetic data are taken from *Sander et al.* [2003], but the Cl_2O_2 photolysis cross-sections from *Burkholder et al.* [1990], with a log linear extrapolation up to 450 nm [*Stimpfle et al.*, 2004]. These are in very good agreement with the Cl_2O_2 spectrum measurements by *Papanastasiou et al.* [2009]. Note that these new measurements form the basis of the JPL 2011 recommendation. A detailed sensitivity study using different Cl_2O_2 scenarios in the model with respect to various Arctic winters, including 2002/2003, has already been presented by *Tripathi et al.* [2006, 2007]. The study shows a difference of about 2% in the estimated ozone loss among the tests. Therefore, to compare with other model results for this winter we have used the results from *Burkholder et al.* [1990] for this model run. The model includes the chemical scheme of REPROBUS that contains 55 species and 160 reactions including gas phase, heterogeneous, and photolytic reactions [*Lefèvre et al.*, 1998]. The Br_y in the model is based on a correlation with CFC-11 that considered supply of bromine from CH_3Br, halons, as well as CH_2Br_2 and CH_2BrCl [*Wamsley et al.*, 1998].

[9] The model has a detailed scheme of PSC formation and growth. The saturation vapor pressure given by *Hanson and Mauersberger* [1988] is used to assume the existence of Nitric Acid Trihydrate (NAT) particles and the one given by *Murray* [1967] is considered for ice particles. Equilibrium composition and volume of binary (H_2SO_4–H_2O) and ternary (HNO_3–H_2SO_4–H_2O) droplets are computed using an analytic expression provided by *Carslaw et al.* [1995]. Liquid supercooled sulphuric acid aerosols, NAT, and ice particles are considered in equilibrium with the gas phase [*Lefèvre et al.*, 1998]. For NAT and ice particles, the number density is set to 5×10^{-3} cm^{-3} and the particle diameter is calculated within the scheme, from available volume of HNO_3 and water. A denitrification scheme is incorporated to account for the sedimentation of HNO_3 containing particles where the NAT particles are assumed to be in equilibrium with gas phase HNO_3. All the three types of particles – NAT, ice, and liquid aerosols – are considered in the sedimentation module and the sedimentation speed of the particles is calculated according to *Pruppacher and Klett* [1997]. Nevertheless, recent studies indicate that PSCs do not frequently exist at NAT temperatures [*WMO*, 2011; *Pitts et al.*, 2007] and liquid aerosols often dominate heterogeneous halogen processing [*Portmann et al.*, 1996]. Therefore, care must be taken when comparing these results with studies using a different PSC scheme.

4. Results

4.1. Evolution of the Winter

[10] Figure 1 presents the temperature, zonal wind and heat flux together with other dynamical entities at 60°N/10 hPa to assess the meteorological situation of the winter. Note that a detailed discussion of the evolution of polar vortex during this winter has been presented by *Günther et al.* [2008]. Therefore, a similar analysis will not be presented here. The minimum temperature extracted from ECMWF analyses at 40°–90°N for the winter shows very low values, below 195 K, from mid-November to mid-January. Further, the temperatures show exceptionally low values and hence, the winter was unusually cold in December and early January. Though there was a warming in late January, the temperatures were again set to cold scales of <195 K, in early and late February. A minor warming in mid-February and early March is also apparent. In short, the winter was remarkably cold in the first half and very warm with three occasional warmings in the second half.

[11] In order to investigate whether the warming was major or not, we now look at the temperatures at 90°N and 60°N (Figure 1, second panel) together with zonal wind at 60°N and 10 hPa (third panel), where the criterion of a MW

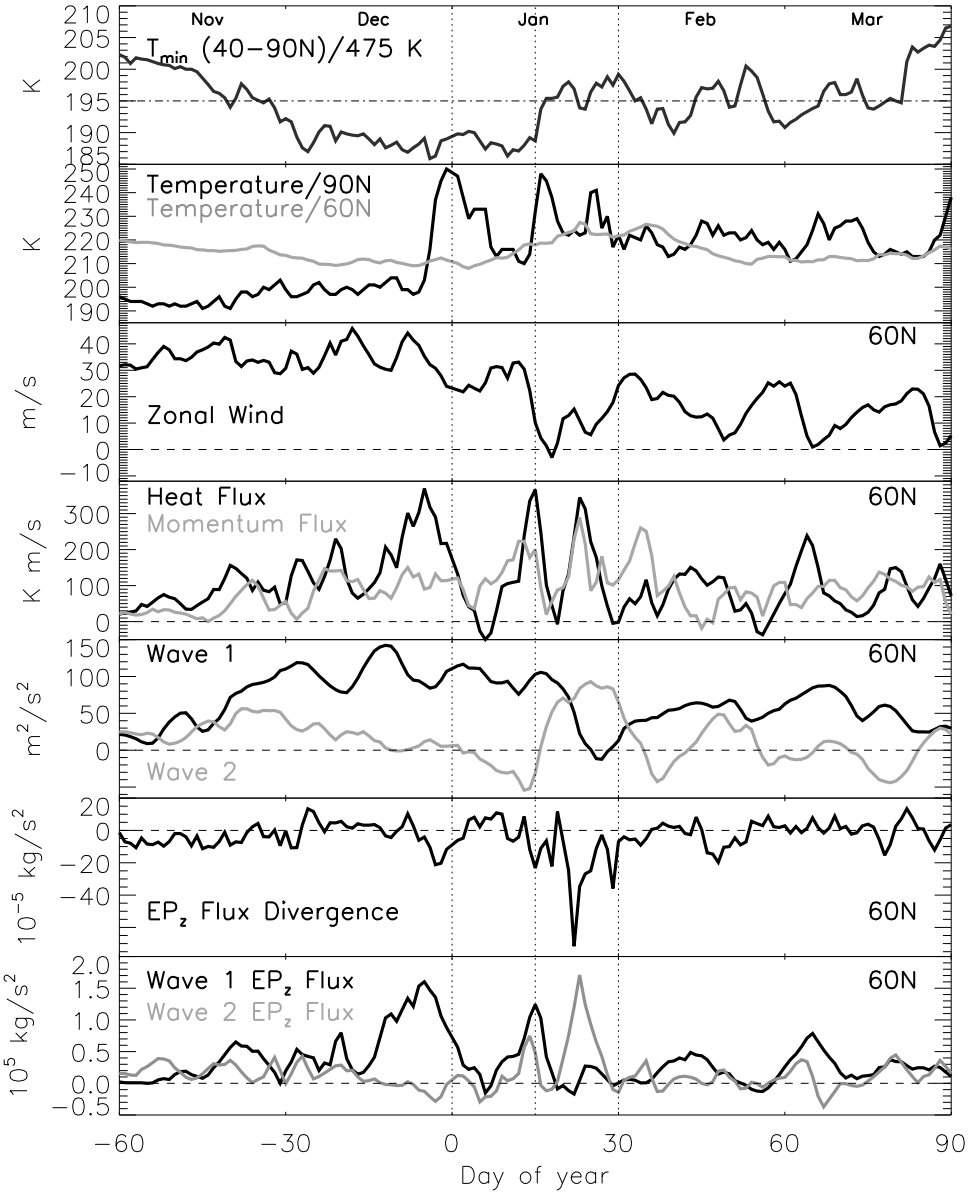

Figure 1. Meteorological situation of the Arctic winter 2002/2003. Minimum temperature extracted from the ECMWF data in 40°–90°N at 475 K, temperature at 60°N and 90°N at 10 hPa, and zonal wind together with other dynamical entities at 60°N and 10 hPa, where the major warming criterion is defined. The dotted vertical lines represent-days 1, 15 and 30 of January 2003, the dashed lines mark 0, and the dash-dotted line demarcates 195 K.

is generally examined [*McInturff*, 1978]. As shown by the minimum temperature distributions (Figure 1, top panel), very cold temperatures are evident in November, December and early February at both latitudes (Figure 1, second panel). However, an abrupt increase in temperature was found in late December, from 198 K to 252 K, within a few days time at 90°N, apart from the minor warmings in mid-February and early March. The warming at 60°N was comparatively slow, where it showed an increment of 18 K in a couple of weeks; from 208 K in early January to 226 K by late January. In conjunction with the high temperatures, the zonal wind reversed on 18 January 2003 and thus, fulfilled the condition for a MW. However, the easterlies lasted for a single day only, though relatively diminished amplitudes of westerlies were present afterwards in January and mid/late February.

