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

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ABSTRACT: In this paper we investigate the evolution of the northern polar vortex during the winter 2002–2003 in the lower stratosphere by using assimilated fields of ozone (O_3) and nitrous oxide (N_2O). Both O_3 and N_2O used in this study are obtained from the Sub-Millimetre Radiometer (SMR) aboard the Odin satellite and are assimilated into the global three-dimensional chemistry transport model of M´et´eo-France, MOCAGE. O_3 is assimilated into the 'full' model including both advection and chemistry whereas N_2O is only assimilated with advection since it is characterized by good chemical stability in the lower stratosphere. We show the ability of the assimilated N_2O field to localize the edge of the polar vortex. The results are compared to the use of the maximum gradient of modified potential vorticity as a vortex edge criterion. The O_3 assimilated field serves to evaluate the ozone evolution and to deduce the ozone depletion inside the vortex. The chemical ozone loss is estimated using the vortex-average technique. The N_2O assimilated field is also used to substract out the effect of subsidence in order to extract the actual chemical ozone loss. Results show that the chemical ozone loss is 1.1 ± 0.3 ppmv on the 25 ppbv N_2O level between mid-November and mid-January, and 0.9 ± 0.2 ppmv on the 50 ppbv N_2O level between mid-November and the end of January. A linear fit over the same periods gives a chemical ozone loss rate of $\sim$18 ppbv day^{-1} and $\sim$9.3 ppbv day^{-1} on the 25 ppbv and 50 ppbv N_2O levels, respectively. The vortex-averaged ozone loss profile from the O_3 assimilated field shows a maximum of 0.98 ppmv at 475 K. Comparisons to other results reported by different authors using different techniques and different observations give satisfactory results.

KEY WORDS chemical data assimilation; diabatic descent; chemical ozone loss

1. Introduction

The evolution of ozone in the Arctic vortex depends on both dynamical and chemical processes. The understanding of the ozone loss processes in the Northern Hemisphere (NH) stratospheric vortex is essential for a reliable prediction of the future evolution of the polar ozone layer (Streibel *et al.*, 2006). The determination of the chemical ozone loss over the Arctic in the stratosphere is highly difficult due to the dynamical variability caused by vertical and horizontal transport of air masses. Different approaches within many sets of observations have been proposed in order to remove the contribution of transport and thus to quantify the ozone loss in the winter stratospheric vortex. They have been described and compared by Harris *et al.* (2002). The 'tracer correlation technique' consists of removing the effect of transport by comparing the pre-winter and post-winter relations between ozone volume mixing ratios and an inert tracer (Singleton *et al.*, 2005). Proffitt *et al.* (1990) was the first to use *in situ* high-altitude aircraft measurements to deduce the ozone loss in the Arctic polar vortex by using this technique. The 'vortex-average technique' calculates the average of all vortex ozone data on an isentropic surface as a function of time (Grooss and Müller, 2003). It assumes that the dynamical contribution to ozone inside the vortex is dominated by diabatic descent. The 'Match' method is a pseudo-Lagrangian technique, which permits us to quantify the chemical ozone loss by measuring the difference in ozone in an air parcel sampled at different times (e.g. von der Gathen *et al.*, 1995; Rex *et al.*, 2002; Harris *et al.*, 2002). The 'passive tracer method' consists of comparing modelled ozone (run with and without chemistry) to observations. It has been widely used in order to quantify the Arctic ozone loss (Goutail *et al.*, 1999; Sinnhuber *et al.*, 2000; Hoppel *et al.*, 2002). Although this method is probably the

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most popular technique used to determine the ozone loss in the polar vortex, it can present some defects. Indeed, limitations arise from uncertainties in dynamical forcings and in resolved-scale representation of subgrid-scale transport and of homogenous and heterogenous chemistry, as well as in the vertical and horizontal resolution of the chemistry and transport model (CTM). Previous studies also indicate that current CTMs cannot give a satisfactory observed partial column ozone loss (Feng *et al*., 2005). Many models underestimate ozone loss in cold winters when compared to ozone loss inferred from observations (e.g. Goutail *et al*., 2005).

Data assimilation consists of combining in an optimal way observations provided by instruments with an *a priori* knowledge about a physical system such as a model output. It has the advantage that observational and model errors are accounted for and can be verified *a posteriori* by considering e.g. observation minus forecast (OMF) statistics or a χ^2 test (e.g. El Amraoui *et al*., 2004). Thus, it is a powerful tool for improving model outputs by constraining the *a priori* knowledge of the model with the observations in order to better estimate the state of the atmosphere.

Data assimilation has proven to be useful in various fields of applications and was successfully applied for studies of atmospheric chemistry and trace gas distribution. Siegmund *et al*. (2005) demonstrated the performance of the assimilation to study the ozone budgets related to transport and chemistry in the Antarctic region during the stratospheric warming of 2002. A notable example of the power of data assimilation is the successful prediction of the split Antarctic ozone hole in September 2002 by Eskes *et al*. (2003) using a tracer transport and assimilation model. Geer *et al*. (2006) have assessed the assimilation of ozone data using various technique and models in the Assimilation of ENVISAT Data (ASSET) ozone intercomparison project. They validate their analysis by comparing them with independent data from ozonesondes and HALOE (the Halogen Occultation Experiment on the Upper Atmosphere Research Satellite), and also with the assimilated fields of observations from the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) aboard the ENVISAT satellite. Recently, Bencherif *et al*. (2007) demonstrated the ability of Odin/SMR N_2O assimilated fields to describe the tropic−midlatitude exchanges during the 2002 major warming in the Southern Hemisphere.

This paper aims to:

- show the capability of assimilated fields to describe the large-scale evolution of the NH stratospheric vortex during the 2002−2003 winter, and
- evaluate the vortex-average technique with N_2O as a dynamical tracer.

In this paper, we assimilate O_3 and N_2O measurements from the Odin/SMR instrument in order to describe the evolution of the NH stratospheric vortex during 2002−2003 Arctic winter. The N_2O assimilated fields are used as a dynamical tracer in order to determine the edge of the vortex using their maximum gradient following the method suggested by Nash *et al*. (1996). They are also used to quantify the diabatic descent inside the vortex. The O_3 assimilated fields serve to evaluate the ozone evolution as well as the chemical ozone loss inside the vortex.

The outline is as follows. Section 2 presents the measurements of O_3 and N_2O as well as the assimilation system used in this study. In Section 3, we present the meteorological situation of the 2002−2003 Arctic vortex. Main results including the validation of assimilated fields, the diabatic descent, the ozone evolution as well as the chemical ozone depletion inside the vortex are presented in Section 4. Some comparisons with other results are done in Section 5. Conclusions are given in Section 6.

