

Estimation of Arctic O₃ loss during winter 2006/2007 using data assimilation and comparison with a chemical transport model

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The evolution of Arctic stratospheric O₃ throughout the winter and spring of 2006/2007 is estimated by a state-of-the-art chemical transport model (CTM) and by a 3D-Var assimilation system using O₃ data from the Earth Observing System (EOS) Microwave Limb Sounder (MLS) and Solar Backscatter Ultraviolet Radiometer (SBUV/2) satellites. Modelled and assimilated O₃ compare well with MLS measurements. The aim of this article is to compare O₃ loss estimates derived from the CTM and data-assimilation results, and as a result point to further developments in the method of inferring O₃ loss using data assimilation. The methods for inferring O₃ loss are discussed and compared with other published methods. The assimilation-system vertical transport is found to be too fast, in agreement with an earlier study, although this affects only the O₃ reference field used for the loss estimation. Improving the O₃ reference in the assimilation method used here provides a maximum vortex average O₃ loss range of 0.8–1.2 ppmv at 68 hPa and 1.0–1.5 ppmv at 46 hPa, peaking at the beginning of March. The corresponding CTM values are 1.4 and 1.6 ppmv, respectively, with the peak lagging the assimilation by a few days at 46 hPa. We show that using a passive tracer as reference for O₃ loss does not provide the best estimate for polar stratospheric cloud (PSC)-related loss for this winter; up to 40% of total O₃ loss is shown not to be related to PSCs or heterogeneous chemistry. Hence, the use of a passive O₃ reference for estimating PSC-related O₃ loss should be made with care. In addition to the vortex average losses, we estimate an innermost vortex O₃ loss of 1.4 ppmv due to PSCs only. Transport effects and differences between the CTM and the assimilation system are discussed, and possible improvements for both models are suggested. Copyright © Royal Meteorological Society and Crown Copyright, 2011

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1. Introduction

Chemical O₃ loss in the Arctic polar winter stratosphere has been estimated by a variety of methods, including compact

correlations between O₃ and long-lived tracers measured by balloons and satellites, the profile-descent method, the Lagrangian match technique and the passive-subtraction method (see WMO, 2006 for a summary). In recent years,

data-assimilation-based methods have been developed to infer polar O₃ loss, based on the assimilation of O₃ satellite observations into chemical transport models (Rösevall *et al.*, 2007; El Amraoui *et al.*, 2008) or into general circulation models (Jackson and Orsolini, 2008). One advantage of the data-assimilation (DA) method is that it makes optimal use of all available observations. Arctic O₃ loss has been derived from the latter methods for only a few winters. It is therefore of great interest to further infer O₃ loss using DA methods during other winters, to compare the results with other observation-based methods and transport models with comprehensive gas-phase and heterogeneous chemistry. The latter have been widely used in O₃ depletion studies (recent examples include Tripathi *et al.*, 2007; Singleton *et al.*, 2007; Konopka *et al.*, 2007; Søvde *et al.*, 2008).

In polar winter, the term O₃ loss is generally used for halogen-induced O₃ loss, but in this study we will use the term for total O₃ loss and specify the halogen-related loss when needed.

It is difficult to carry out accurate comparisons of O₃ loss between the different methods described in WMO (2006), because the same definition of vortex edge, altitude range, periods and so forth would have to be used. When inferring O₃ loss a reference is always needed, representing how O₃ would evolve if the loss processes were absent. Chemical transport model (CTM) studies often use a passive tracer (Harris *et al.*, 2002); however, as pointed out by Singleton *et al.* (2005), the passive tracer introduces possible errors in the O₃ reference: gas-phase NO_x chemistry, including catalytic cycles destroying O₃ (Crutzen, 1970), will not be accounted for, giving too high an O₃ reference (from subsidence), and O₃ from production regions at low latitudes will be unaccounted for giving too low an O₃ reference. For a polar vortex displaced from the Pole, the Sun may reach large parts of the vortex and the NO_x effect may be quite large even in early to mid-February (Tilmes *et al.*, 2006; Konopka *et al.*, 2007). There may also be transport of NO_x-processed O₃ from regions further above or through the weak edges of the vortex (at all altitudes), and models may artificially transport air (and O₃) through the vortex edges. Hence the use of a passive tracer may not provide the best reference for estimating O₃ loss due to heterogeneous processes, but will diagnose all changes due to both loss and production. The passive O₃ method, as used by Singleton *et al.* (2007), is amenable to a detailed comparison with our data-assimilation method, which also relies on Microwave Limb Sounder (MLS) observations and on a passively transported O₃ reference field.

The aim of this study is to test the data-assimilation method to infer O₃ loss and to point the way to future developments of this method described by Jackson and Orsolini (2008), based on the UK Met Office (UKMO) assimilation system. To this end we compare the assimilated O₃ and its inferred O₃ loss with those from the state-of-the-art Oslo CTM2 described by Søvde *et al.* (2008), which is driven by meteorology from the European Centre for Medium-Range Weather Forecasts (ECMWF), and we draw detailed comparisons with MLS measurements.

The assimilation method is described in section 2 and comprises two simulations, one in which dynamics and O₃ are assimilated and one in which dynamics only is assimilated to transport a reference O₃ field passively. For the Oslo CTM2, which, in contrast to the UK model, describes all known important chemical processes in detail,

the O₃ loss due to polar stratospheric cloud (PSC) related processes is derived, as well as differences from a passive tracer simulation. For technical and economic reasons, it was not possible to run the two models with the same meteorology. However, comparing transport of an initially identical O₃ field by both models has allowed us to assess the transport differences caused by the combined effect of different meteorology and transport schemes. The different simulations carried out in this study are listed in Table I.

Here we study O₃ depletion during the Arctic winter and early spring of 2006/2007. The Arctic lower stratospheric temperatures were below the 25-year average during most of this period. Low O₃ abundance in the polar vortex and high levels of ClO, indicative of chlorine activation and O₃ loss, have been observed by the Earth Observing System (EOS) Microwave Limb Sounder (MLS) aboard the AURA satellite and the Sub-Millimetre Radiometer (SMR) about the Odin satellite (Rösevall *et al.*, 2007). The vortex temperatures were low enough for PSC formation in the first part of December, until a strong warming event in late December. After that, no PSCs were detected until 11 January. A major final warming occurred at the end of February, with a few short-lived drops below PSC threshold values in mid-March, although the lower stratosphere vortex remained stable until mid-April. We are interested in the bulk O₃ loss, which occurs in late spring, and therefore start our simulations in early January, just before the January onset of PSC formation. We will not estimate the early winter loss.