[12] To scale the intensity of the warmings, we now derive various fluxes and wave amplitudes (the four bottom panels). As depicted in the figure, large heat and momentum fluxes are found in the MW period. The heat flux follows the temperature distributions of the winter, as expected, and shows large fluxes of about 380 K m/s in late December, mid and late January in accordance with the increase in temperature. The EP flux divergence shows strikingly higher values of around -65×10^{-5} kg/s^2 during the warming periods, indicating the source of profound wave activities. The waves extracted from geopotential fields also show the presence of wave 1 before and during the MW, and both wave 1 and 2

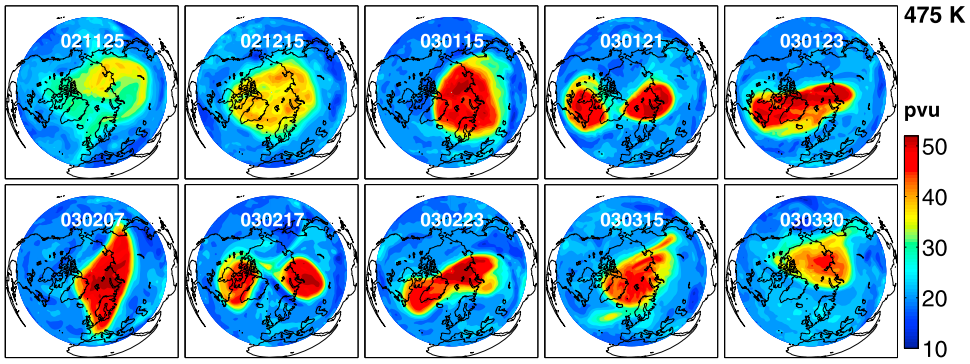

Figure 2. Temporal evolution of the polar vortex in the Arctic winter 2002/2003 at 475 K. The dates are selected by analyzing the complete record of the winter from November through the end of March.

after the MW with comparatively large amplitudes of around 90 m^2 s^2. The evolution of planetary waves and their estimated amplitudes are in very good agreement with those of *Günther et al.* [2008], though the scale of the amplitudes is different. The EP flux calculated for the waves exhibits very large values of 1.7×10^5 kg/s^2 for wave 1 in late December and mid-January, just before the MW. The momentum flux and wave EP flux also show an advanced shift in time with the warming periods, indicating considerable wave forcing prior to the MW. This is also manifested with the wave 1 amplitude, where a peak amplitude of 150 m^2 s^2 was estimated a few days before the MW. Though wave 1 amplitudes are small, wave 2 amplitudes are larger in late January and that triggered the minor warming in late January, just after the MW. It is interesting to note that the wave 2 EP flux during this minor warming is equal to or higher than that found for wave 1 during the MW. However, it is clear that the wave 1 amplification led to the MW in mid-January and the minor warmings in mid-February and early March.

[13] We now analyze how this particular meteorological situation affected the temporal evolution of the polar vortex in the lower stratosphere as our goal is to calculate the ozone loss inside the vortex. Figure 2 shows the PV maps at 475 K constructed from the ECMWF data for selected days of the winter. It shows that the vortex has already formed in November and strengthened by December, consistent with the very low temperatures. A strong concentric vortex was found in early January and it slightly elongated by 10 January 2003 as the wave 1 got amplified. Subsequently, the vortex split on 20 January 2003, just after the MW with the reversal of westerlies. Since the easterlies were not strong and did not prevail more than a day, the warming did not dissipate the vortex. *Kleinböhl et al.* [2005] also report that although there was rapid meridional transport of tropical air into the Arctic during this period, the low latitude air did not mix with the vortex air. So the vortex merged again by early February as the winds were westerlies and temperatures were cold. Nevertheless, the lower stratospheric vortex split again in mid-February due to a wave 1 event. The separated vortices, however, joined again to form a pole centered strong vortex by early March and sustained intact until the end of March. These results are also consistent with the discussion of polar vortex presented by *Günther et al.* [2008].

4.2. Meteorology During the ASUR and POAM Sampling

[14] The ASUR observations of the polar vortex in 2002/2003 are limited to 16 days with about 180 ozone profile measurements. Though measurements are sampled between 50°N and 79°N (65°–90°N EqL) these are mostly around a few longitudes (60°E–60°W). Therefore, the general evolution of the polar processes and vortex situations, that was discussed in the previous section, might not be applicable for all days of the ASUR flights. So in this section we discuss the situation of the polar vortex during the specific days of ASUR sampling. The ASUR measurements started on 14 January 2003, on which the vortex and the cold pool was on the same axis of the flight (similar to the one shown for 15 January 2003 in Figure 2, but tilted to the left) and therefore, a large number of measurements were performed in the vortex. However, the outside edge of the vortex was sampled on the following day, 15 January 2003, and thus, no measurement was found inside the vortex. The vortex disturbances started by 17 January 2003 and the vortex modified like a dumb-bell in the east–west direction (similar to the one shown in Figure 2 for 21 January 2003), and therefore, only a part of the vortex was sampled on 19 January 2003. The warming intensified and the vortex moved up toward the pole and merged afterwards, and hence, the measurements in 23–26 January 2003 sampled the vortex air near to the center of the pole only. The vortex became cold and near-concentric again and began to stabilize, as depicted in Figure 2 for 3 February 2003. Thus, the measurements taken during 7–9 February 2003 were mostly inside the vortex. The vortex shrunk by 12 February 2003 due to severe warming and therefore, no measurement was found inside the vortex during the transit flights on 12 February and 10 March 2003.

[15] Even if the vortex split on 17 February 2003, it had strengthened again by merging its parts during the second leg of the flights (SCIA-VALUE 2003). So relatively large, strong, and concentric vortices were observed for the following days of flights. Therefore, a majority of the measurements were inside the vortex in 13–19 March 2003, except for the longitudinal flight on 14 March, where no vortex sampling was found at 475 K, though some measurements were found inside the vortex at higher altitudes. This indicates that the vortex was tilted up, and was not

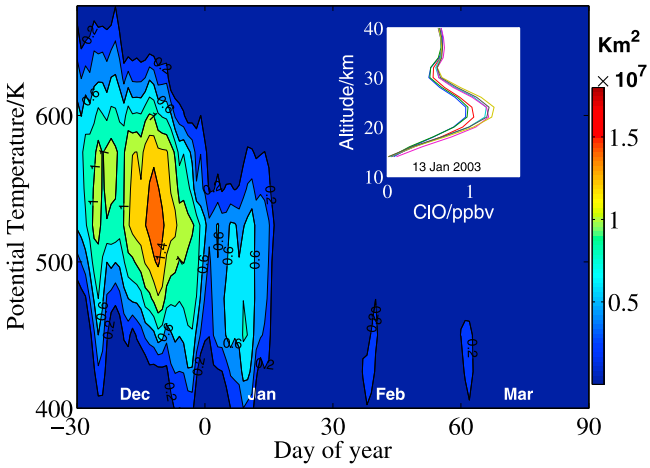

Figure 3. Areas of PSCs as defined by the area below NAT temperatures, which were calculated by the method of *Hanson and Mauersberger* [1988], using ECMWF meteorological analyses with 4.5 ppmv of H_2O and climatological profiles of HNO_3 for the Arctic winter 2002/2003. Inset: The ASUR ClO measurements taken inside the vortex [*Nash et al.*, 1996] with solar zenith angle <89° on 13 January 2003. The ClO measurements were separated by a few minutes as illustrated with different colors. Though ASUR has ClO measurements on other flight days, profiles with the highest ClO VMRs are displayed here.

symmetric with altitude. For instance at 450–550 K, there were 13 days (out of 16) of vortex sampling with more than 100 (out of ~180) ozone measurements.

[16] The POAM measurements, however, have global coverage between 51°N and 71°N (65°–90°N EqL) and therefore, the general meteorological situation described (Section 4.1) is fairly applicable to those observations. Despite limited to 71°N, as far as the nature of the winter and the Arctic vortex are concerned, this sampling pattern is sufficient to make a reasonable analysis of ozone loss, as the vortex was often displaced to midlatitude regions due to the frequent warmings. Consequently, a large number of ozone measurements were found inside the vortex, i.e., around 550 out of ~1500 measurements. The sampling pattern of these vortex observations can be found in Figure 3 of *Singleton et al.* [2005]. Further, these are also the best vortex-sampled satellite measurements available for this winter as compared to other satellite observations.