2. Data analysis

2.1. O_3 and N_2O measurements

The Odin satellite was launched on 20 February 2001 into a polar sun-synchronous circular orbit with the ascending node at 18 hours and an inclination of about 97.8°. The satellite makes about 14 orbits per day and provides a global coverage in the sense that all longitudinal ranges are sampled. Each orbit contains about 60 limb profiles of each measured species. Measurements are made in the plane of the orbit covering the latitude range from 83 °S to 83 °N. Odin has the unique mission to make both astronomy and aeronomy measurements. It includes two instruments: the Optical Spectrograph and Infrared Imager System (OSIRIS) and the Sub-Millimetre Radiometer (SMR). For the aeronomy part of the Odin mission, SMR in the main stratospheric mode is able to scan the limb of the atmosphere from 7 to 70 km with a vertical resolution of about 1.5 km. Both O_3 and N_2O used in this study are retrieved in the 501.8 GHz band. Typically N_2O is retrieved in the stratosphere above 15 km with a single-scan precision of the order of 5% (10−20 ppbv), and O_3 between ∼19 and ∼50 km with a single-scan precision of about 25% (0.5−1.5 ppmv) (Urban *et al*., 2005).

2.2. Assimilation system

The assimilation system used in this study is MOCAGE−PALM developed jointly between Météo-France and CERFACS (Centre Européen de Recherche et de Formation Avancée en Calcul Scientifique) in the framework of the ASSET European project (Geer *et al*., 2006).

MOCAGE (Modèle de Chimie Atmosphérique à Grande Echelle; Peuch *et al*., 1999) is a three-dimensional CTM of the troposphere and stratosphere forced by external wind and temperature fields from the ARPEGE model, the operational meteorological model of Météo-France (Courtier *et al*., 1991). The MOCAGE horizontal resolution is 2° both in latitude and longitude and the transport scheme is semi-Lagrangian. It includes 47

hybrid (σ, P) levels from the surface up to 5 hPa (Cathala *et al.*, 2003), where $\sigma = P/P_s$; P and P_s are the pressure and the surface pressure, respectively. MOCAGE has a vertical resolution of about 800 m in the vicinity of the tropopause and in the lower stratosphere. It has the flexibility that it can be used for stratospheric as well as for tropospheric studies, including detailed chemical schemes (e.g. Dufour *et al.*, 2004; Pradier *et al.*, 2006).

The assimilation module is PALM (Projet d'Assimilation par Logiciel Multiméthode), a modular and flexible software developed at CERFACS, which consists of elementary components that exchange data (Lagarde *et al.*, 2001). It manages the dynamic launching of the coupled components (forecast model, algebra operators and input/output of observational data) and the parallel data exchanges.

The technique implemented within PALM and used for the assimilation of O_3 and N_2O profiles from Odin/SMR is the 3D-FGAT method (First Guess at Appropriate Time). This method is a cheap compromise between the well-known 3D-Var and 4D-Var techniques (Fisher and Andersson, 2001). It compares the observation and background at the correct time and assumes that the increment to be added to the background state is constant over all the assimilation window, instead of propagating it with a linear model. The choice of this assimilation technique limits the size of the assimilation window, since it has to be short enough compared to chemistry and transport time-scales. Using ozone profiles from the MIPAS instrument, this technique has produced good-quality results compared to independent data and many other assimilation systems (Geer *et al.*, 2006).

3. 2002–2003 Arctic vortex

During the Arctic winter, the vortex is often affected by stratospheric sudden warmings associated with planetary-scale wave perturbations that originate in the troposphere (Harris *et al.*, 2002). The Arctic vortex is less stable than its Antarctic counterpart and is often displaced off the pole. The area and the position of the Arctic vortex vary enormously from year to year. At temperatures below 195 K, heterogenous reactions can occur on the surface of polar stratospheric clouds (PSCs). Figure 1 shows the minimum temperatures north of 40 °N (from ARPEGE analyses) at four different potential temperature levels from 475 K to 625 K corresponding to the lower stratosphere from November 2002 to March 2003. The 2002–2003 Arctic winter was dominated by a very cold vortex in December and during the first half of January. ARPEGE analyses show an exceptionally low minimum temperature especially from early December to mid-January in the lower stratosphere. The temperature during this period is below the formation temperature of PSCs. This is in agrement with other results using ECMWF analysis (Raffalski *et al.*, 2005; Streibel *et al.*, 2006), Met Office analysis (Singleton *et al.*, 2005) and lidar PSC observations (EORCU, 2003). By the end of December, a minor warming developed in the upper-middle stratosphere. This had practically no effect on the lower-stratospheric polar vortex since it remained strong and stable (EORCU, 2003). In mid-January, a major warming appeared perturbing the vortex strongly. Around 20 January the vortex split into two small vortices. Two other minor warmings appeared around the middle of February and the middle of March. A final warming began at the end of March and the vortex broke down by mid-April. Our analysis indicate that the Arctic polar vortex formed in November 2002 at 625 K, 575 K and 525 K, but at 475 K the vortex was established by 5 December 2002.

4. Results

4.1. Validation of assimilated fields

To assimilate ozone in the model, we have used the linear ozone parametrization developed by Cariolle and Teyssèdre (2007). The scheme is implemented with activation of the ozone destruction due to heterogeneous chemistry with the simplest formulation that uses only local temperature. On the other hand, N_2O is assimilated in the same model but with advection only (i.e. all chemical reactions are switched off).