2. The Met Office O₃ assimilation system

The approach by which data assimilation is used to estimate chemical O₃ loss is detailed in Jackson and Orsolini (2008), but is summarized here. The O₃ assimilation scheme is based on a 3D-Var version of the operational Met Office assimilation system (Lorenc *et al.*, 2000), and uses a forecast model that is semi-Lagrangian with a height-based vertical coordinate (Davies *et al.*, 2005). The model version used here has 50 levels from the surface to around 63 km and a horizontal resolution of 2.5° latitude × 3.75° longitude. EOS MLS and Solar Backscatter Ultraviolet Radiometer (SBUV/2) O₃ observations are assimilated, together with dynamical observations from satellites, aircraft, radiosondes and surface stations. Further details of the O₃ assimilation scheme and the impact of the assimilation of EOS MLS and SBUV/2 data on O₃ analyses are given in Jackson (2007) and references therein.

A realistic O₃ field is established before the bulk O₃ destruction due to PSCs occurs, and is produced by running the O₃ assimilation (referred to as UKASSIM) for 7 days, starting on 3 January 2007. From 10 January, the O₃ assimilation run is continued until 20 March, while a reference run is also started using the same initial conditions. This reference run (referred to as UKREF) is like the assimilation run, except that no O₃ observations are assimilated. Hence in UKREF O₃ is initialized with EOS MLS and SBUV/2 observations and thereafter transported passively by assimilated winds. The seven-day initialization step ensures that the reference O₃ is realistic and shares the same potential observational biases as the assimilated O₃ (UKASSIM).

UKREF O₃ is subtracted from the UKASSIM O₃ field to provide an estimate of O₃ loss. The dynamical fields in both runs are near-identical and thus differences between the

Table I. Description of the simulations in this study.

Simulation	Description	Winds	Start date
CTMFULL	Oslo CTM2 with full chemistry	ECMWF IFS	1 January
CTMnoPSC	Oslo CTM2 with no heterogeneous reactions on PSCs	ECMWF IFS	10 January
CTMPASS	Oslo CTM2 with passively transported O ₃	ECMWF IFS	10 January
UKASSIM	Met Office stratospheric assimilation of dynamics and MLS and SBUV O ₃	Met Office	3 January
UKREF	UK stratospheric assimilation of dynamics, transporting O ₃ passively; initial O ₃ is from UKASSIM at 10 January.	Met Office	10 January
CTMUKREF	Initial UKREF O ₃ passively transported by the Oslo CTM2	ECMWF IFS	10 January

assimilated and reference O₃ fields should in principle not be caused by differences in transport between the runs and can therefore be attributed to chemistry. In the middle and high-latitude winter lower stratosphere, it is likely that these chemical changes will be overwhelmingly due to heterogeneous PSC-related chemistry. It is important to note that the reference run does not capture NO_x-related O₃ chemistry; hence the method diagnoses all loss processes caused by either gas-phase or heterogeneous chemistry. As noted in section 1, O₃ production due to NO_x at lower latitudes is also not captured.

3. The Oslo CTM2

The Oslo CTM2 is a global chemical transport model comprising comprehensive chemistry for the troposphere and the stratosphere. The Oslo CTM2 has previously been tested against observations and applied in O₃ depletion studies (Søvde *et al.*, 2008 and references therein). The Oslo CTM2 is driven by three-hourly meteorological forecast data from the ECMWF Integrated Forecast System (IFS) model, which are produced with 12 hours of spin-up starting from an ERA-Interim analysis at noon on the previous day. Advective transport is carried out with the low-diffusive second-order moments scheme (Prather, 1986).

Surface emissions are set at year 2000 (POET: Granier *et al.*, 2005). Boundary conditions for stratospheric species are set from observations at the surface and model top as explained by Søvde *et al.* (2008). Chemical kinetics are calculated based on the meteorological temperature and pressure, while calculation of photodissociation coefficients is carried out by the Fast-J2 method (Wild *et al.*, 2000; Bian and Prather, 2002).

Recently, some improvements have been made to the Oslo CTM2. The vertical span of the model has been increased to 60 sigma-pressure hybrid layers, extending from the surface up to 0.1 hPa, to improve the stratospheric circulation. The Oslo CTM2 has recently been run with 60 layer meteorology using only tropospheric chemistry (Dalsøren *et al.*, 2010), and also using full stratospheric chemistry (Eleftheratos *et al.*, 2010). The vertical resolution in the tropopause region varies between roughly 0.8 and 1.2 km. In this study the horizontal resolution is T42 (2.8° × 2.8°). Another improvement is that the microphysics of PSCs has been updated since Søvde *et al.* (2008), as described in the appendix.

Starting from a previously steady-state simulation for year 2000, the Oslo CTM2 was spun up using meteorological data from ECMWF IFS cycle 29, in T21 resolution for 1 January 2000–1 November 2004, followed by T42 carried out for 2 years (1 November 2004–1 December 2006). Cycle 36

Table II. Heterogeneous reactions on PSCs in the Oslo CTM2. Reactions marked ‘*’ also occur on background aerosols (included in CTMFULL and CTMnoPSC), with other kinetics.

N ₂ O ₅ + H ₂ O	→	2HNO ₃	*
N ₂ O ₅ + HCl	→	ClONO + HNO ₃	
ClONO ₂ + H ₂ O	→	HOCl + HNO ₃	*
ClONO ₂ + HCl	→	Cl ₂ + HNO ₃	*
HOCl + HCl	→	Cl ₂ + H ₂ O	*
BrONO ₂ + H ₂ O	→	HOBr + HNO ₃	*
BrONO ₂ + HCl	→	BrCl + HNO ₃	*
HOBr + HCl	→	BrCl + H ₂ O	*
ClONO ₂ + HBr	→	BrCl + HNO ₃	

Table III. Description of the O₃ loss inferences in this study.

Inferred O ₃ loss	Diagnosed processes
CTMFULL – CTMnoPSC	PSC
UKASSIM – UKREF	PSC + NO _x
CTMFULL – CTMPASS	PSC + NO _x
UKASSIM – CTMUKREF	PSC + NO _x

data was used for the rest of the Arctic winter 2006/2007. The CTM simulations presented here cover the period 1 January–30 April 2007.

When the Oslo CTM2 is used to infer O₃ loss, two methods are used. One is the passive tracer method which infers O₃ change from all processes (e.g. NO_x and transport), while the other diagnoses only PSC processes by carrying out a separate full-chemistry simulation where heterogeneous chemical reactions on PSCs (listed in Table II) have been switched off. These reactions are responsible for the halogen activation causing the O₃ depletion, and also cause some conversion of NO_x to HNO₃.

The first experiment with full chemistry (CTMFULL) starts on 1 January 2007 and lasts until 30 April 2007, while the second experiment (CTMnoPSC) starts in parallel from 0000 UTC on 10 January, to match the assimilation period. Background aerosols (1999 values) are included in both the CTMFULL and CTMnoPSC runs, and only the three types of PSCs are switched off in the latter. O₃ differences between CTMFULL and CTMnoPSC runs indicate O₃ loss due to PSCs.