4.3. Potential PSC Areas and Chlorine Activation

[17] In general, low temperatures initiate the formation of PSCs, on which chlorine is activated to deplete ozone in the lower stratosphere [*WMO*, 2007]. So we now look at the distribution of PSCs in the Arctic winter 2002/2003. In this study, the area of PSCs (A_{PSC}) is defined as the area characterized by temperatures less than the NAT formation temperature, T_{NAT}. The T_{NAT} calculation is performed by using the formula of *Hanson and Mauersberger* [1988], for which the temperature and pressure data are taken from ECMWF operational analyses, with a constant value of 4.5 ppmv of H_2O and a HNO_3 climatology for the Arctic winter stratosphere [*Kleinböhl et al.*, 2002]. The resulting calculation is displayed in Figure 3. Nevertheless, as emphasized earlier, recent studies indicate that the PSCs may not always present at T_{NAT} in the Arctic [*Pitts et al.*, 2007; *WMO*, 2011]. In order to compare and to be consistent with other works for this winter, we use the above mentioned PSC calculation and hence, care must be taken when these PSC calculations are compared to other studies.

[18] In line with colder temperatures, large areas of PSCs are found in December and January. The maximum areas of these PSCs are found in the second half of December with values of 1.4–1.7 × 10^7 km^2. The area of PSCs cover a large vertical extent of 450–625 K. As the temperatures began to increase by early January, the A_{PSC} is reduced considerably and shrunk to a small area at 450–525 K with peak values of about 0.7 × 10^7 km^2. After the MW, small areas of occasional PSCs are found in mid-February and early March for a few days, below 475 K.

[19] The ASUR ClO mixing ratio profiles observed inside the vortex (with solar zenith angle <89°) on 13 January 2003 are presented in the inset of Figure 3. The measurements with activated ClO profiles are shown here. In agreement with the PSC calculations, the ASUR ClO measurements show activated chlorine of about 1.3 ppbv at around 22 km. These results are in good accordance with the findings of *Tilmes et al.* [2003] and *Urban et al.* [2004], who report high chlorine activation in early and mid-January in tune with large areas of PSC in the early winter. Further, *Tripathi et al.* [2006] also find a similar amount of ClO, about 1.2 ppbv at around 450 K, in the HALOX measurements [*von Hobe et al.*, 2005; *Günther et al.*, 2008] and Mimosa-Chim simulations on 15 January 2003.

4.4. Ozone and Ozone Loss

[20] To derive ozone loss from the ASUR measurements, the passive tracer technique is applied [e.g., *Kuttippurath et al.*, 2009]. This method uses passive ozone tracer calculations from a CTM and then the loss is computed as tracer minus (measured) ozone. The method assumes that there has been no ozone loss until the initial day. If there is some loss on the initial day, it will be propagated and hence, it should be corrected with respect to ozone measurements. As the ASUR measurements are discontinuous and start in early January, we use the POAM ozone observations to correct the initialization error and to get the complete evolution of the ozone and ozone loss from November through the end of March. This gives an opportunity to compare ozone and ozone loss from ASUR with those of POAM and the model. Our analysis with ASUR concentrates on the lower stratospheric isentropes of 450, 475, 500 and 550 K, where most of the loss happens in the majority of the Arctic winters [*Rex et al.*, 2004; *Kuttippurath et al.*, 2010]. The model ozone and tracer profiles are interpolated to the ASUR and POAM measurement locations. The comparisons are performed for each profile measurement and then averaged for each day if the measurements are inside the vortex. The vortex edge is taken as the maximum PV gradient as described by *Nash et al.* [1996]. In order to compare the ASUR and model ozone, and to compute ozone loss from the ASUR measurements, the model ozone and tracer profiles are convolved with ASUR ozone averaging kernels to account for the lower vertical resolution of the ASUR measurements [*Kuttippurath et al.*, 2007].

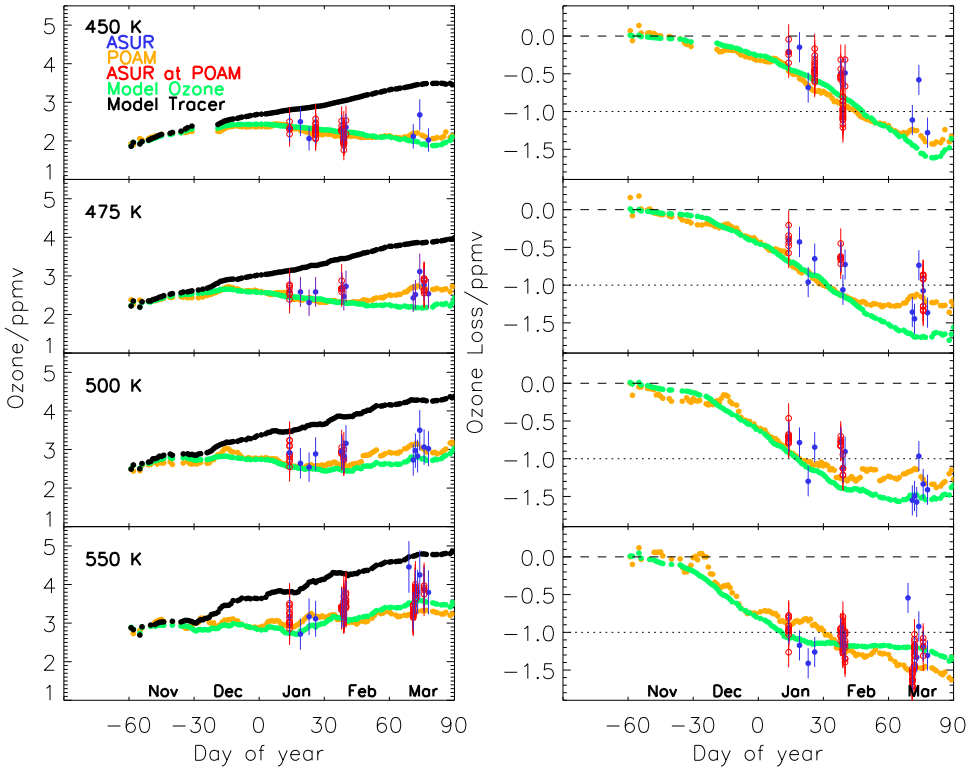

Figure 4. (left) Vortex averaged ozone and (right) ozone loss from ASUR compared to that of POAM and Mimosa-Chim CTM for the Arctic winter 2002/2003. The ozone loss is computed as ASUR ozone-tracer at ASUR locations, POAM ozone-tracer at POAM locations, and modeled ozone-tracer at POAM overpass for each measurement inside the vortex, and then averaged for each day. The ASUR ozone observations sampled near to POAM measurements, within 200 km, and the corresponding ozone loss estimated using the model tracer are also shown (red points: ASUR at POAM). The model ozone and tracer shown are the interpolated data at the POAM overpass locations. The horizontal dotted lines in the right panel represent 1 ppmv of ozone loss. The data shown are smoothed for 7 days, except for ASUR.

[21] Figure 4 (left) illustrates the distribution of ozone from ASUR, POAM and the model, and the ozone loss computed from the ASUR, POAM and Mimosa-Chim data at 450–550 K for the Arctic winter 2002/2003. The ASUR ozone and ozone depletion at POAM overpass points, within 200 km, are also shown for comparison. However, since both ASUR data sets (all ASUR measurements and ASUR at POAM overpass measurements) show very similar values for ozone, the ASUR observations irrespective of POAM overpass (shown in blue) are discussed throughout this study. Instead of average values, all individual ASUR measurements close to the POAM locations inside the vortex are shown here to demonstrate the spread of ASUR measurements around the POAM data. This also illustrates the inhomogeneity of the ozone distribution inside the vortex.

[22] The ozone data from ASUR and POAM and the model agree quite well at all altitudes in November through mid-February. However, the model underestimates the measured ozone by about 0.5 ± 0.2 ppmv in March at 475 K and 500 K. Furthermore, the model overestimates the POAM and underestimates ASUR ozone by about 0.3 ± 0.2 ppmv in March at 550 K.