The initialization field for both the chemically integrated and passive fields is obtained by one month of ozone assimilation. Thus, for the 3 November initialization date, ozone was assimilated between 3 October and 3 November. Separately, the same exercise was done for N_2O within the same period. O_3 is assimilated between $\sim$60 and $\sim$10 hPa ($\sim$19–28 km), whereas N_2O is assimilated in the pressure range $\sim$100–10 hPa ($\sim$16–28 km). At this stage, it is essential to evaluate the consistency of the assimilated fields. We then give a brief overview

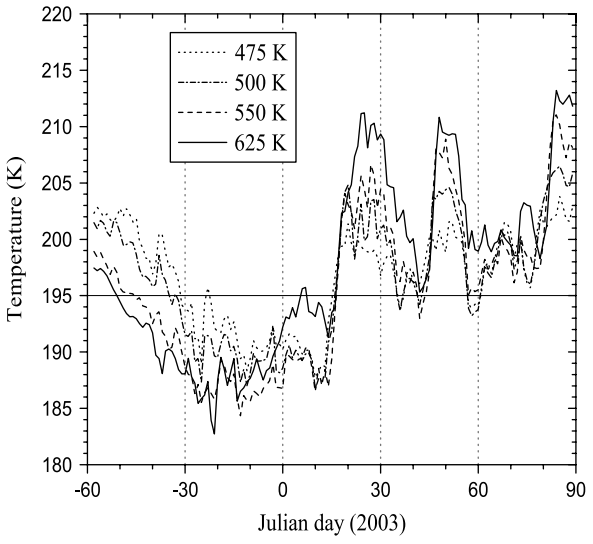


Figure 1. Minimum temperatures in the Northern Hemisphere (north of 40 °N) deduced from ARPEGE analyses from November 2002 to March 2003 at 475, 500, 550 and 625 K potential temperature levels (corresponding to the lower and the middle stratosphere).

about the validation of assimilated measurements from Odin/SMR within MOCAGE–PALM. The comparison of the total ozone derived from O_3 assimilated field and the Total Ozone Mapping Spectrometer (TOMS) shows a difference of 5–10% at high latitudes in the NH. The comparison with ozonesondes in the NH in terms of vertical profiles indicates that the difference is below 2% for the whole pressure range (60–10 hPa). The deviation between O_3 analysis and HALOE is less than 12% between 60 and 10 hPa. The comparison with MIPAS shows that the largest discrepancies are found at low latitudes around 15 hPa where analysis overestimates the MIPAS ozone by 10%. In the NH, comparison of N_2O assimilated field with data obtained from MIPAS yields a good agreement within 5% for the altitude range 100–10 hPa. For more details about the validation of assimilated fields from Odin/SMR, the reader is referred to Massart *et al.* (2007).

In conclusion, the assimilated fields in the lower stratosphere generally have a precision of about 12% and 5% for O_3 and N_2O, respectively. Note that all the above differences are expressed in terms of standard deviation.

Figure 2 presents the evolution of ozonesonde measurements at Ny-Ålesund (79 °N, 12 °E) at the 57.2 hPa level (∼19.7 km) compared to the O_3 assimilated field, the modelled field with the 'full' model and the ozone passive tracer. The comparison is made between the beginning of November 2002 and the end of March 2003. The 'full' model (including advection + linear ozone chemistry) and modelled passive ozone tracer are in reasonable agreement with ozonesonde observations from the beginning of November to practically mid-December 2002. From mid-December 2002, the agreement between the chemistry/no chemistry runs disappears due to the chemical depletion of ozone. In comparison to ozonesonde observations, the model with chemistry behaves relatively well, but sometimes overestimates the ozone at this altitude. However, the use of the assimilation of Odin/SMR ozone in the same model gives satisfactory results; the maximum absolute difference between the two fields does not exceed 0.7 ppmv over the whole period, except on one day (Julian day = 52) where the difference reaches 1.6 ppmv. The correlation between the two fields is about 0.82. The ozone assimilated field and the independent ozonesonde observations are generally in good agreement over the whole assimilation period (November 2002–March 2003).

4.2. Assimilation versus model

In this paragraph, we quantify the added value of the assimilation process on the transport as well as on the chemistry modelled fields. A correlation plot of the averaged values inside the vortex of modelled N_2O versus assimilated N_2O at 625 K potential temperature level is shown in Figure 3(a). A linear regression slope of 0.90 is observed, which is very satisfactory. Since N_2O is assimilated without chemistry, this significant agreement suggests that the transport is well modelled within MOCAGE.

In order to have an idea about how the assimilation improves the chemistry parametrization used in the model, we present in Figure 3(b) the time evolution of the averaged values inside the vortex of O_3 (modelled and assimilated) at the same isentropic level 625 K. From early November to mid-January, the difference between both fields is small (0.2–0.45 ppmv). From mid-January until the end of March, the difference is much more

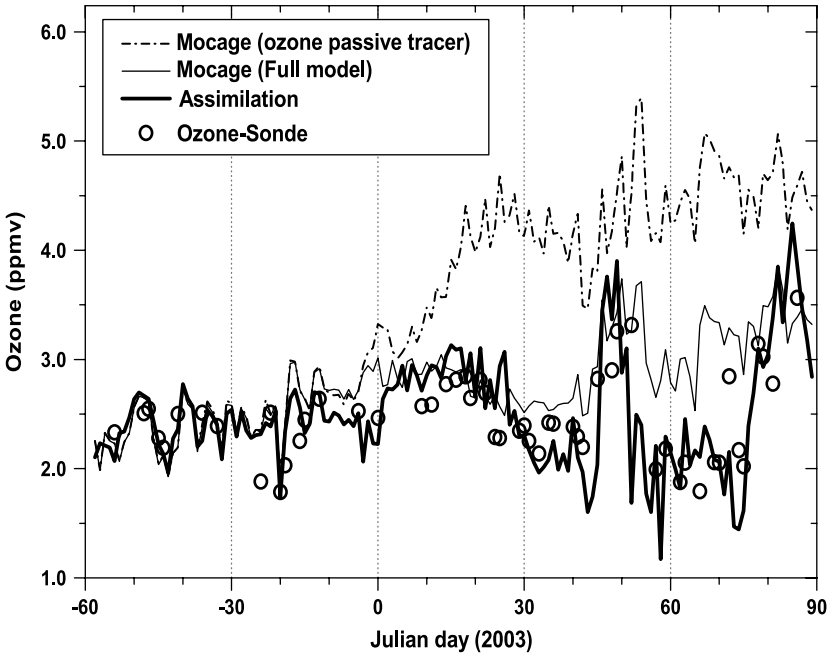


Figure 2. Ozonesonde observations (open circles) at Ny-Ålesund (79 °N, 12 °E) at the 57.2 hPa level compared to MOCAGE ozone passive tracer (dash-dotted line), the modelled field with the 'full' model (thin solid line) and the Odin/SMR ozone assimilated fields (bold solid line).