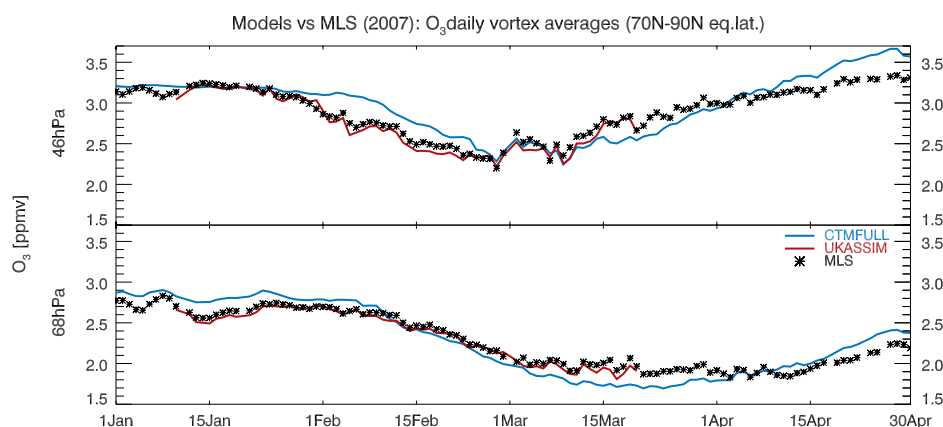


Figure 1. Measured and modelled vortex-averaged O₃ at the MLS pressure levels 46 and 68 hPa. In this figure and the following figures, the blue line refers to the Oslo CTM2 simulations, the red line to the UKMO assimilation. MLS measurements are represented by black asterisks. 46 hPa potential temperature ranges from 480–520 K, while at 68 hPa it ranges from 430–470 K.

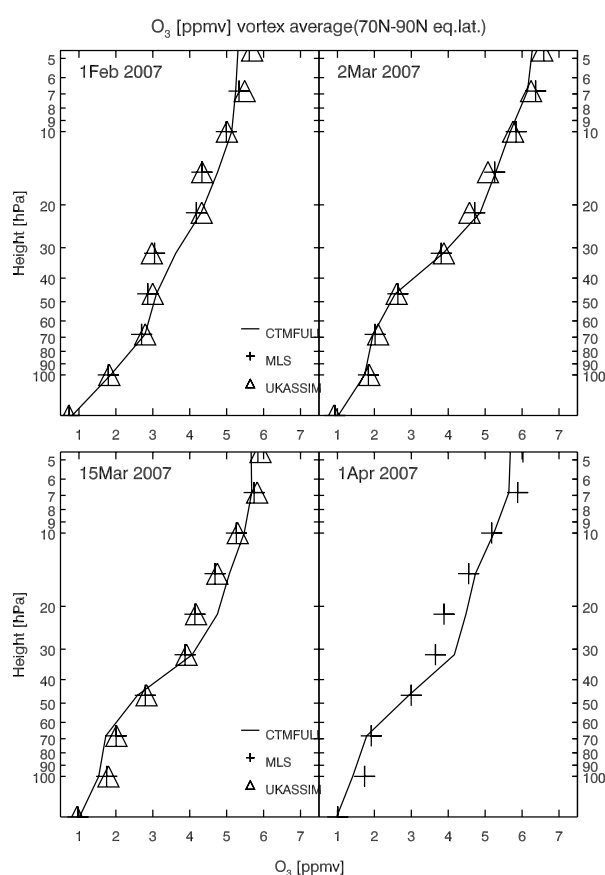


Figure 2. Modelled vortex-averaged O₃ as vertical profiles at MLS pressure levels for dates given in the figure. The Oslo CTM2 is represented by solid lines, the assimilation by triangles and the MLS measurements by plus signs. There are no assimilation data for 1 April.

The passive tracer (CTMPASS) also starts at 0000 UTC on 10 January 2007, so that differences between CTMnoPSC and CTMPASS can be assessed. These should be similar as long as the vortex O₃ loss is only due to heterogeneous PSC processes. To diagnose transport differences between the UK model and the Oslo CTM2 caused by differences in winds and transport schemes, we also interpolate the UK assimilated field from 10 January into the Oslo CTM2 grid and transport this field with the CTM passively (referred to as CTMUKREF). CTMUKREF is then compared with

UKREF. At the time when UKREF starts, the CTMFULL O₃ is very similar to UKASSIM (not shown).

All simulations are listed in Table I, and how we use these simulations for inferring O₃ losses is detailed in Table III.

4. Reproducing satellite measurements

Simulated and assimilated O₃ are compared with O₃ observations from MLS (V2.2) at standard pressure levels. Completing 14 orbits per day (ascending and descending), each day comprising 3495 vertical profiles, the instrument provides daily, near-global measurements. The vertical resolution is near 3 km in the lower stratosphere. The accuracy of retrievals in the lower and middle stratosphere is 3–10% for O₃ (Froidevaux *et al.*, 2008). In comparisons with the model, we only select MLS O₃ data described as valid by Froidevaux *et al.* (2008).

Within the closest hour and interpolated from the model's four closest grid points, the CTM was sampled at the times and locations of the MLS measurements to provide a profile-by-profile comparison. In this way, a daily average of profiles will take into account the asynoptic character of MLS observations.

For potential temperatures (θ) between 400 and 650 K, at 25 K intervals, the equivalent latitude (Nash *et al.*, 1996) was calculated in the Oslo CTM2. Daily vortex averages are constructed by selecting only profiles (corresponding CTM and MLS profiles) with equivalent latitude higher than a certain value. We use a restrictive limit at the vortex edge, 70°N, and discuss the effect of this choice. Typically, this produces about 300 profiles inside the vortex each day. For the assimilation, where the data available are snapshots at 1200 UTC every day, 'daily-like' vortex averages are generated slightly differently: from the selected MLS profiles (according to CTM equivalent latitude), we retrieve the geographical longitudes and latitudes to select the corresponding assimilation grid boxes. In this way, the same profiles are used for both models, even though the assimilated vortex may differ slightly from the Oslo CTM2 due to the different meteorology.