[23] Figure 4 (right) delineates the ASUR, POAM and Mimosa-Chim ozone loss at various lower stratospheric altitudes for the Arctic winter 2002/2003. The ASUR ozone loss shows 0.7 ± 0.2, 1 ± 0.2, 1.2 ± 0.2, and 0.9 ± 0.2 ppmv at 450, 475, 500 and 550 K, respectively, by late January. *Tripathi et al.* [2006] and *Streibel et al.* [2006] report correspondingly large ozone loss rates of around 4 ppbv/sh (parts per billion in volume/sunlit hour) and 6 ppbv/sh at 475 K and 500 K, respectively, by the end of January. This much ozone loss in mid-winter is uncommon in the Arctic [*Newman et al.*, 2002; *Goutail et al.*, 2005; *Kuttippurath et al.*, 2010]. The loss rates are also markedly higher than that found in other Arctic winters during this period of the winter [e.g., *Rex et al.*, 2004; *Kuttippurath et al.*, 2010]. The large areas of PSC occurrence and high chlorine activation at those parts of the vortex displaced into sunlight triggered this unusual ozone depletion. The ozone loss continued to occur at 450 K and reached its maximum of 1.3 ± 0.2 ppmv by late March, in conjunction with cold temperatures found at this level, as shown in Figure 3. However, further loss in ozone was mitigated by relatively higher temperatures and the absence of PSCs at higher altitudes. Therefore, the maximum depletion was limited to 1.4–1.5 ± 0.2 ppmv at 475–550 K. This excludes a single day measurement that showed a loss of about 1.6 ± 0.2 ppmv by late January at 550 K.

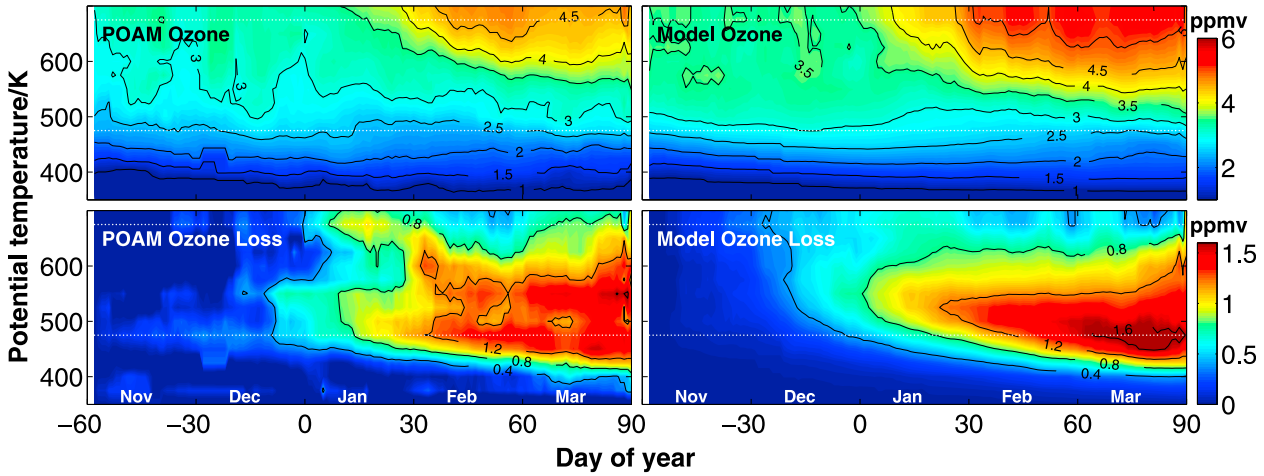

Figure 5. Temporal evolution of the vertical distribution of ozone and ozone loss in POAM and Mimosa-Chim CTM for the Arctic winter 2002/2003. The loss is estimated inside the vortex using the *Nash et al.* [1996] criterion for each altitude and the data are smoothed for 7 days. The dotted horizontal lines represent 475 K and 675 K.

[24] The POAM observations find almost the same value of ozone loss for the ASUR measurement days at 450–500 K. In agreement with the lower ozone, the ozone loss is slightly higher in POAM at 550 K in March. The ASUR ozone loss is in good accordance with modeled loss for the respective days, except for March at 475 K and 500 K, where the model overestimates both the ASUR and POAM ozone loss. POAM and Mimosa-Chim, however, find slightly higher depletion at some altitudes as the analysis extends to the end of March. Therefore, the ozone loss reaches 1.5 ± 0.2 ppmv at 550 K in POAM and 1.6/1.5 ± 0.2 ppmv at 475/550 K in Mimosa-Chim by the end of March, which are still in the error bounds of ASUR ozone, even though the model underestimates the measured ozone at 475 K. In general, the estimated ozone loss from ASUR is in good agreement with that of POAM and Mimosa-Chim.

[25] It has to be noted that the ozone loss in December is unusually large as it scales about 0.5 ± 0.2 ppmv at 450–550 K in both POAM and Mimosa-Chim. *Feng et al.* [2005] report high chlorine activation down to 400 K in early December. Further, as shown by the PV maps the vortex was very cold and elongated, which allowed vortex air to be frequently exposed to sunlight, and eventually caused ozone depletion in December. *Grooß et al.* [2005] too observe that the vortex spent more time at the sunlit parts of the midlatitudes during this period compared to that of other Arctic winters. Therefore, the unusually cold temperatures initiated large areas of PSCs and subsequent chlorine activation, and the vortex excursions to sunlit parts of the midlatitudes led to the large ozone depletion in the early winter.

4.5. Vertical Distribution of Ozone and Ozone Loss

[26] Apart from the interannual variability of ozone loss and the difference in estimated ozone loss by various techniques, there is also a spread in the altitude of maximum loss estimated by different instruments/methods for the same winter, as discussed by *Kuttippurath et al.* [2010]. In this study we have seen that ASUR, POAM and Mimosa-Chim show similar altitudes of maximum ozone loss. In order to study the vertical distribution of ozone loss closely, we use POAM measurements as ASUR observations are rather sporadic.

[27] Figure 5 illustrates the resulting ozone (upper panel) and ozone loss (Figure 5, bottom) evolution during the winter with respect to potential temperature. The model results are interpolated to each POAM overpass measurement and are sorted inside the vortex with respect to each altitude. The ozone measurements show relatively small values (less than 2 ppmv) in the lower stratosphere in November–January. After the vortex split during the MW, the split vortices in the midlatitudes constituted little higher ozone in February–March for the same altitudes. Nevertheless, these polar processes did not affect the vortex chemistry greatly as emphasized earlier and hence, the change in ozone is not very large. The simulations show comparable values below 600 K, where the deviations are mostly within ±0.2 ppmv, but slightly larger above.

[28] Regarding the ozone loss, POAM and Mimosa-Chim follow similar timing and vertical extent in its distribution. The depletion started by early December and intensified with the presence of large areas of PSCs by January around 475 K. As stated previously, the high ozone loss (0.7 ± 0.2 ppmv) in the early winter is unusual compared to the previous [*Goutail et al.*, 2005] and following winters [*Kuttippurath et al.*, 2010], and this makes the winter very distinct. The ozone loss in January–March is vertically spread at 425–650 K, and both POAM and Mimosa-Chim show the peak loss in late March. This highly vertically spread ozone loss coincides with the timing and location of the area of PSCs and chlorine activation. Studies with Mimosa-Chim [*Tripathi et al.*, 2006] have already shown that there was significant denitrification in the vortex in 450–650 K in this winter. Since denitrification enhances accumulated ozone loss by removing HNO_3, which otherwise deactivates ClO into its reservoirs [*Waibel et al.*, 1999], the contribution of denitrification to the additional ozone loss was estimated to be about 11–17% around 475 K in this winter [*Tripathi et al.*, 2006; *Grooß et al.*, 2005]. It indicates that, in addition to the chlorine activation, substantial denitrification is also responsible for this large ozone loss.