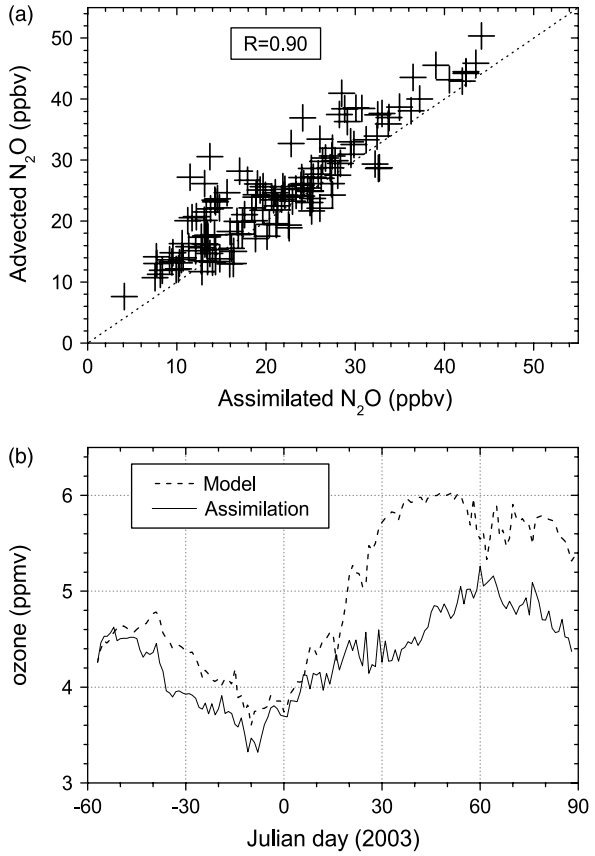


Figure 3. (a) Correlation plot of the averaged values inside the vortex of modelled N_2O versus assimilated N_2O at the 625 K potential temperature level. The linear regression slope is 0.90. (b) Time evolution of the averaged values inside the vortex of O_3 (modelled and assimilated) at the 625 K potential temperature level. In this study, the model uses the linear ozone parametrization of Cariolle and Teyssèdre (2007) to estimate the O_3 evolution (see text for details).

important. Note that the temperature inside the vortex during this period at this potential temperature level is above 195 K (Figure 1). The ozone destruction inside the vortex due to PSC chemistry in the linear ozone parametrization is based on the local temperature. From mid-January, the model is thus unable to destroy ozone through the heterogeneous reactions. This explains the rapid increase of ozone inside the vortex from mid-January in the modelled field, which does not appear in the assimilated field. This example shows that the used linear parametrization of ozone is unable to reproduce the evolution of ozone inside the vortex during the whole period (November 2002–March 2003). In conclusion, the assimilation of Odin data within MOCAGE brings a much more important correction to the chemistry part of the model than to its transport part.

4.3. Vortex edge definition

The vortex edge is the term commonly used to refer to the location of the barrier between the air masses inside and outside the vortex (Müller and Günther, 2003). The determination of the ozone loss inside the vortex with the vortex-average method requires that the edge of the vortex is well localized. Many authors define the vortex edge by using the potential vorticity (PV) field according to Nash criterion (e.g. Rex *et al.*, 1999). However, Greenblatt *et al.* (2002) have shown that small-scale vortex edge features might not be properly represented in analysis of PV directly derived from meteorological fields. Moreover, the degree of isolation of the polar vortex as defined by the PV field is not well known (Jost *et al.*, 2002). The PV field is poorly conserved in small-scale filaments due to radiative and dynamical processes (Hauchecorne *et al.*, 2002). Allen and Nakamura (2003) also argue that the tracer-based method has several advantages over the PV method in determining the location of the vortex; in particular, PV is not conserved isentropically beyond the time-scale in which diabatic effects are negligible.

Another parameter used to diagnose the polar vortex is the modified PV (MPV) introduced by Lait (1994) in order to remove the altitude dependence of PV without destroying its structure on a given isentropic surface. This parameter has been recently used for polar vortex studies (e.g. Christensen *et al.*, 2005). It is defined as a function of PV and potential temperature θ as

$$MPV = PV \left(\frac{\theta}{475} \right)^{-\frac{9}{2}} .$$

In this study we determine the vortex edge by using the N_2O assimilated field as a dynamical tracer since this molecule is chemically inert in the lower stratosphere. Moreover, the sharp gradient in N_2O at the vortex edge is likewise noticeable in satellite measurements (e.g. Manney *et al.*, 1999). In order to evaluate this method, the results are compared to the use of the maximum gradient of the MPV field as a vortex edge criterion. Figure 4 (left-hand side) shows maps of the assimilated N_2O field at different potential temperature levels on selected days. The same figure (right-hand side) gives the corresponding MPV fields. In both cases, the edge of the vortex determined by the maximum gradient is denoted by a solid black contour. The comparison between the two fields shows that in the NH the minimum values of N_2O correspond to the maximum values of MPV. Both fields also show almost the same structure during December and January at 475 K and 525 K when the vortex was strong (first and second row). By mid-February, the vortex was perturbed and began to split, this being more marked at 575 K and 625 K (third row). In this case the comparison between the two fields shows that the MPV field is smooth, while N_2O assimilated fields show more detailed structures and filaments. The equivalent latitude corresponding to the maximum gradient of both fields during this period shows some differences (e.g. Table I for 20 January 2003 for 575 K and 625 K). With the N_2O method, we clearly see midlatitude intrusions of air masses in the inner vortex (Figure 4, third row). Qualitatively there is good agreement between the two methods. However, when calculating the equivalent latitude corresponding to their maximum gradient (Table I), we note some differences

Table I. Equivalent latitude (in degrees) corresponding to the maximum gradient of N_2O and MPV fields at different potential temperature levels for specific days.

	475 K		525 K		575 K		625 K	
	N_2O	MPV	N_2O	MPV	N_2O	MPV	N_2O	MPV
14 December 2002	63.0	63.0	65.0	63.8	**63.0**	**67.7**	**61.0**	**69.8**
30 December 2002	64.0	65.0	63.0	63.4	**63.1**	**70.8**	**58.2**	**62.5**
20 January 2003	68.5	68.6	65.2	66.4	**62.0**	**66.4**	**63.0**	**67.5**
28 January 2003	67.7	68.1	67.0	67.3	67.2	68.3	65.1	66.0
10 February 2003	67.1	66.5	69.2	68.8	67.1	67.5	66.2	68.1
25 February 2003	69.0	69.1	67.7	69.4	**65.0**	**69.6**	**65.3**	**69.6**
5 March 2003	69.2	69.1	67.3	68.9	67.2	69.7	67.7	69.2
23 March 2003	69.0	70.2	69.0	69.7	69.4	69.8	69.0	70.2

Bold denotes that the values determined by the two methods differ by more than 4 degrees.