4.1. Time series and vertical profiles of vortex-averaged O₃

Vortex-averaged time series of O₃ on two MLS pressure levels are shown in Figure 1 for MLS observations (stars),

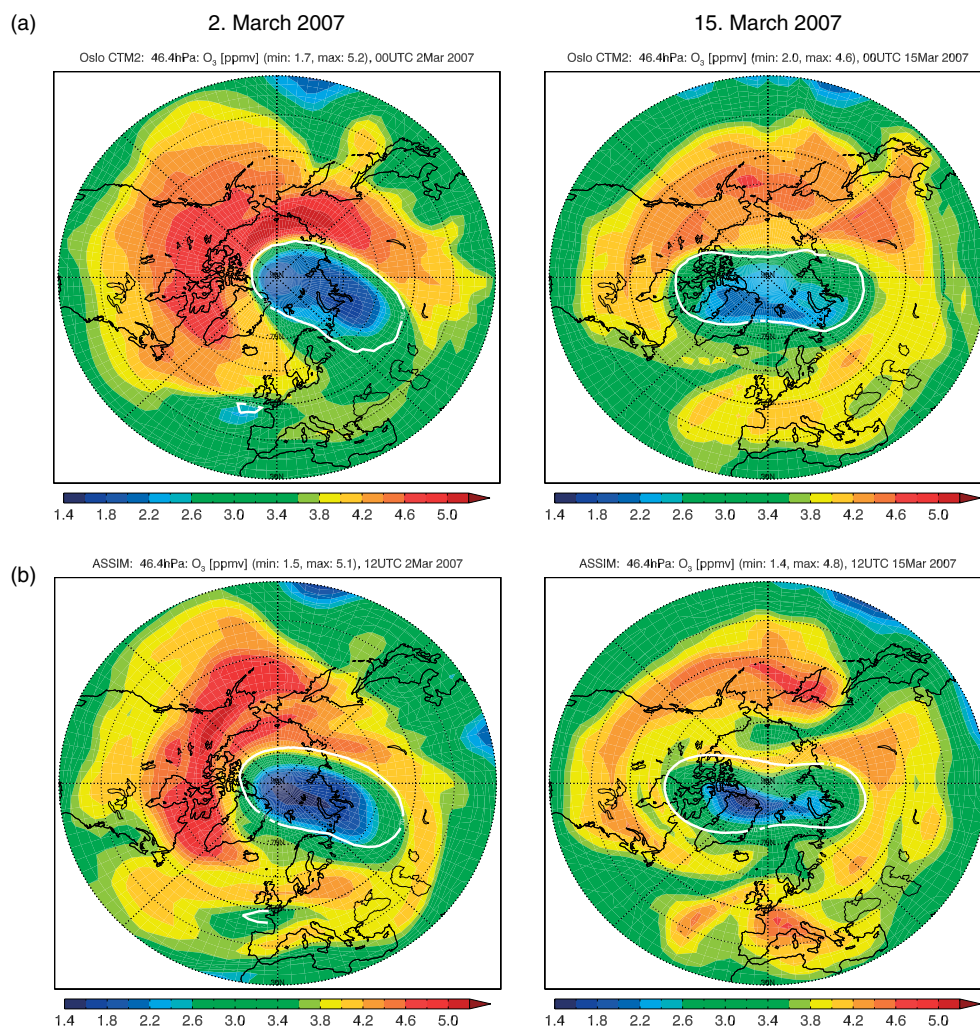


Figure 3. Modelled and assimilated O_3 at 46 hPa on 2 and 15 March. (a) Oslo CTM2, (b) assimilated O_3 .

CTMFULL simulation (blue line) and the assimilation run UKASSIM (red line). UKASSIM follows MLS very closely for both pressure levels, indicating that the assimilation method performs accurately. Also, CTMFULL follows MLS closely, although it slightly overestimates values at 46 hPa in February and at 68 hPa in January. In March the CTM underestimates values at both levels until the beginning of April where it again slightly overestimates.

Vertical profiles of the vortex-averaged O_3 are shown in Figure 2 for specific dates throughout the winter. Both the Oslo CTM2 and the assimilation reproduce the measurements very well at all levels. The fact that the Oslo CTM2 performs well indicates that the most important processes for transport and chemistry are included.

4.2. Horizontal distribution of O_3

Maps of O_3 modelled from the CTMFULL simulation (at 0000 UTC) and UKASSIM (at 1200 UTC) are shown in Figure 3(a) and (b), respectively, for 2 and 15 March 2007.

The 70°N equivalent latitude is shown on the maps, and since the models are driven by different meteorology they differ slightly. Due to the slight temporal displacement, the assimilation is somewhat rotated eastwards.

On 2 March, the O_3 levels in and near the vortex are very similar in both CTMFULL and UKASSIM. On 15 March, the CTMFULL O_3 is slightly higher in the inner vortex than

in UKASSIM, whereas closer to the vortex edge the CTM is lower. This explains why the CTM slightly underestimates the vortex average O_3 compared with UKASSIM at 46 hPa in Figure 1.

Compared with the assimilated O_3 , the CTM overestimates levels in the areas of O_3 maximum outside the vortex and at mid-latitudes. There is a remnant of processed and depleted O_3 over the eastern Atlantic on 2 March, which is slightly less prominent in the assimilation. On the other hand, low O_3 in intrusions from low latitudes is more apparent in the latter; for example, on 15 March one can see intrusions from low latitudes over the Atlantic and Pacific sectors, and the intrusion over East Asia (around 135°E and 60°N) seems stronger in the UKMO-driven assimilation.

Some of the differences between models in Figure 1 may originate from differences in transport, which will be discussed more closely in section 6.

5. Inferred O_3 loss

In Figure 4, we present vortex-average time series of all our reference fields at 46 and 68 hPa. The difference between the two CTM runs CTMPASS and CTMnoPSC indicates that the NO_x destruction of O_3 is clearly important; by mid-March the vortex average difference is ~ 0.5 ppbv at both levels, continuously increasing with the amount of sunlight in spring.

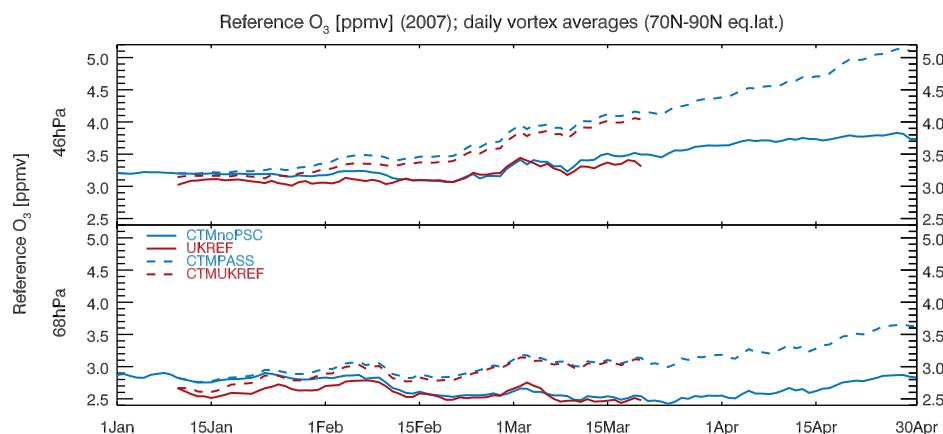


Figure 4. Modelled vortex-averaged O₃ references at the MLS pressure levels 46 and 68 hPa. The blue solid line is CTMnoPSC, the blue dashed line is the CTM2 passive tracer (CTMPASS), while the red solid line is the UK reference (UKREF) and the red dashed line is the UK initial reference transported by the Oslo CTM2 (CTMUKREF).