[29] Some significant ozone loss is also observed at higher altitudes, especially above 600 K in January. This is due to the NO_x catalyzed ozone destruction [*Grooß et al.*, 2005; *Kuttippurath et al.*, 2010], as large NO_x-rich air descended into the polar vortex from the mesosphere [*Konopka et al.*, 2007] and subtropical air was transported to the Arctic [*Kleinböhl et al.*, 2005] during the period. Because, in the PSC free stratosphere in 600–900 K, most ozone loss occurs through the NO_x catalytic cycle with a rate limiting step between NO_2 and O [*Kuttippurath et al.*, 2010]. The box model calculations of *Konopka et al.* [2007] also confirm that ~76% of ozone depletion at this altitude range was contributed by the NO_x cycle in this winter. This observed ozone loss in POAM is reasonably represented in the Mimosa-Chim simulations. Apparently all data sets show the maximum loss of 1.3–1.5 ± 0.2 ppmv in the 450–500 K altitude range. A recent study by *Kuttippurath et al.* [2010] notes that the altitude of maximum ozone depletion of Arctic warm winters is slightly higher than that for cold winters. This feature is found in this warm winter too, which is also reported by *Tripathi et al.* [2006] in comparison with the cold Arctic winter of 1999/2000.

4.6. Ozone Partial Column Loss

[30] In order to get a comprehensive overview of ozone loss in the winter, we now compute the partial ozone column loss from the measurements and simulations inside the vortex using the passive method. As most ozone loss occurs in the lower stratosphere, and to compare with other loss estimations, we have computed ozone loss in the vertical column range of 400–550 K. The partial ozone column calculated from the available ASUR measurements shows 61 ± 4 DU at 400–550 K in late March. The maximum loss estimated from the POAM measurements shows 63 ± 4 DU, and the modeled depletion at the POAM overpass points shows 65 ± 4 DU at the aforesaid column range for the same period. All data show a similar evolution of column loss, such that they exhibit a loss of around 12 ± 1 DU in December, 20–30 ± 2 DU in January, 30–50 ± 3 DU in February, and 50–65 ± 4 DU in March at 400–550 K. These exclude a single day POAM measurement that shows about 71 ± 4 DU of ozone loss in mid-March in the same altitude range. The large loss in December, as discussed in Section 4.4, is also shown by the column values. Consistent with the good agreement in ozone and ozone loss comparison in VMRs, the ASUR and POAM measurements show similar partial column loss. The slight difference between the ozone measurements is also reflected in their column ozone loss computations, but are still within the error bars. Even if there are some differences in sampling patterns of both instruments, the sampled vortex air between 50°N and 75°N shows similar loss. The modeled ozone loss at the observed points is in excellent agreement with the estimated column loss from the respective measurements.

4.7. Uncertainty in the Estimated Ozone Loss

[31] In this section we discuss the possible error sources in the estimated ozone loss. The first and foremost factor that can significantly affect the computation is the initialization of the model runs for the tracer calculations. The model should be initialized with respect to the status of vortex in the early winter, i.e., in order to catch the early ozone loss as in the case of this winter, the model run has to be initialized sufficiently early. Additionally, the passive method relies on the assumption that the ozone loss until the initial day is zero. So if there is an offset between the measured ozone and modeled tracer, the tracer/model ozone should be corrected with respect to the measured ozone. Otherwise, the ozone loss offset will be propagated and the derived loss will be corrupted. Another important factor to be considered is the proper selection of a vortex edge, as it is necessary to isolate the vortex from midlatitude air. In this study we have tested three different criteria (above 65°N EqL, the *Nash et al.* [1996] edge and poleward criteria), which all yield very similar results (not shown). The accuracy of the measurements (about 5–12%) is also to be accounted for. Therefore, to compute the uncertainty of the estimated ozone loss, we consider all the above mentioned parameters, i.e., the accuracies of the ASUR and POAM, the mean difference between model and ASUR/POAM ozone, the average difference among the ozone loss computed by different vortex criteria, and the initial offset in ozone loss that used to correct the inferred ozone loss, and take root square sum (RSS) of them. The RSS of these quantities shows about 0.17 ppmv or 3.5–5.1% at the studied altitudes, and is considered as the accuracy of the loss estimated from ASUR, POAM and the model.

5. Comparison With Other Estimations

[32] There are several published results available for comparison with the ozone loss estimated in this study, which are listed in Table 2. The ASUR ozone loss of 1.3 ± 0.2 ppmv at 400–500 K by late March is in good agreement with that of *Singleton et al.* [2005], *Christensen et al.* [2005], *Grooß et al.* [2005] and *Tilmes et al.* [2003], as they show the maximum loss within 1.2–1.5 ppmv between 400 K and 450 K. Further, as found with the ASUR observations, *El Amraoui et al.* [2008] also estimate the same ozone loss of 1.1 ± 0.2 ppmv by mid-January at 475 K from the measurements of the Sub-millimeter Radiometer (SMR) on the Odin satellite. The loss estimated, 1.6 ± 0.2 ppmv at 435 K by mid-March, from ozonesonde measurements by the Match method [*Streibel et al.*, 2006] is also close to the ASUR ozone loss. The ozone depletion deduced from SCIAMACHY measurements, i.e., 0.7 ppmv at 425–475 K in late March, shows the lowest loss among the various estimations for this winter. This can be due to the sampling limitation of the sensor, as it cannot observe high latitudes in early winter [*Sonkaew et al.*, 2011]. As expected, the loss determined from local measurements – Kiruna and Kola peninsula – [*Raffalski et al.*, 2005; *Ryskin and Kulikov*, 2008] departs slightly from the ASUR estimations because of the uneven or undersampling of the vortex over a single station, which essentially delineates the developments of the polar vortex over the stations. Since the POAM and Mimosa-Chim ozone losses are similar to that of ASUR, the above mentioned comparisons hold good for POAM and Mimosa-Chim as well. Nevertheless, it has to be borne in mind that the model underestimates the measured ozone at 450–500 K and overestimates at 550 K. In agreement with the ASUR ozone loss, the maximum loss is estimated in the lower stratosphere by other methods too, with slight differences in the peak ozone loss altitudes (±25 K). Such discrepancies in the maximum

Table 2. Vortex Averaged Ozone Loss Estimated (By Late March) From the ASUR, POAM and Mimosa-Chim Ozone Data Compared to Different Studies for the Arctic Winter 2002/2003[a]

	Ozone Loss in VMR				
Study	Method[a]	Loss (ppmv)	Peak Altitude	Period	Data
This study	PS	1.3–1.5 ± 0.2	450–475 K	Jan–Mar	ASUR
This study	PS	1.3–1.5 ± 0.2	500–550 K	Nov–Mar	POAM
Streibel et al. [2006]	Match	1.6 ± 0.2	407 K	Dec–15 Mar	Match
Singleton et al. [2005]	PS	1.2 ± 0.3	435 K	Dec–Mar	POAM
Christensen et al. [2005]	VAO	1.3 ± 0.1	435 K	10 Dec–10 Mar	ozonesondes
Tilmes et al. [2003]	TC	1.5 ± 0.0	440 K	16 Dec–Feb	HALOE
Ryskin and Kulikov [2008]	VAO	1.86 ± 0.33	530 K	Dec–5 Mar	MWR[b]
El Amraoui et al. [2008]	VAO	1.1 ± 0.2	25 ppbv/N_2O	15 Nov–15 Jan	SMR[d]
Raffalski et al. [2005]	VAO	1.1 ± 0.1	150 ppbv/N_2O	mid/Dec–mid/Feb	MWR[b,d]
Sonkaew et al. [2011]	VAO	0.7 ± 0.3	450–475 K	Dec–Mar	SCIAMACHY
Singleton et al. [2005]	PS	1.2 ± 0.3	425–450 K	Dec–15 Mar	SLIMCAT[c]
Grooß et al. [2005]	PS	1.3 ± 0.1	460 K	Dec–Mar	CLaMS[c]
Tripathi et al. [2006]	PS	1.3–1.5 ± 0.2	450–475 K	Nov–Mar	Mimosa-Chim[c]
This study	PS	1.3–1.5 ± 0.2	450–475 K	Nov–Mar	Mimosa-Chim @ POAM[c]
	Ozone Loss in Column				
Study	Method[a]	Loss (DU)	Peak Altitude	Period	Data
This study	PS	61 ± 4	400–550 K	Jan–Mar	ASUR
This study	PS	63 ± 4	400–550 K	Nov-Mar	POAM
Streibel et al. [2006]	Match	56 ± 4	407–501 K	Dec–15 Mar	Match
Christensen et al. [2005]	VAO	68 ± 7	380–525 K	10 Dec–10 Mar	ozonesondes
Tilmes et al. [2003]	TC	48 ± 4	416–510 K	16 Dec–Feb	HALOE
Müller et al. [2007]	TC	43–47 ± 6	380–550 K	Dec–22 Feb	HALOE/ILAS
Goutail et al. [2005]	PS	90 ± 5	Total Col.	Dec–10 Mar	SAOZ[e]
Grooß et al. [2005]	PS	46	380–550 K	Dec–15 Mar	CLaMS[c]
Feng et al. [2005]	PS	65	345–670 K	Dec–Mar	SLIMCAT[c]
Tripathi et al. [2006]	PS	63 ± 4	400–550 K	Nov–Mar	Mimosa-Chim[c]
This study	PS	65 ± 4	400–550 K	Nov–Mar	Mimosa-Chim @ POAM[c]