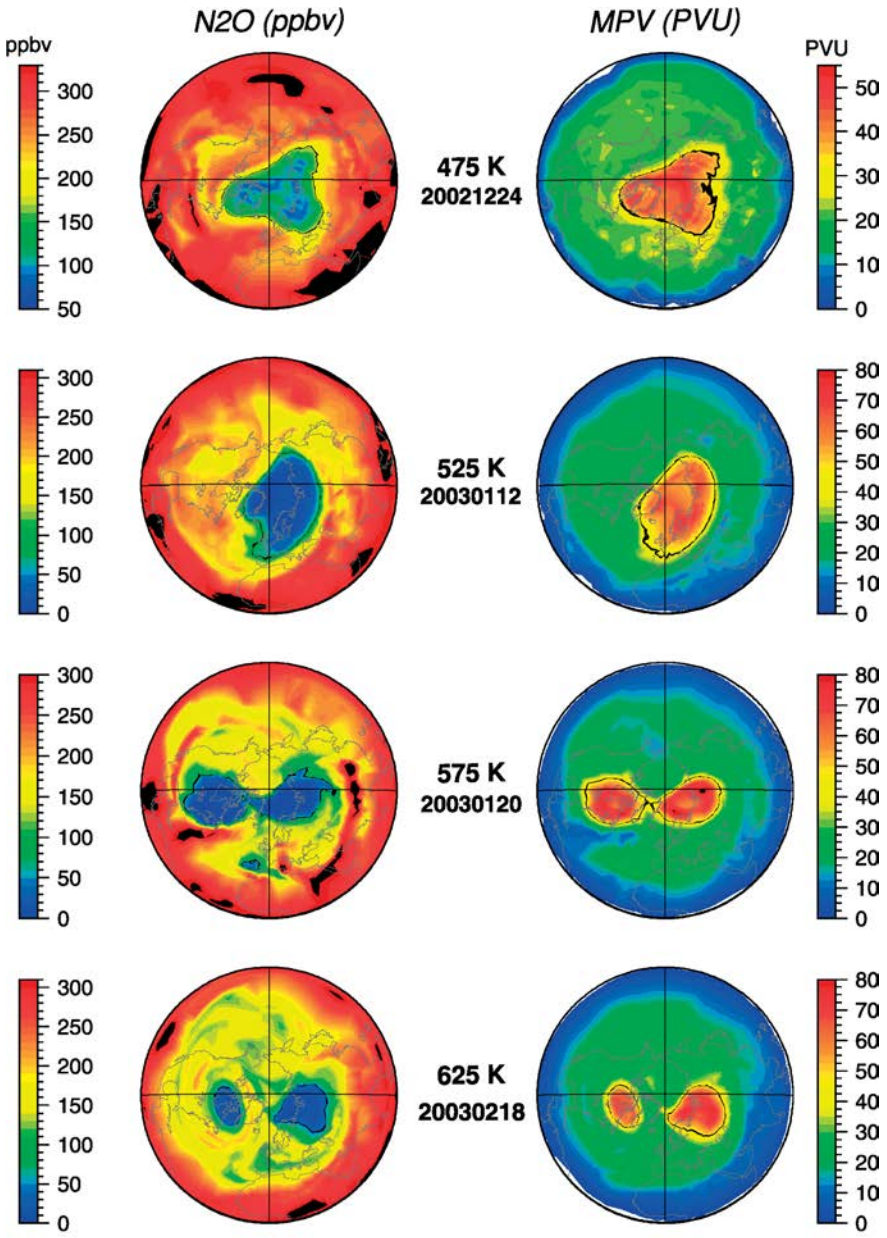


Figure 4. Odin/SMR assimilated N_2O field (left-hand side) and the corresponding modified potential vorticity (MPV) field from ARPEGE analyses (right-hand side) for potential temperature levels (475, 525, 575 and 625 K) for the selected days (24 December 2002, 12 January 2003, 20 January 2003 and 6 March 2003). The edge of the vortex determined by both methods is denoted by the solid black contour.

especially at 575 K and 625 K during the minor warming of December 2002, the major warming of January 2003 and the minor warming of February. An investigation of the difference between the two methods in determining the vortex edge is beyond the scope of this paper and is the subject of ongoing work.

4.4. Diabatic descent

The evolution of stratospheric ozone in the winter Arctic vortex is governed by both chemistry and transport. Ozone variations due to transport are often of the same magnitude as those due to chemical ozone destruction (Tilmes *et al*., 2004). Subsidence always masks the chemical ozone loss in the lower stratosphere. Descent of air tends to increase the ozone mixing ratio at a given altitude. Air with large mixing ratios of ozone is transported downwards into the lower stratosphere, the region where chemical ozone depletion occurs.

N_2O 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 and of 100 years up to 22 km (WMO, 1986). Therefore N_2O is well suited to study the vertical motion of the air masses inside the polar vortex. The evolution of N_2O inside the vortex is a combination of both diabatic descent and mixing. Figure 5 shows the time evolution from 10 December 2002 to 20 March 2003 of analyzed N_2O mixing ratio averaged over ten-day intervals over the vortex area at the 475, 525, 575 and 625 K isentropic levels. The descent at 475 K and 525 K inside the vortex was stronger, and extended over a longer period, than the descent at 575 K and 625 K levels. Further, the N_2O values inside the vortex decreased rapidly in December, and stabilized from mid-January to mid-February for 475 K and 525 K. The value of N_2O inside the vortex increased for the 575 K and 625 K levels during the same period. This is associated with the mixing with air from outside the vortex that occurred after the major warming of mid-January as reported by Urban *et al*. (2004). N_2O values decreased during early March, and after that time we see a final increase of N_2O inside the vortex at all the studied levels, which is in connection with the vortex break-up.

Figure 6 shows mean potential temperature levels corresponding to the vortex averages of the 25, 50, 75 and 100 ppbv levels of N_2O. The descent of all these levels is clearly seen from the beginning of November until the beginning of February. The 25 ppbv N_2O level descended from a potential temperature level of 595 K to ∼525 K. The 50 ppbv level descended from 560 K to ∼495 K. During the beginning of February, the potential temperature level of all N_2O levels hardly varied (∼530 K and ∼500 K for 25 and 50 ppbv N_2O levels, respectively). This indicates that the diabatic descent in this period was weak. The 75 and 100 ppbv N_2O levels descended below 500 K from the middle of December.