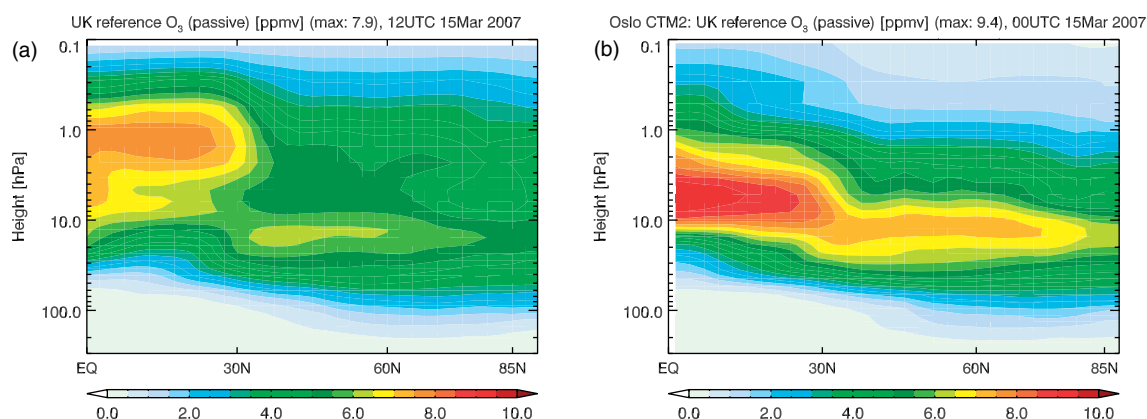


Figure 5. Zonal means of (a) UKREF and (b) CTMUKREF O₃ on 15 March.

The fact that CTMPASS and CTMUKPASS evolve very similarly confirms that on 10 January the O₃ is very well modelled in the Oslo CTM2.

UKREF and CTMnoPSC are remarkably similar. Since the latter includes NO_x chemistry and the winds differ, the similarity is accidental, due to excess vertical transport in UKREF compensating for the NO_x chemistry not included in UKREF. In order to get a first estimate of the errors in inferred O₃ loss from the data-assimilation method, we use both UKREF and CTMUKREF as reference O₃. From Figure 4 we see that there are clear differences in UKREF versus CTMUKREF, which should have been equal if transport was similar in both models. Their differences may be explained by too fast a Brewer–Dobson circulation in the UKMO analyses, as reported by Monge-Sanz *et al.* (2007). This is clearly illustrated in Figure 5, where we show zonal means for UKREF and CTMUKREF on 15 March. Whereas the starting fields were almost identical (not shown), the UK model contains more vigorous vertical transport, thereby diluting O₃ vertically. The faster vertical transport is mainly evident at low latitudes, but also dilutes the passive O₃ vertically at higher latitudes. This feature is also present in UKASSIM transport, but O₃ is corrected continuously by the O₃ assimilation. In other words, the UKREF is only suited for short simulations. A suitable length can be assessed from Figure 4, e.g. ~10% error in UKREF versus CTMUKREF. This would give a suitable length of about one month (in Jackson and Orsolini (2008) the assimilation period was 35 days).

In Figure 6, we show time series of the inferred daily vortex average O₃ losses at 46 and 68 hPa, using two references for each model, as explained in Table III. CTMFULL–CTMnoPSC provides our best estimate of O₃ loss from PSC-related processes. This loss is accidentally similar to the loss inferred by UKASSIM–UKREF, which both peak at 1.0 ppmv (30%) at 46 hPa, although delayed by a few days in the CTM. It may be that the CTM loss is prolonged, or that the UKASSIM–UKREF drops off because the passive O₃ is becoming too low due to transport effects (too little O₃ is transported into the vortex).

Because the data-assimilation method diagnoses all O₃ losses, it is better to compare UKASSIM–CTMUKREF with CTMFULL–CTMPASS, also plotted in Figure 6, where we see that the prolonged loss is not so prominent. These are also quite similar; at 68 hPa the former loss peaks at 1.2 ppmv (40%), whereas the latter peaks at 1.4 ppmv (41%), both in mid-March. At 46 hPa, the former loss peaks at 1.5 ppmv (40%) in early March, whereas the latter peaks at 1.6 ppmv (45%) a few days later.

Absolute maximum daily vortex average O₃ losses inferred from the simulations are presented in Table IV for 46 and 68 hPa. Results using various choices of vortex edge are also listed. Relaxing the vortex edge definition to 65°N produces a smaller vortex average O₃ loss, since the largest loss is in the vortex core. Similarly, increasing the edge to 75°N produces a higher average loss since the profiles are then in the innermost vortex. The modelled local maximum losses are found close to the vortex centre (not listed).

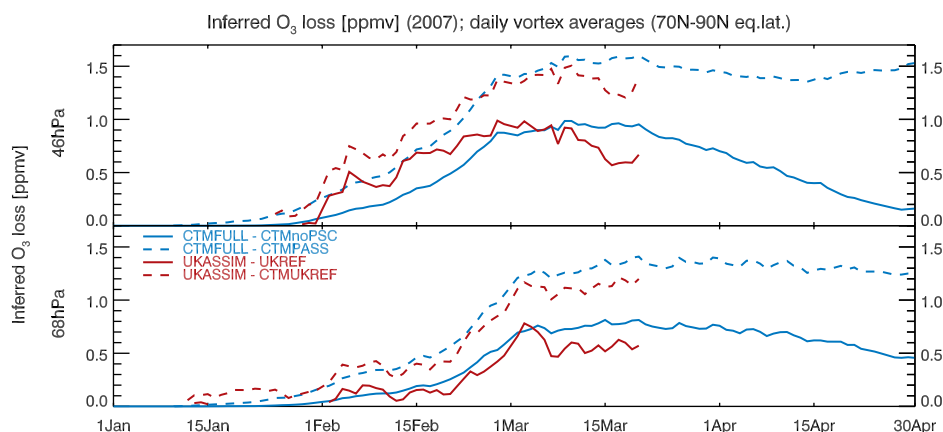


Figure 6. Inferred vortex-averaged O_3 loss at the MLS pressure levels 46 and 68 hPa. The blue solid line is CTMFULL–CTMnoPSC, the blue dashed line is CTMFULL–CTMPASS, while the red solid line is the assimilated loss UKASSIM–UKREF and the red dashed line is the UKASSIM–CTMUKREF.