[a]The passive tracer method is denoted by PS, tracer correlation method is marked by TC, and the vortex averaged/profile descent method is denoted by VAO.
[b]The ozone loss analyses based on station measurements (for, e.g., Kiruna).
[c]Model simulations.
[d]Analyses based on N_2O levels instead of altitude.
[e]Analyses with total column measurements.

ozone loss altitudes were also reported by *Grooß et al.* [2005] for this winter and *Kuttippurath et al.* [2010] for 2004/2005. It is interesting to note that the POAM measurements with the SLIMCAT passive tracer also infer a similar ozone depletion of 1.2–1.5 ± 0.3 ppmv by late March at 425–450 K [*Singleton et al.*, 2005], as found in this study. These results corroborate the strength and consistency of the loss computation method and POAM data.

[33] The ozone column loss computed from the ASUR observations is also generally in good agreement with that from other techniques. For instance, the column loss calculated using ozonesonde measurements in 400–550 K around mid-March by *Christensen et al.* [2005] is in excellent agreement with that estimated from ASUR, POAM and Mimosa-Chim as they show a loss of 68 ± 7 DU for the same period and altitude range. The ozone loss calculated by *Feng et al.* [2005] and *Tripathi et al.* [2006] too find a similar loss of 65–67 ± 4 DU in 400–550 K in late March. The loss estimate of *Müller et al.* [2007], 43–47 ± 6 DU in 400–500 K, from the Halogen Occultation Experiment (HALOE) and the Improved Limb Atmospheric Spectrometer (ILAS)–II satellite measurements is in excellent agreement with our estimate of 45 ± 3 DU in 400–500 K, around 22 February 2003. Their ozone loss estimate of 55 ± 6 DU in 380–550 K by 22 March 2003 is also close to our estimate for the same period, i.e., 60 ± 3 DU in 400–550 K. The ASUR/POAM estimation of 55 ± 3 DU in 400–500 K by late March is in very good agreement with that inferred from the Match method, about 56 ± 4 DU in 407–501 K in mid-March [*Streibel et al.*, 2006]. Nevertheless, the loss estimated by *Tilmes et al.* [2003], i.e., 48 ± 4 DU in 416–510 K in mid-April, is lower than the above mentioned loss estimates. The difference between our estimates in mid-March (55 ± 3 DU) and the estimates of *Grooß et al.* [2005] (about 46 DU) is also beyond the error bars of the compared data sets. In short, except these two loss computations [i.e., *Tilmes et al.*, 2003; *Grooß et al.*, 2005], all partial column estimates agree well within their error bars, and they show an average loss of 65 ± 5 DU in 400–550 K in the Arctic winter 2002/2003.

[34] When our partial column estimates are compared to the total column estimates of *Goutail et al.* [2005], i.e., 90 ± 5 DU, there is a difference of 25–30 DU. The total column loss equals to the loss usually estimated for a cold or moderately cold winter [*Kuttippurath et al.*, 2010; *Harris et al.*, 2010; *WMO*, 2011]. A recent study by *Kuttippurath et al.* [2010] reports an average difference of 19 ± 7 DU between the partial column loss calculated below and above 550 K. Apparently, this also demonstrates the difference between

the ozone loss contributed by halogens in the lower stratosphere and NO_x in the middle stratosphere. In the Arctic winter 2002/2003, the halogen dominated loss in the lower stratosphere is about 60–65 DU at 400–550 K. The difference between this partial column loss and the total column loss (i.e., 25–30 DU) is much larger than the expected average loss (19 ± 7 DU) above 550 K. This hints further at the special dynamics of the winter, as there was large mesospheric descent of NO_x-rich air masses and rapid meridional transport of subtropical air masses, which offered a conducive atmosphere for ozone depletion by the NO_x chemical cycle at higher altitudes. The study by *Konopka et al.* [2007] also confirms this ozone loss feature as they compute ~27–30 DU (or 76%) column loss by the NO_x cycle, from satellite measurements above 550 K. This column loss (27–30 DU) matches exactly the difference computed between the partial column loss from our study and total column loss from *Goutail et al.* [2005] (i.e., 25–30 DU), and thus, it affirms that the large loss above 550 K was due to the NO_x chemistry activated on a NO_x-rich air influx from the mesosphere and sub-tropics. To check this additional loss above 550 K, we computed the loss at 350–950 K (to the topmost level of the model) from the POAM and Mimosa-Chim data and it yielded 86 ± 5 DU and 70 ± 4 DU, respectively. These calculations are very close to the total column estimate of 90 ± 5 DU by *Goutail et al.* [2005]. Since the model overestimates the POAM ozone above 550 K and there is no upper stratosphere and mesosphere in the model, the deficit in the simulated column loss is reasonably justified. This additional ozone loss above 550 K further manifests that the winter was very special in various aspects of stratospheric transport and chemistry.

6. Comparison With Other Arctic Winters

[35] As emphasized in the introduction, the Arctic winter stratosphere intermittently experiences major and minor warmings, which make large interannual variability in the Arctic ozone loss. Ozone loss estimation in the Arctic is available from various measurement sources for each winter since 1989 to compare with our calculations [*Hofmann et al.*, 1989; *Goutail et al.*, 2005; *Tilmes et al.*, 2006; *Harris et al.*, 2010; *Kuttippurath et al.*, 2010; *WMO*, 2011]. Among these winters, 1995, 1996, 2000, and 2005 were very cold [*WMO*, 2007] and hence, the total column loss calculated from ground-based total ozone measurements showed >80–90 DU [*Goutail et al.*, 2005; *Kuttippurath et al.*, 2010]. The winters 1992, 1994, 1997, 1998, 2007, and 2008 were moderately cold and thus, the total column loss was in an average scale of about 60 DU [*Andersen and Knudsen*, 2002; *Goutail et al.*, 2005; *Kuttippurath et al.*, 2010]. On the other hand, the winters 2001, 2004, 2006 and 2009 were subjected to minor and major warming events with subsequent break in the persistence of the polar vortex. Therefore, ozone depletion computed in these warm winters showed the lowest values of about 25–30 DU [*Goutail et al.*, 2005; *Kuttippurath et al.*, 2010]. In addition to the total column, similar amounts of ozone depletion were also estimated in the partial column range of 380–550 K from ozonesonde measurements for all winters [*Rex et al.*, 2004; *Harris et al.*, 2010] and hence, they also express analogous features of ozone loss. Although the loss estimated from satellite measurements [*Tilmes et al.*, 2006] show slightly lower values than those estimated from the ground-based/Match [*Goutail et al.*, 2005; *Rex et al.*, 2004] measurements for individual years, those analyses still show a clear difference between the loss derived in the warm and cold winters [*Andersen and Knudsen*, 2002], as discussed with the total column measurements.

[36] While comparing the ozone loss of 65 ± 5 DU in 400–550 K in this winter, as analyzed from the ASUR, POAM and Mimosa-Chim results together with other published works, with the loss estimated in the Arctic winters since 1989, the estimated loss in 2002/2003 is close to the estimates for the moderately cold winters. Further, in line with the column ozone loss, the loss found in mixing ratio also exposes a distinct difference between the range of depletions observed in the cold and warm winters, with a loss of ~1.8–2.1 ppmv in cold winters [*Rex et al.*, 2004; *Kuttippurath et al.*, 2010] and about 0.5–0.7 ppmv in warm winters [*Manney et al.*, 2003; *Rex et al.*, 2004; *Kuttippurath et al.*, 2010; *Sonkaew et al.*, 2011]. Therefore, the ozone depletion computed in 2002/2003, i.e., 1.5 ± 0.3 ppmv, stays between these cold and warm winter estimates. Apart from the significant ozone loss in December (0.5 ± 0.2 ppmv at 450–500 K or 12 ± 1 DU in 400–550 K), such a large scale ozone loss in a winter with three minor and a MW is exceptional, and is happened for the first time in the Arctic in 1989–2010, and this makes the Arctic winter 2002/2003 unique.