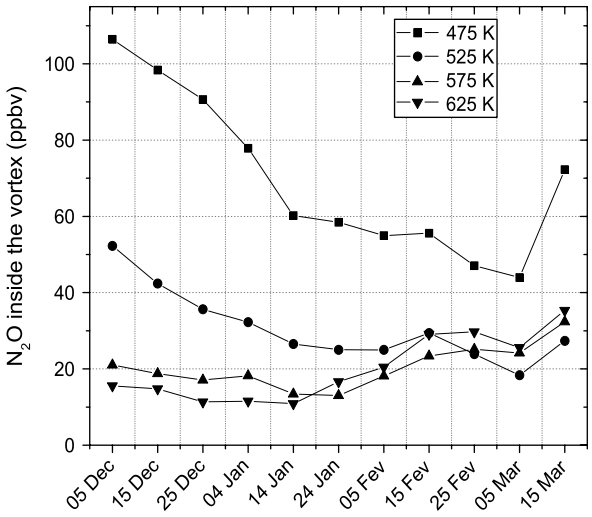


Figure 5. Time evolution of the assimilated N_2O mixing ratios inside the vortex averaged over ten-day periods at 475, 525, 575 and 625 K for the whole assimilation period (November 2002–March 2003).

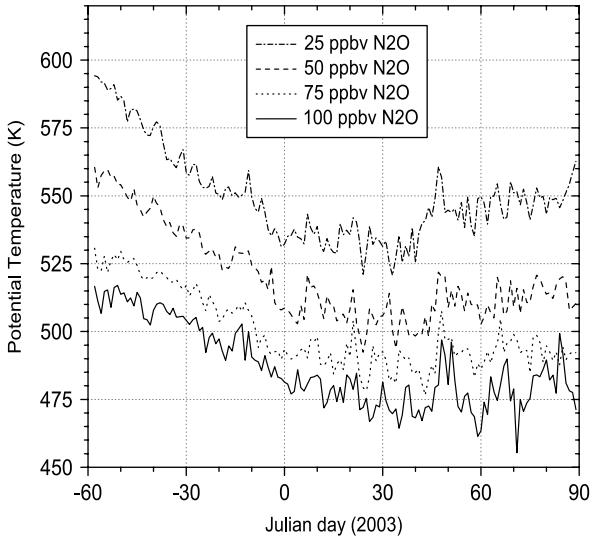


Figure 6. Diabatic descent of the averaged values of different N_2O levels (25, 50, 75 and 100 ppbv) for the whole assimilation period (November 2002–March 2003). The 25 and 50 ppbv N_2O level air masses are above the 500 K potential temperature level for the whole assimilation period, whereas the 75 and 100 ppbv N_2O level air masses descend below 500 K starting in mid-December 2002.

4.5. Evolution of ozone inside the vortex

In order to investigate the evolution of ozone inside the polar vortex, the average values were calculated at different potential temperature levels. Figure 7 represents the evolution of the assimilated ozone averaged inside the vortex from November 2002 to March 2003 at 475, 525, 575 and 625 K potential temperature levels. The enhancement of the average ozone value at the beginning of formation of the vortex can be attributed to the diabatic subsidence, which brought ozone-rich air masses from higher to lower levels. After a few days, the ozone average value inside the vortex decreased substantially. This is due to the chemical ozone depletion,

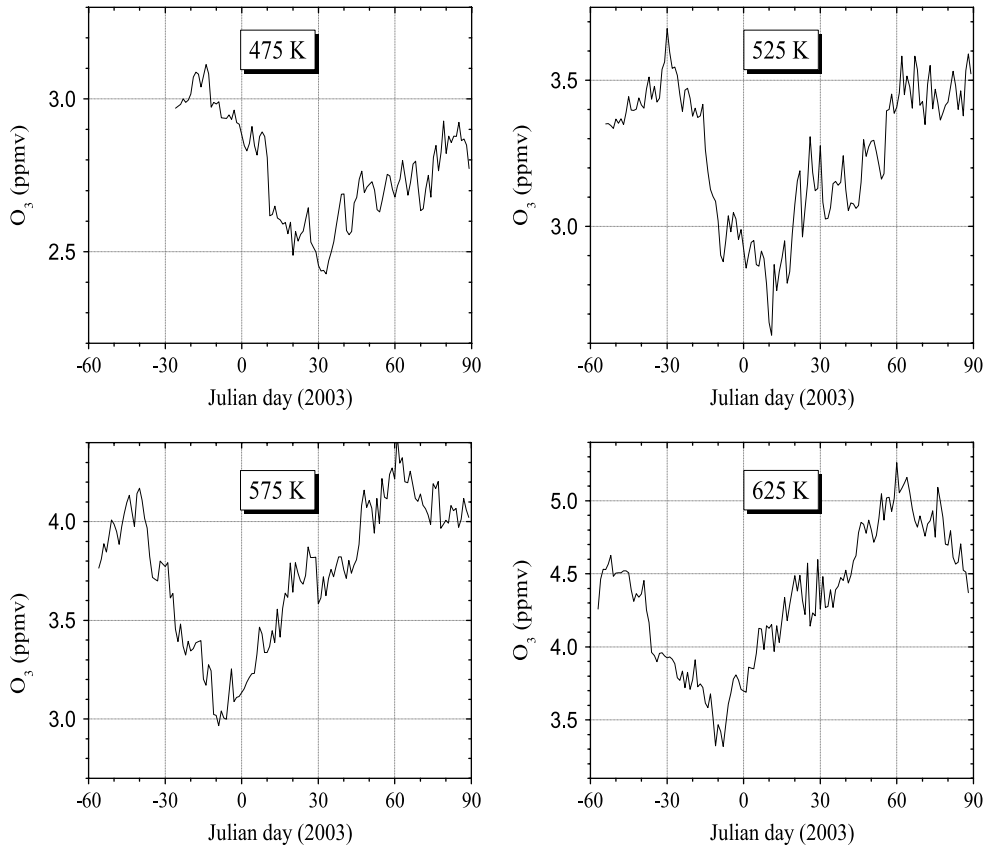


Figure 7. Time evolution of the assimilated ozone averaged inside the vortex for different potential temperature levels (475, 525, 575 and 625 K) from November 2002 to March 2003. The vortex edge is determined by the maximum gradient of the N_2O assimilated field on each potential temperature level.

which is more important than the diabatic subsidence. The major ozone decrease inside the vortex is shown in December 2002 and early January 2003. During this period the vortex was somewhat centred off the pole and moved towards lower latitudes (e.g. Figure 4, upper three rows). Singleton *et al*. (2005) reported that during December 2002 and January 2003 the vortex was often elongated allowing in-vortex air to make frequent excursions into the sunlight at lower latitudes. This situation as well as the low temperatures allowed the early advent of the ozone loss.