Vertical profiles of the inferred vortex-averaged O_3 losses for 15 March, in percent, are shown in Figure 7 around the period of maximum loss. From these simulations, the estimated NO_x +PSC related O_3 loss at 46–68 hPa is 30–35% from UKASSIM–CTMUKREF and 35–45% for CTMFULL–CTMPASS. Losses due to PSC-related processes are estimated by the Oslo CTM2 to be 25–30% at this altitude; however, before March this amounts to up to 40%. PSC processes are confined below 20 hPa, and above 10 hPa there is O_3 gain due to too little O_3 coming in from low latitudes, where the passive O_3 is lower than normal since the production is shut off.

The O_3 gain in UKASSIM–UKREF can also be due to errors in the reference transport diluting extravortex O_3 vertically, giving too little O_3 coming from either above or through the vortex edge.

6. Discussion

By transporting the initial field of UKREF (UKASSIM on 10 January) by the Oslo CTM2 (in CTMUKREF), combining the advantages of having accurately initialized O_3 (from 7 days of O_3 assimilation) and a superior transport representation, we have shown that the transport in the two models differs substantially. One of the main differences in meteorology is that UKMO uses 3D-Var assimilation, while the ECMWF winds are produced with 4D-Var. This will result in transport differences between the models. In addition, the transport schemes of our models differ. The UKMO model uses a semi-Lagrangian transport scheme, more diffusive than the second-order moments scheme in the Oslo CTM2. Comparing UKREF with CTMUKREF, we have assessed the combined effect of different winds and transport schemes. Ideally, we would like to single out these two effects by running the Oslo CTM2 with UKMO winds, which as already explained has not been possible in this study.

In UKASSIM, however, the transport errors are masked due to O_3 assimilation. The large amount of MLS data largely controls UKASSIM O_3 , minimizing the impacts of potential deficiencies in transport in the UKMO assimilation system. UKASSIM is therefore virtually a globally gridded MLS field.

For CTMFULL to be similar to UKASSIM, the Oslo CTM2 would have to cover most of the important atmospheric processes. In fact, this is what we see, giving confidence in the CTM transport and chemistry and suggesting that

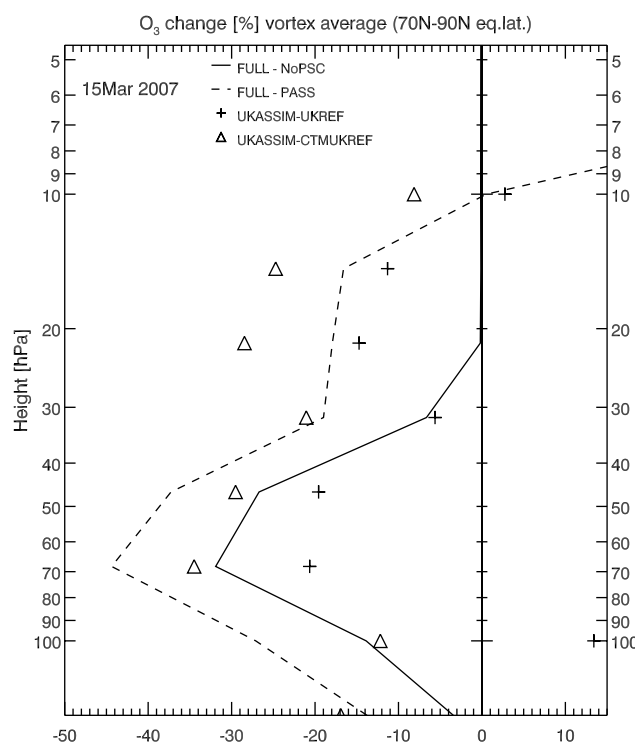


Figure 7. Inferred change in vortex-averaged O_3 as vertical profiles at MLS pressure levels for 15 March. Assimilation inferred loss UKASSIM–UKREF is shown by plus signs and UKASSIM–CTMUKREF as triangles. CTM losses are given as lines.

the Oslo CTM2 transport is more correct than the UKMO transport used here.

To study the transport differences more closely, we present in Figure 8 the MLS and modelled O_3 as a function of equivalent latitude, at 46 hPa for 2 and 15 March. The vertical dashed line denotes our choice of vortex edge. The horizontal gradients at 46 hPa are quite similar for all model runs. In all runs, including UKREF, the horizontal gradients are maintained at least until the beginning of March, indicating that the horizontal transport may be better represented. However, the difference between UKREF and CTMUKREF is about 0.5 ppmv at all latitudes north of 30°N, and greater at lower latitudes where the largest enhancement of vertical transport is located. This suggests that the enhanced vertical transport is a bigger problem in UKREF than is horizontal transport.

Table IV. Maximum daily vortex average O₃ loss (ppmv) at 46 and 68 hPa, for different equivalent latitude vortex-edge definitions. Model data are sampled as MLS, i.e. the values are based on profile-by-profile calculations.

Diagnosed simulations		46 hPa			68 hPa		
Name	Diagnosed processes	75°N	70°N	65°N	75°N	70°N	65°N
CTMFULL – CTMnoPSC	PSC	1.1	1.0	0.9	0.9	0.8	0.8
UKASSIM – UKREF	PSC + NO _x	1.2	1.0	0.9	0.9	0.8	0.7
CTMFULL – CTMPASS	PSC + NO _x	1.8	1.6	1.5	1.5	1.4	1.3
UKASSIM – CTMUKREF	PSC + NO _x	1.7	1.5	1.3	1.4	1.2	1.1

Were the vertical transport less vigorous in UKREF, extravortex air would be richer in O₃. The more diffusive scheme would then transport more O₃-rich air into the vortex, where it would increase the reference O₃. Too high a reference vortex O₃ in the assimilation method would overestimate the O₃ loss; however, due to the O₃ diluted extravortex air, the effect of diffusive cross-vortex transport is reduced.

Over the first few weeks of March, the vortex momentarily stretches out and almost splits in two, as can be seen in Figure 3 for 15 March. In addition to being stretched, the vortex edge crosses the North Pole, with the vortex centre close to it. Most globally gridded models will have problems resolving such a vortex properly in T42 resolution. This is likely why the Oslo CTM2 slightly weakens the gradient across the vortex edge in Figure 8(b), since it applies a polar grid box averaging in the transport scheme, a well-known treatment by CTMs.