7. Conclusions

[37] The Arctic winter 2002/2003 was exceptional as it was characterized by an unusual cold spell in the first half and a MW in the second half. Therefore, large areas of PSCs are found at 450–625 K from December through mid-January. A wave 1 event led to the MW around 18 January 2003 and thus, the high temperatures inhibited the formation of PSCs afterwards. However, the easterlies did not prevail, though only diminished amplitudes of westerlies were present in the later part of the winter. Apart from the MW in mid-January, there were three minor warmings in mid-December, mid-February and early March. Though the vortex split during the MW in mid-January and during the minor warming in mid-February, it did not disappear until early April. Since the vortex split was confined mostly to the lower stratosphere, the MW can be classified as a vortex displacement event instead of a vortex split event.

[38] The ozone loss determined with the ASUR measurements taken during the EuPLEx and SCIA–VALUE 2003 airborne campaigns shows high values in the mid-winter. The ASUR measurements show the maximum ozone loss of 1.3 ± 0.2 ppmv at 450–500 K, from the available measurements until late March. The partial column loss calculated from ASUR observations in 400–550 K shows about 61 ± 4 DU in the same period. These ozone loss computations, both in mixing ratios and partial column, are in very good agreement with those estimated from POAM, Mimosa-Chim CTM and other available published results for this winter. The POAM/Mimosa-Chim loss amounts to 0.5 ± 0.2 ppmv at 450–550 K or 12 ± 1 DU in 400–550 K in December, which is uncharacteristically high during this period in the Arctic polar winter stratosphere. The uncommon ozone depletion in the early winter was due to very low temperatures, large areas of PSCs, significant vortex wide

denitrification and high chlorine activation, as the vortex moved to sunlit parts of the adjacent midlatitude regions.

[39] In this study we have presented both the dynamical processes during the minor and major warmings, and the chemical ozone loss in the Arctic winter 2002/2003. The ASUR measurements used for the diagnosis of ozone loss have not hitherto been used for the study of this winter. The heat flux, momentum flux, EP flux, EP flux divergence and PV maps were used for the description of the dynamical situation, which were not presented together to characterize this winter before. On average, in conjunction with all published results, this winter was inflicted with a maximum ozone loss of 1.5 ± 0.3 ppmv at 450–550 K or 65 ± 5 DU in 400–550 K by late March. Interestingly, these values inferred from a number of computations rightly coincide with those derived in our study (1.3 ± 0.2 ppmv or 63 ± 4 DU at the same altitude ranges). When compared to other Arctic winters, as analyzed from this work, this winter has a unique feature of three minor warmings, a MW and large ozone loss that usually observed in a moderately cold winter, in addition to its unusually large ozone depletion in December–January. Therefore, this study offers some interesting analyses for future studies of polar processing and ozone loss.

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References

Andersen, S. B., and B. M. Knudsen (2002), The influence of vortex ozone depletion on Arctic ozone trends, *Geophys. Res. Lett.*, *29*(21), 2013, doi:10.1029/2001GL014595.

Blumenstock, T., F. Hase, I. Kramer, S. Mikuteit, H. Fischer, F. Goutail, and U. Raffalski (2009), Winter to winter variability of chlorine activation and ozone loss as observed by ground-based FTIR measurements at Kiruna since winter 1993/94, *Int. J. Remote Sens.*, *30*(15), 4055–4064.

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 (2002), Ozone depletion observed by ASUR during the Arctic winter 1999/2000, *J. Geophys. Res.*, *107*(D20), 8277, doi:10.1029/2001JD000546.

Burkholder, J. B., J. J. Orlando, and C. J. Howard (1990), Ultraviolet absorption cross-sections of Cl_2 O_2 between 210 and 410 nm, *J. Phys. Chem.*, *94*, 687–695.

Carslaw, K., B. Luo, and T. Peter (1995), An analytic expression for the composition of aqueous HNO_3–H_2SO_4 stratospheric aerosols including gas phase removal of HNO_3, *Geophys. Res. Lett.*, *22*(14), 1877–1880, doi:10.1029/95GL01668.

Christensen, T., et al. (2005), Vortex-averaged Arctic ozone depletion in the winter 2002/2003, *Atmos. Chem. Phys.*, *5*, 131–138.

El Amraoui, L., V.-H. Peuch, P. Ricaud, S. Massart, N. Semane, H. Teyssèdre, D. Cariolle, and F. Karcher (2008), Ozone loss in the 2002–2003 Arctic vortex deduced from the assimilation of Odin/SMR O_3 and N_2O measurements: N_2O as a dynamical tracer, *Q. J. R. Meteorol. Soc.*, *134*(630), 217–228, doi:10.1002/qj.191.

Feng, W., et al. (2005), Three-dimensional model study of the arctic ozone loss in 2002/2003 and comparison with 1999/2000 and 2003/2004, *Atmos. Chem. Phys.*, *5*, 139–152.

Goutail, F., J.-P. Pommereau, F. Lefèvre, M. Van Roozendael, S. B. Andersen, B.-A. Kåstad Høiskar, V. Dorokhov, E. Kyrö, M. P. Chipperfield, and W. Feng (2005), Early unusual ozone loss during the Arctic winter 2002/2003 compared to other winters, *Atmos. Chem. Phys.*, *5*, 665–677.

Grooß, J.-U., G. Günther, M. Müller, P. Konopka, S. Bausch, H. Schlager, C. Voigt, C. M. Volk, and G. C. Toon (2005), Simulation of denitrification and ozone loss for the Arctic winter 2002/2003, *Atmos. Chem. Phys.*, *5*, 1437–1448.

Günther, G., R. Müller, M. von Hobe, F. Stroh, P. Konopka, and C. M. Volk (2008), Quantification of transport across the boundary of the lower stratospheric vortex during Arctic winter 2002/2003, *Atmos. Chem. Phys.*, *8*, 3655–3670.

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

Harris, N. R. P., M. Rex, F. Goutail, B. M. Knudsen, G. L. Manney, R. Müller, and P. von der Gathen (2002), Comparison of empirically derived ozone losses in the Arctic vortex, *J. Geophys. Res.*, *107*(D20), 8264, doi:10.1029/2001JD000482.

Harris, N. R. P., R. Lehmann, M. Rex, and P. von der Gathen (2010), A closer look at Arctic ozone loss and polar stratospheric clouds, *Atmos. Chem. Phys.*, *10*, 8499–8510, doi:10.5194/acp-10-8499-2010.

Hofmann, D. J., T. L. Deshler, P. Aimedieu, W. A. Matthews, P. V. Johnston, Y. Kondo, W. R. Sheldon, G. J. Byrne, and J. R. Benbrook (1989), Stratospheric clouds and ozone depletion in the Arctic during January 1989, *Nature*, *340*, 117–121.

Kleinböhl, A., et al. (2002), Vortexwide denitrification of the Arctic polar stratosphere in winter 1999/2000 determined by remote observations, *J. Geophys. Res.*, *107*, 8305, doi:10.1029/2001JD001042, [printed *108*(D5), 2003].

Kleinböhl, A., J. Kuttippurath, M. Sinnhuber, B.-M. Sinnhuber, H. Küllmann, K. Künzi, and J. Notholt (2005), Rapid meridional transport of tropical air masses to the Arctic during the major stratospheric warming in January 2003, *Atmos. Chem. Phys.*, *5*, 1291–1299, doi:10.5194/acp-5-1291-2005.

Konopka, P., et al. (2007), Ozone loss driven by nitrogen oxides and triggered by stratospheric warmings can outweigh the effect of halogens, *J. Geophys. Res.*, *112*, D05105, doi:10.1029/2006JD007064.

Kuttippurath, J., et al. (2007), Intercomparison of ozone profile measurements from ASUR, SCIAMACHY, MIPAS, OSIRIS, and SMR, *J. Geophys. Res.*, *112*, D09311, doi:10.1029/2006JD007830.