The evolution of O_3 inside the vortex in Figure 7 shows that on the 475 K level, ozone inside the vortex decreased from mid-December until the beginning of February when the assimilated ozone averaged inside the vortex was a minimum. For this level, ozone inside the vortex was reduced by 21.8 ± 5% by the beginning of February with respect to its average value at the end of November. At 525 K, ozone was reduced by 1.0 ±0.2 ppmv between the end of December and the middle of January. At 575 K, at the beginning of formation of the vortex by the middle of November, the average value of ozone was 4.2 ±0.3 ppmv, and by the end of December it was 3.0 ±0.3 ppmv. The ozone reduction on this potential temperature level was 29 ± 6%. At 625 K, we find that ozone was reduced by ∼28 ± 6% between the beginning of November and the end of December.

The evolution of O_3 inside the vortex in Figure 7 includes both chemical loss and diabatic descent. In order to extract the actual chemical ozone loss, we use the N_2O assimilated field to subtract out the effect of subsidence, which masks the chemical ozone loss.

4.6. Chemical ozone loss

During winter, diabatic cooling in the vortex results in subsidence. Diabatic descent must then be accounted for in the calculation of chemical ozone loss. The time evolution of N_2O inside the vortex is used to remove the contribution of subsidence. Figure 8 shows time series of ozone inside the vortex at 25 ppbv and 50 ppbv N_2O levels. On the 25 ppbv N_2O level, ozone was chemically depleted by 28 ± 6% (1.1 ± 0.3 ppmv) by mid-January with respect to its average value at the middle of November. During this period, air masses on this level descended from ∼581 K down to ∼531 K (Figure 6). On the 50 ppbv level, the ozone was chemically depleted by 26 ± 5% (0.9 ± 0.2 ppmv) between the middle of November and the end of January. A linear fit over the same periods as the chemical ozone loss gives a chemical ozone loss rate of ∼18 ppbv day^{-1} and ∼9.3 ppbv day^{-1} on the 25 ppbv and 50 ppbv N_2O levels, respectively.

In order to deduce the chemical ozone loss profile, the time evolution of N_2O inside the vortex is used to remove the contribution of subsidence using the same

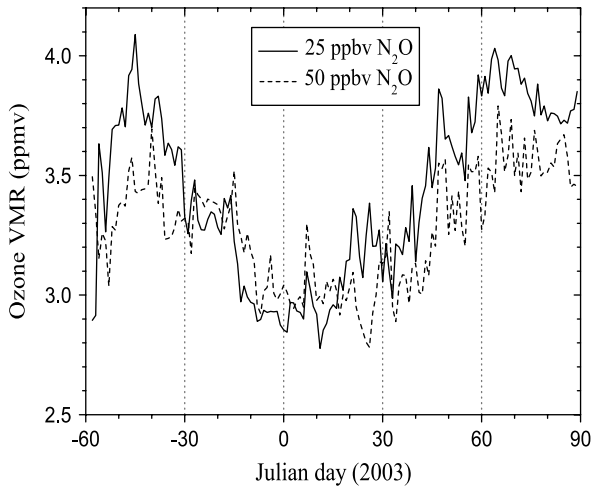


Figure 8. Time series of averaged ozone inside the vortex at 25 and 50 ppbv N_2O levels from November 2002 to March 2003.

method as Rex *et al.* (2002). Figure 9 presents the vortex-averaged ozone loss profile between mid-December and early February as a function of potential temperature. The profile starts at 475 K (∼19 km); this altitude is the lower limit of Odin/SMR O_3 measurements in the retrieved band (501.8 GHz). The largest chemical ozone loss deduced from the O_3 assimilated field is observed at 475 K at 0.98 ± 0.2 ppmv.

Since the transport is well modelled within MOCAGE, the modelled ozone without chemistry and the ozone assimilated field are used through the passive tracer method in order to infer the chemical ozone loss and validate the results obtained with the vortex-average technique. The advantage of the passive tracer method is that no correction related to the diabatic descent is required. The chemical ozone loss is directly inferred on each selected isentropic level.

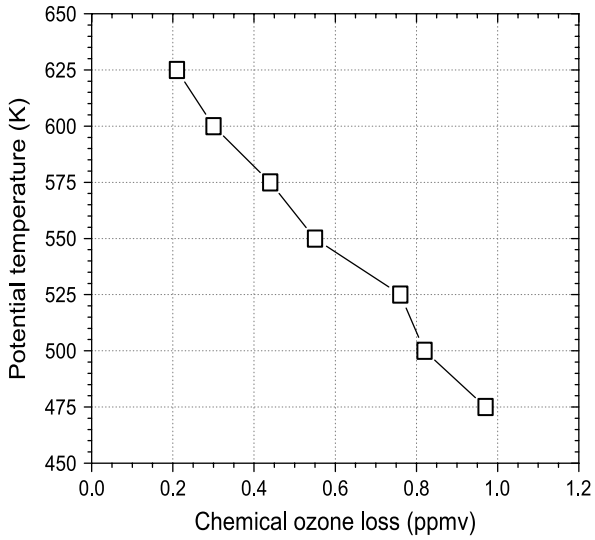


Figure 9. Estimated ozone loss mixing ratio between mid-December and early February versus potential temperature after removing the effects of diabatic descent. In the studied domain, the maximum ozone loss of ∼1 ppmv is observed at 475 K.

Results indicate that the passive tracer loss is quite similar to the vortex-average results above 500 K. Nevertheless, the passive tracer gives slightly higher estimates of the ozone loss at 475 (∼1.15 ppmv) and 500 K (∼0.9 ppmv).

The objective of the next Section is to compare our results with the findings from other independent observations.

5. Comparison with other results

The ozone loss of the winter 2002–2003 was reported by many authors using many datasets and a wide range of methods.

Raffalski *et al.* (2005) used ozone measurements from the millimetre wave radiometer at Kiruna, Sweden to deduce the ozone decrease. They used N_2O measurements from the Odin/SMR instrument in order to remove the effect of subsidence. They found that between mid-December and mid-February, the chemical ozone loss was 0.5 and 0.9 ppmv on the 25 ppbv and 50 ppbv levels, respectively. In comparison to these results, we found that between the middle of December and the middle of January, the chemical ozone loss was ∼0.7 and ∼0.9 ppmv on the 25 and 50 ppbv N_2O levels, respectively.

Using the passive tracer technique, Singleton *et al.* (2005) used Polar Ozone and Aerosol Measurement (POAM III) satellite observations and the SLIMCAT-CTM to deduce the ozone loss. They found that the ozone loss was approximately 0.7 ppmv between December and mid-January at the 500 K isentropic level. Our results indicate that on the same potential temperature level, ozone is chemically depleted by 0.82 ppmv between mid-December and early February.