As already noted, the Oslo CTM2 seems to catch both chemistry and transport in the CTMFULL simulation. There may, however, be errors in the CTM transport that could be offset by errors in heterogeneous processes. With a horizontal resolution of T42 there may e.g. be artificial diffusion/transport through the vortex edge. The amount of artificial diffusion should be studied in future, by increasing the horizontal resolution. It is, however, reasonable that artificial diffusion increases the more asymmetric the vortex is, due to the polar grid as described above, which could certainly be the case in mid-March. If such cross-vortex artificial diffusion is present in UKREF, to a large extent it will be masked by the already-mentioned O₃-diluted extravortex air. If there is high O₃ artificially transported into the vortex in all simulations, an O₃ loss estimate would be somewhat overestimated. Although we argue that for the assimilation method in this study CTMUKREF is a better reference than UKREF, it is worth noting that both UKREF and CTMUKREF may contain some artificial transport of extravortex high O₃ through the vortex edge.

The data-assimilation methods used (3D-Var for UK model and 4D-Var for ECMWF IFS) to determine the meteorological fields have different effects on the representation of transport in the analyses. In this study it has not been possible to disentangle the effects of the diffusive characteristics of the numerical schemes from the transport differences induced by misrepresentativeness of the ECMWF and UK assimilated winds. The transport scheme differences would be better diagnosed by driving the Oslo CTM2 by the UKMO winds, which has not been possible for technical and economic reasons. We have, however, assessed the combined effect of different meteorology and different transport schemes in the two models.

CIO overestimation leads to excess O₃ loss, and if modelled O₃ compares well with observations, overestimated CIO would indicate that the PSC scheme is too effective, and thereby counteracting artificial transport of high O₃ into the vortex. We have compared (but not shown) the CTM vortex CIO with MLS measurements (Santee *et al.*, 2008), also by selecting model profiles for MLS locations and times. In that comparison the Oslo CTM2 slightly overestimates CIO at 46 hPa in mid-March. The efficiency of the PSC-related chemistry and the amount of artificial diffusion into the vortex should be studied at higher resolution.

A way to reduce chlorine activation would be to reduce PSC surface area density (SAD). As PSCs are not transported in the Oslo CTM2, the PSC SAD could be slightly overestimated. This should be resolved in the future, and would allow PSCs to follow the vortex rather than being constantly formed at the front of an asymmetric vortex and evaporated behind. By assuming that droplets of supercooled ternary solutions (PSC1b/STS, described in Appendix A) are most important for halogen activation, we have tested a doubling of the STS radius, which does not change the vortex O₃ loss noticeably. Nonetheless, it should probably be allowed to grow.

Background aerosols also convert NO_x to HNO₃ and, as noted, we apply a background aerosol SAD from 1999, since there have been no major volcanic injections into the stratosphere since then. However, recent observations (Hofmann *et al.*, 2009) suggest that the background aerosol layer has been steadily increasing since 2000, probably due to anthropogenic sulphur emissions. So far our CTM NO_x levels do not indicate too low an aerosol SAD; however, the background aerosol distribution should be studied more closely to resolve this uncertainty.

Since the atmospheric gas chemistry is better known than the heterogeneous PSC chemistry, it can be argued that O₃ loss could be inferred from CTMnoPSC–UKASSIM. In our vortex averages, the CTMFULL and O₃ assimilation are very similar, although Figure 8 shows MLS and UKASSIM O₃ continuously decreasing towards the vortex centre. Using this approach instead of vortex averages, an inner vortex loss due to PSCs would then amount to 1.4 ppmv, as in Rex *et al.* (personal communication).

7. Conclusion

To improve the O₃ loss inference from the UKMO assimilation system, we have combined the assimilation with the use of the Oslo CTM2, comprising comprehensive tropospheric and stratospheric chemistry. We have found the O₃ assimilation to be virtually a gridded MLS field due to the large number of MLS measurements going into the assimilation system. The overall evolution of the

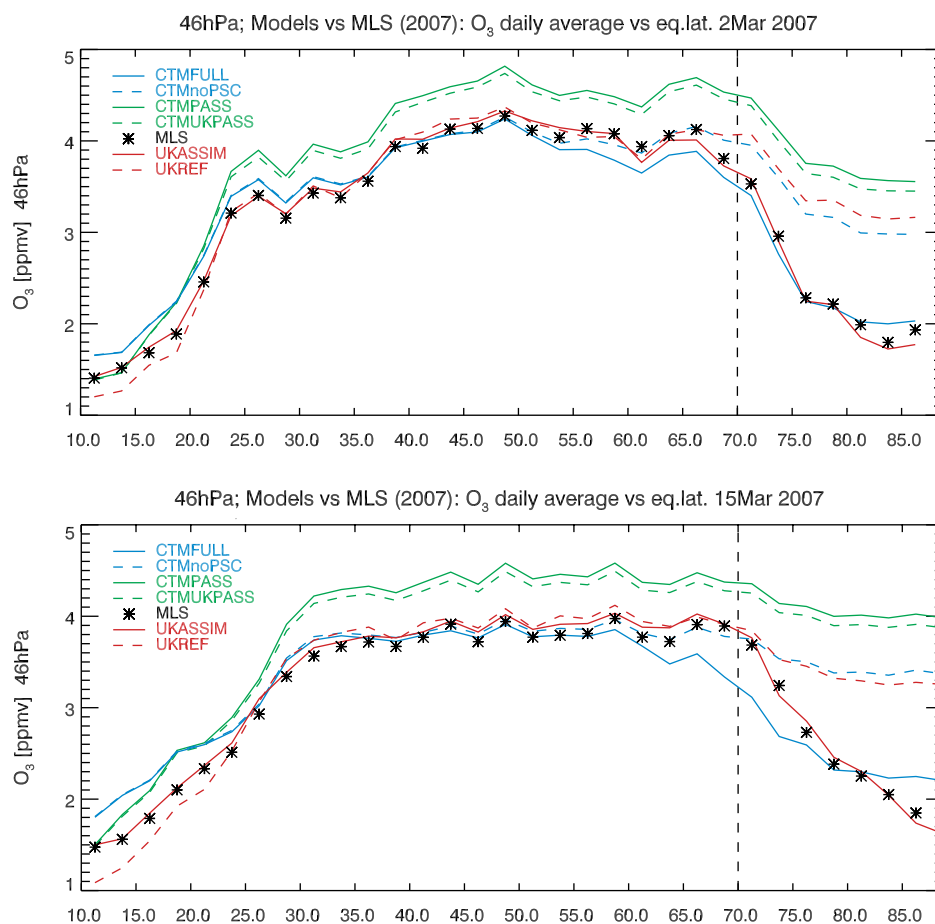


Figure 8. Modelled, assimilated and measured (MLS) daily average O_3 versus equivalent latitude at 46 hPa on (a) 2 March and (b) 15 March 2007.

vortex average O_3 in both the CTM and the assimilation is remarkably similar to that of MLS measurements, which gives confidence in the Oslo CTM2, although the inner vortex O_3 gradient is not caught by the CTM after mid-March.