Kuttippurath, J., S. Godin-Beekmann, F. Lefèvre, and A. Pazmiño (2009), Ozone depletion in the Arctic winter 2007–2008, *Int. J. Remote Sens.*, *30*(15), 4071–4082, doi:10.1080/01431160902821965.

Kuttippurath, J., S. Godin-Beekmann, F. Lefèvre, and F. Goutail (2010), Spatial, temporal, and vertical variability of polar stratospheric ozone loss in the Arctic winters 2004/2005–2009/2010, *Atmos. Chem. Phys.*, *10*, 9915–9930, doi:10.5194/acp-10-9915-2010.

Lefèvre, F., F. Figarol, K. S. Carslaw, and T. Peter (1998), The 1997 Arctic ozone depletion quantified from three-dimensional model simulations, *Geophys. Res. Lett.*, *25*, 2425–2428.

Manney, G. L., L. Froidevaux, M. L. Santee, N. J. Livesey, J. L. Sabutis, and J. W. Waters (2003), Variability of ozone loss during Arctic winter (1991–2000) estimated from UARS Microwave Limb Sounder measurements, *J. Geophys. Res.*, *108*(D4), 4149, doi:10.1029/2002JD002634.

McInturff, R. M. (1978), Stratospheric warmings: Synoptic, dynamic and general-circulation aspects, *NASA Ref. Publ.*, *1017*, 50 pp.

Müller, R., et al. (2007), Impact of mesospheric intrusions on ozone–tracer relations in the stratospheric polar vortex, *J. Geophys. Res.*, *112*, D23307, doi:10.1029/2006JD008315.

Murray, F. W. (1967), On the computation of saturation vapour pressure, *J. Appl. Meterol.*, *6*, 203–204.

Nash, E. R., P. A. Newman, J. E. Rosenfield, and M. R. Schoeberl (1996), An objective determination of the polar vortex using Ertel's potential vorticity, *J. Geophys. Res.*, *101*, 9471–9478.

Newman, P. A., et al. (2002), An overview of the SOLVE/THESEO 2000 campaign, *J. Geophys. Res.*, *107*(D20), 8259, doi:10.1029/2001JD001303.

Papanastasiou, D. K., V. C. Papadimitriou, D. W. Fahey, and J. B. Burkholder (2009), UV absorption spectrum of the ClO dimer (Cl_2O_2) between 200 and 420 nm, *J. Phys. Chem. A*, *113*, 13,711–13,726.

Pitts, M. C., L. W. Thomason, L. R. Poole, and D. M. Winker (2007), Characterization of polar stratospheric clouds with spaceborne lidar: CALIPSO and the 2006 Antarctic season, *Atmos. Chem. Phys.*, *7*, 5207–5228, doi:10.5194/acp-7-5207-2007.

Portmann, R., S. Solomon, R. Garcia, L. Thomason, L. Poole, and M. McCormick (1996), Role of aerosol variations in anthropogenic ozone depletion in the polar regions, *J. Geophys. Res.*, *101*(D17), 22,991–23,006.

Pruppacher, H. R., and J. D. Klett (1997), *Microstructure of Atmospheric Clouds and Precipitations*, 2nd ed., Kluwer Acad., Norwell, Mass.

Raffalski, U., G. Hochschild, G. Kopp, and J. Urban (2005), Evolution of stratospheric ozone during winter 2002/2003 as observed by a

ground-based millimetre wave radiometer at Kiruna, Sweden, *Atmos. Chem. Phys.*, *5*, 1399–1407.

Randall, C. E., D. Rusch, R. Bevilacqua, K. W. Hoppel, and J. D. Lumpe (2003), Validation of POAM-3 O_3: Comparison to ozonesonde and satellite data, *J. Geophys. Res.*, *108*(D12), 4367, doi:10.1029/2002JD002944.

Rex, M., et al. (2002), Chemical depletion of Arctic ozone in winter 1999/2000, *J. Geophys. Res.*, *107*(D20), 8276, doi:10.1029/2001JD000533.

Rex, M., R. J. Salawitch, P. von der Gathen, N. R. P. Harris, M. P. Chipperfield, and B. Naujokat (2004), Arctic ozone loss and climate change, *Geophys. Res. Lett.*, *31*, L04116, doi:10.1029/2003GL018844.

Rodgers, C. D. (1976), Retrieval of atmospheric temperature and composition from remote measurements of thermal radiation, *Rev. Geophys.*, *14*, 609–624.

Ryskin, V. G., and Y. Y. Kulikov (2008), Evaluation of chemical ozone loss in the stratosphere over the Kola Peninsula in the 2002/2003 winter from microwave sounding data, *Izv. Atmos. Oceanic Phys.*, *44*, 199–205, doi:10.1134/S0001433808020060.

Sander, S. P., et al. (2003), Chemical kinetics and photochemical data for use in stratospheric modeling: Evaluation 13, *JPL Publ.*, *00-003*.

Shine, K. P. (1987), The middle atmosphere in the absence of dynamical heat fluxes, *Q. J. R. Meteorol. Soc.*, *113*, 603–633.

Singleton, C. S., C. E. Randall, M. P. Chipperfield, S. Davies, W. Feng, R. M. Bevilacqua, K. W. Hoppel, M. D. Fromm, G. L. Manney, and V. L. Harvey (2005), 2002–2003 Arctic ozone loss deduced from POAM III satellite observations and SLIMCAT chemical transport model, *Atmos. Chem. Phys.*, *5*, 597–609.

Sonkaew, T., C. von Savigny, K.-U. Eichmann, M. Weber, A. Rozanov, H. Bovensmann, and J. P. Burrows (2011), Chemical ozone loss in Arctic and Antarctic polar winter/spring season derived from SCIAMACHY limb measurements 2002–2009, *Atmos. Chem. Phys. Discuss.*, *11*, 6555–6599, doi:10.5194/acpd-11-6555-2011.

Stimpfle, R. M., D. M. Wilmouth, R. J. Salawitch, and J. G. Anderson (2004), First measurements of ClOOCl in the stratosphere: The coupling of ClOOCl and ClO in the Arctic polar vortex, *J. Geophys. Res.*, *109*, D03301, doi:10.1029/2003JD003811.

Streibel, M., et al. (2006), Chemical ozone loss in the Arctic winter 2002/2003 determined with Match, *Atmos. Chem. Phys.*, *6*, 2783–2792.

Tilmes, S., M. Müller, J.-U. Grooss, M. Höpfner, G. C. Toon, and J. M. Russel III (2003), Very early chlorine activation and ozone loss in the Arctic winter 2002–2003, *Geophys. Res. Lett.*, *30*(23), 2201, doi:10.1029/2003GL018079.

Tilmes, S., R. Müller, A. Engel, M. Rex, and J. M. Russell III (2006), Chemical ozone loss in the Arctic and Antarctic stratosphere between 1992 and 2005, *Geophys. Res. Lett.*, *33*, L20812, doi:10.1029/2006GL026925.

Tripathi, O. P., et al. (2006), High resolution simulation of recent Arctic and Antarctic stratospheric chemical ozone loss compared to observations, *J. Atmos. Chem.*, *55*, 205–226.

Tripathi, O. P., et al. (2007), Comparison of polar ozone loss rates simulated by one-dimensional and three-dimensional models with Match observations in recent Antarctic and Arctic winters, *J. Geophys. Res.*, *112*, D12307, doi:10.1029/2006JD008370.

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.

von Hobe, M., J.-U. Grooß, R. Müller, S. Hrechanyy, U. Winkler, and F. Stroh (2005), A re-evaluation of the ClO/Cl_2O_2 equilibrium constant based on stratospheric in-situ observations, *Atmos. Chem. Phys.*, *5*, 693–702.

Waibel, A. E., et al. (1999), Arctic ozone loss due to denitrification, *Science*, *283*, 2064–2069.

Wamsley, P. R., et al. (1998), Distribution of halon-1211 in the upper troposphere and lower stratosphere and the 1994 total bromine budget, *J. Geophys. Res.*, *103*, 1513–1526.

World Meteorological Organization (WMO) (2007), Scientific assessment of ozone depletion: 2006, *Global Ozone Monit. Res. Proj. Rep. 50*, 572 pp., Geneva, Switzerland.

World Meteorological Organization (WMO) (2011), Scientific assessment of ozone depletion: 2010, *Global Ozone Monit. Res. Proj. Rep. 52*, 516 pp., Geneva, Switzerland.

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