Tilmes *et al.* (2003) reported a very early chlorine activation and ozone loss from the analysis of the HALOE data using the ozone-tracer correlation method. They found a maximum local ozone loss of 1.5 ppmv at ∼440 K between mid-December and February. Their analysis shows that the ozone loss during the same period was ∼1.1 and ∼0.8 ppmv at 475 K and 500 K, respectively. Unfortunately, in the used band, Odin/SMR data do not allow ozone loss calculation for lower altitudes (below 475 K) due to the limited range of O_3 measurements. Nevertheless, in comparison to these results, we found that the chemical ozone loss obtained at 475 K and 500 K was 0.98 and 0.82 ppmv, respectively.

Christensen *et al.* (2005) have used the vortex-average technique to infer the ozone loss in the Arctic vortex (as defined by Nash *et al.*, 1996) from ozone sonde data. We find quite good agreement when we compare our results to their results. In fact, they found that ozone loss is 1.0 ± 0.2 ppmv at 479 K, while we find 0.98 ± 0.2 ppmv at the 475 K potential temperature level.

Streibel *et al.* (2006) used the Match technique to determine the chemical ozone loss inside the vortex during winter 2002–2003. Their results reveal that the ozone destruction rates varied between 10 and 15 ppbv day^{-1}.

They also found that the accumulated ozone loss was 1.3 and 0.55 ppmv at the 475 K and 500 K potential temperature levels, respectively. We found a chemical ozone loss rates of the same magnitude ($\sim$9.3 and $\sim$18 ppbv day^{-1} on the 50 ppbv and 25 ppbv levels, respectively). The chemical ozone loss obtained by our analysis is 0.98 and 0.82 ppmv at 475 K and 500 K, respectively. The differences between our method and the Match technique may be explained by the fact that the two methods do not use the same vortex edge definition (Christensen *et al.*, 2005).

Ozone loss results inferred from the modelled ozone without chemistry and the ozone assimilated field within the passive tracer method are quite similar to the vortex-average results above 500 K. However at 475 and 500 K, results indicate that the ozone loss deduced by the passive tracer is slightly higher. The differences between the two methods is not the subject of this study. To understand the discrepancies between the two methods, additional investigations are required.

Generally, the comparison of our results regarding the 2002–2003 Arctic vortex to those of other authors using different datasets and different techniques generally gives quite good agreement. However, Feng *et al.* (2005) reported that the high-resolution model shows large ozone loss in the lower stratosphere. The models still fail to reproduce many aspects of polar chemistry and transport (e.g. chemical ozone loss and diabatic descent). Indeed, in this study, the model with the implemented parametrization of ozone is unable to reproduce the ozone evolution inside the vortex (Figures 2 and 3). Consequently, it is unable to provide a better estimate of the chemical ozone loss inside the vortex. In this case, the use of chemical data assimilation can be a valuable tool to overcome these possible deficiencies of the models. In particular, chemical data assimilation is mainly used here to correct the model heterogeneous ozone depletion and reproduce a near-complete ozone destruction inside the vortex. Consequently, the assimilated fields can better describe the large-scale evolution of the NH stratospheric vortex in comparison to the model alone.

6. Conclusions

We have used assimilated fields of O_3 and N_2O from Odin/SMR in order to describe the large-scale evolution of the polar stratospheric vortex during the winter 2002–2003.

The N_2O assimilated fields were used to estimate the location of the vortex edge using their maximum gradient according to Nash *et al.* (1996). The results are compared to the maximum gradient of modified potential vorticity as a vortex edge criterion in terms of equivalent latitude. Both methods agree well during the whole assimilation period for all studied levels except at high levels (575 K and 625 K) during the minor warming of December, the major warming of January and after the minor warming during February. A detailed comparison between the two methods is not the subject of this paper. It is the subject of an ongoing work.

The N_2O assimilated fields were also used to remove the contribution of subsidence. Results indicate that diabatic descent was important from the beginning of November to early February. In particular, during this period, air masses on the 25 ppbv N_2O level subside from $\sim$595 K down to $\sim$525 K. Those of 50 ppbv N_2O level descend from 560 K down to 495 K.

The assimilation of ozone enabled us to overcome the imperfections of the MOCAGE model in connection with the linear ozone parametrization used in this study. Chemical data assimilation of ozone is mainly used here to describe the ozone evolution and to correct the model heterogeneous ozone depletion and reproduce a near-complete ozone destruction in the vortex. Results indicate that ozone is chemically depleted by 1.1 $\pm$ 0.2 ppmv on the 25 ppbv N_2O level between the middle of November and mid-January. On the 50 ppbv N_2O level, ozone is chemically depleted by 0.9 $\pm$ 0.2 ppmv between the middle of November and the end of January. A linear fit over the same periods gives a chemical ozone loss rate of $\sim$18 ppbv day^{-1} and $\sim$9.3 ppbv day^{-1} on the 25 ppbv and 50 ppbv N_2O levels, respectively. The vortex-averaged ozone loss profile from the O_3 assimilated field shows a maximum of 0.98 ppmv at 475 K. Additionally, the passive tracer method indicates that the ozone loss results are quite similar to the vortex-average results above 500 K. At 475 and 500 K, the passive tracer method gives slightly higher values.

Comparisons of ozone loss at different isentropic levels with the Match technique reveal that the Match technique gives slightly higher estimates, particularly at 475 K. This discrepancy may be explained by the fact that the two methods do not use the same vortex edge definition. Thus, great care should to be taken in order to have a careful comparison with the Match technique (Christensen *et al.*, 2005).

Comparisons with other results from the winter 2002–2003 show quite good agreement with chemical ozone loss inferred from HALOE data using the the ozone-tracer correlation method (Tilmes *et al.*, 2003). Results derived from POAM III and the SLIMCAT-CTM using the passive tracer method are slightly smaller than our results, while we find very good agreement with the results of Christensen *et al.* (2005) using ozonesonde data within the vortex-average technique.

The comparison of our results to others using different techniques and different measurements during the same period shows generally good agreement. This demonstrates the power of chemical data assimilation to overcome the possible deficiencies of the model and to describe the large-scale behaviour of the NH stratospheric vortex. This is particularly interesting insofar as the specific objective of chemical data assimilation is to produce a self-consistent picture of the atmosphere taking into account both the available observations and our theoretical understanding of the chemical and transport system.

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