We find the transport in reference UKREF to be too fast, vertically diluting the passive O_3 tracer, in agreement with Monge-Sanz *et al.* (2007). This may be due to either the 3D-Var assimilation or, to some extent, the transport scheme. The erroneous transport is not visible in the assimilation UKASSIM, because the O_3 assimilation corrects virtually all erroneous transport. The ability of the Oslo CTM2 to reproduce measurements indicates that both its chemistry and transport are good. We therefore suggest that for this study, transporting the UK reference field in the Oslo CTM2 provides a more realistic passive tracer for the assimilation method.

For the assimilation method, the two references provide a range of NO_x +PSC related O_3 loss between 0.8 (UKREF) and 1.2 ppmv (CTMUKREF) at 68 hPa, whereas the Oslo CTM2 produces 1.4 ppmv loss. At 46 hPa the respective values are 1.0–1.5 ppmv for the assimilation and 1.6 ppmv for the CTM. Both methods suggest O_3 loss peaking near 68 hPa or close above. Defining the vortex north of $70^\circ N$ equivalent latitude, the CTM-inferred loss peaks in early March at 46 hPa and in mid-March at 68 hPa, while the loss inferred from data assimilation peaks in early March.

Several studies have indicated that NO_x is important for O_3 loss in the vortex (Singleton *et al.*, 2005; Tilmes *et al.*, 2006; Konopka *et al.*, 2007), and from our CTM

simulations we find PSC-related O_3 destruction amounting to up to 0.8–1.0 ppmv between 46 and 68 hPa. NO_x is therefore responsible for up to 40% of the O_3 loss. This large contribution from NO_x means that a passive tracer is not a good reference for estimating PSC-related O_3 .

The assimilated loss estimates compare well with those in Rösevall *et al.* (2007), who assimilated Odin/SMR O_3 observations in an isentropic transport model, driven offline by ECMWF analyses. They found a vortex-averaged maximum loss of the order of 1 ppmv at 475 K, with a vortex edge defined at an equivalent latitude of $72^\circ N$. As in our assimilation-based method, they found maximum loss in early March. Using the MATCH approach, Rex *et al.* (personal communication) estimated the loss to be near 40% (1.4 ppmv) at 450–475 K. These slightly higher losses are Lagrangian estimates throughout winter that are not exactly equivalent to vortex average loss at a fixed pressure level. However, it should be noted that CTMnoPSC–UKASSIM would yield similar loss values for the innermost vortex O_3 loss due to PSCs, and a vortex average will yield O_3 loss similar to CTMnoPSC–CTMFULL, as shown in Figure 6.

Rösevall *et al.* (2007) also compared losses in 2006/2007 with those in 2004/05, and found that peak O_3 losses were comparable at 475 K but not as prolonged in 2006/2007. The losses inferred with our assimilation method are indeed comparable in both winters (Jackson and Orsolini, 2008) at the lower levels (450–500 K). In Rösevall *et al.* (2007) the losses at higher levels (525–575 K) were higher in 2006/2007 (around 1.2 ppmv), peaking in early April, and were probably induced by NO_x . We also find losses in excess

of 1 ppmv near 20 hPa by mid-March when the assimilation runs stop. This is also seen in the Oslo CTM2 (Figure 7).

Estimating O₃ loss via assimilation is a technique that is in its infancy and potential deficiencies remain. Jackson and Orsolini (2008) also pointed out issues related to smearing at the vortex edge and lack of dynamical balance in the 3D-Var analyses, and we find that the quality of the passively transported O₃ reference is important, not least the accurate representation of vertical hemispheric mean meridional circulation. We have significantly improved the inferred O₃ loss from assimilation by improving the reference O₃, as shown in Figures 4 and 6. In particular, we find this a possible explanation as to why Jackson and Orsolini (2008) underestimated O₃ loss compared with other studies.

For future assimilation studies, the vertical transport needs to be addressed, e.g. by using 4D-Var assimilation. Changing UK assimilation from 3D-Var to 4D-Var reduces excessive vertical transport while keeping the horizontal transport similar (B. Monge-Sanz, personal communication), in agreement with the O₃ gradients with equivalent latitudes calculated here. The diffusiveness of the transport scheme should also be diagnosed.

In our study, on a seasonal time-scale, using the Oslo CTM2 transport for the UK reference field provides consistent O₃ loss estimates for both models, which also compare well with other studies.

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A. Appendix: Oslo CTM2 microphysics

Microphysics and heterogeneous chemistry are treated according to Smyshlyaev *et al.* (1998), representing formation and evolution of PSCs including denitrification and dehydration through sedimentation. We distinguish between PSC1a (nitric acid trihydrate; NAT), PSC1b (supercooled ternary solutions; STS, a mixture of H₂O, HNO₃ and H₂SO₄) and PSC2 (ice H₂O, possibly coated with HNO₃). Treated separately from PSCs are the background aerosols (assumed to be binary solution of H₂O and H₂SO₄); they are taken from a satellite-derived (SAGE I, SAGE II, SAM II and SME) zonal mean monthly climatology produced by David Considine at NASA Langley, primarily based on Thomason *et al.* (1997). The microphysics is described by Søvde *et al.* (2008), but will be briefly described here, as two important improvements have been included.

The ternary solution of H₂O/HNO₃/H₂SO₄ is calculated based on Carslaw *et al.* (1995). If either HNO₃ or H₂SO₄ is absent, a binary solution may be the product. Since the Oslo CTM2 does not include the H₂SO₄ directly, the amount of H₂SO₄ in the stratosphere is calculated from the background aerosols (Søvde *et al.*, 2008).

Below a certain temperature the ternary solution aerosols are assumed to have STS/PSC1b properties. As the first improvement we now use T_{NAT} instead of a fixed temperature for this, calculated from the actual H₂O and

HNO₃ in the model. Ternary solutions have been found to freeze well below T_{NAT} (Voigt *et al.*, 2005), so we assume they stay liquid until a freezing temperature is reached (described by Søvde *et al.*, 2008). Once frozen, they will stay frozen until the melting temperature is reached.

If there is H₂SO₄ present in the PSC, the PSC was formed from background aerosols, from which H₂SO₄ is derived. It is therefore no longer available as background aerosol (i.e. background-aerosol SAD is zeroed). Above T_{NAT} , however, the calculated ternary solution aerosol will replace the background-aerosol SAD, simulating the uptake of HNO₃ on the sulphuric background aerosols.

Size distributions are log-normal and differ for the three PSC types. The second improvement in this study is the fact that NAT particles are allowed to grow – the mean of the log-normal distribution is allowed to change, while the others have a fixed distribution.

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