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Three-dimensional model study of the Arctic ozone loss in 2002/2003 and comparison with 1999/2000 and 2003/2004

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Abstract. We have used the SLIMCAT 3-D off-line chemical transport model (CTM) to quantify the Arctic chemical ozone loss in the year 2002/2003 and compare it with similar calculations for the winters 1999/2000 and 2003/2004. Recent changes to the CTM have improved the model's ability to reproduce polar chemical and dynamical processes. The updated CTM uses σ-θ as a vertical coordinate which allows it to extend down to the surface. The CTM has a detailed stratospheric chemistry scheme and now includes a simple NAT-based denitrification scheme in the stratosphere.

In the model runs presented here the model was forced by ECMWF ERA40 and operational analyses. The model used 24 levels extending from the surface to ∼55 km and a horizontal resolution of either 7.5°×7.5° or 2.8°×2.8°. Two different radiation schemes, MIDRAD and the CCM scheme, were used to diagnose the vertical motion in the stratosphere. Based on tracer observations from balloons and aircraft, the more sophisticated CCM scheme gives a better representation of the vertical transport in this model which includes the troposphere. The higher resolution model generally produces larger chemical O_3 depletion, which agrees better with observations.

The CTM results show that very early chemical ozone loss occurred in December 2002 due to extremely low temperatures and early chlorine activation in the lower stratosphere. Thus, chemical loss in this winter started earlier than in the other two winters studied here. In 2002/2003 the local polar ozone loss in the lower stratosphere was ∼40% before the stratospheric final warming. Larger ozone loss occurred in the cold year 1999/2000 which had a persistently cold and stable vortex during most of the winter. For this winter the current model, at a resolution of 2.8°×2.8°, can reproduce the observed loss of over 70% locally. In the warm and more disturbed winter 2003/2004 the chemical O_3 loss was generally much smaller, except above 620 K where large losses occurred due to a period of very low minimum temperatures at these altitudes.

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1 Introduction

In recent years, three-dimensional (3-D) models have been widely used to study the behaviour of ozone and other constituents during winter in the Arctic and Antarctic (e.g. Brasseur et al., 1997). Many chemical transport models (CTMs), forced by meteorological analyses, can successfully reproduce the general features of the seasonal evolution of total column ozone. However, models still fail to reproduce many aspects of polar chemistry and transport (e.g. Krämer et al., 2003; Stowasser et al., 2002). Moreover, given the large interannual variability in the Arctic it is important that any model is tested under different conditions.

Ozone loss in the Arctic stratosphere has received much attention over the past decade or so (e.g. see WMO, 2003). The significant year-to-year variability in Arctic meteorology leads to significant interannual variability in the polar ozone loss (Chipperfield and Jones, 1999). Thus, it is very difficult

Table 1. SLIMCAT model experiments.

Run	Resolution	Radiation Scheme	Dates	Initialisation	Passive ozone reset
ML	7.5°×7.5°	MIDRAD	01/01/1989–28/04/2004		1 January
CL	7.5°×7.5°	CCM	01/01/1989–04/04/2004		1 January
CH99	2.8°×2.8°	CCM	01/12/1999–19/04/2000	CL	1 December
CH02	2.8°×2.8°	CCM	01/12/2002–20/04/2003	CL	1 December
MH02	2.8°×2.8°	MIDRAD	01/12/2002–19/04/2003	ML	1 December
CH03	2.8°×2.8°	CCM	01/12/2003–19/04/2004	CL	1 December
MH03	2.8°×2.8°	MIDRAD	01/12/2003–05/04/2004	ML	1 December

to determine the chemical ozone loss since it is masked by dynamic variability caused by reversible vertical and horizontal advection and by mixing of air masses (e.g. Grooß and Müller, 2003). In the past, many models have tended to underestimate the chemical O_3 loss during cold Arctic winters (e.g. Hansen et al., 1997; Goutail et al., 1999 and Krämer et al., 2003). Some of these also appear to overestimate the chemical loss during warm winters (e.g. Guirlet et al., 2000). Previous studies also indicate that current CTMs cannot give a satisfactory observed partial column ozone loss (e.g. Rex et al., 2004). All these conclusions are based on the fact that the CTMs cannot reproduce the observed ozone. In fact, successful quantitative simulation of ozone loss depends critically on the realistic combination of horizontal and vertical transport, chemistry, radiative transfer and other processes which must be correctly represented in the models.

In this paper we examine the performance of our recently updated SLIMCAT model in the Arctic stratosphere. We use it to investigate the Arctic chemical ozone loss in 2002/2003 and compared it with the winters 1999/2000 and 2003/2004. Section 2 describes the updated model and experiments performed. The different meteorological conditions related to the polar ozone loss for the three winters are presented in Section 3. Section 4 shows our model results including sensitivity studies to the different radiation scheme and horizontal resolution. We also show how significant improvements were made in the new updated model when compared with observations especially for the cold Arctic winters 1999/2000 and 2002/2003. Section 5 summarises our conclusions.

2 Model and experiments

2.1 SLIMCAT 3-D CTM

SLIMCAT is an off-line 3-D CTM first described in Chipperfield et al. (1996). The original version of SLIMCAT used pure isentropic levels as a vertical coordinate and was thus limited to the domain above ∼330 K (∼9 km). The new version of SLIMCAT (Chipperfield, in preparation, 2005[1]) is now formulated using a hybrid σ-θ vertical coordinate, which means that it can extend down to the surface. Horizontal winds and temperatures are specified using meteorological analyses. Vertical advection is calculated from diabatic heating rates using a radiation scheme. The original SLIMCAT used the MIDRAD middle atmosphere radiation scheme (Shine, 1987) to determine the cross-isentropic flow since the lowest model vertical level was ∼330 K. In this case the lower boundary to the radiation scheme was 700 hPa where climatological upwelling long-wave fluxes are specified in order to model reasonable heating rates above ∼100 hPa. With the extension of the model to the surface the use of MIDRAD is likely no longer appropriate and we have added alternative radiation schemes including code based on the NCAR CCM (Briegleb, 1992). This scheme (hereafter called 'CCM') uses a δ-Eddington approximation (Joseph et al., 1976) and accurately computes absorbed solar radiation when compared to available reference calculations and observations (Briegleb, 1992). The downward and upward fluxes in the CCM radiation scheme can extend from 1000 hPa to the top of atmosphere.

Chemical tracers in SLIMCAT are advected using the scheme of Prather (1986) which conserves second-order moments. The model contains a detailed gas-phase stratospheric chemistry scheme (Chipperfield, 1999). The runs used here are based on photochemical data from JPL 2003 (Sander et al., 2003) with the exception of the absorption cross sections of Cl_2O_2 which were taken from Burkholder et al. (1990) extrapolated to 450 nm. The model also contains a treatment of heterogeneous reactions on liquid aerosols, nitric acid trihydrate (NAT) and ice (see Chipperfield, 1999). For the runs used here a simple NAT-based denitrification scheme was included in the model which assumes NAT forms in two modes. This is the same as the scheme described by Davies et al. (2002) and tested for the cold winter of 1999/2000.

[1] Chipperfield, M. P.: A new version of the TOMCAT/SLIMCAT off-line chemical transport model, Q. J. Roy. Met. Soc., in preparation, 2005.

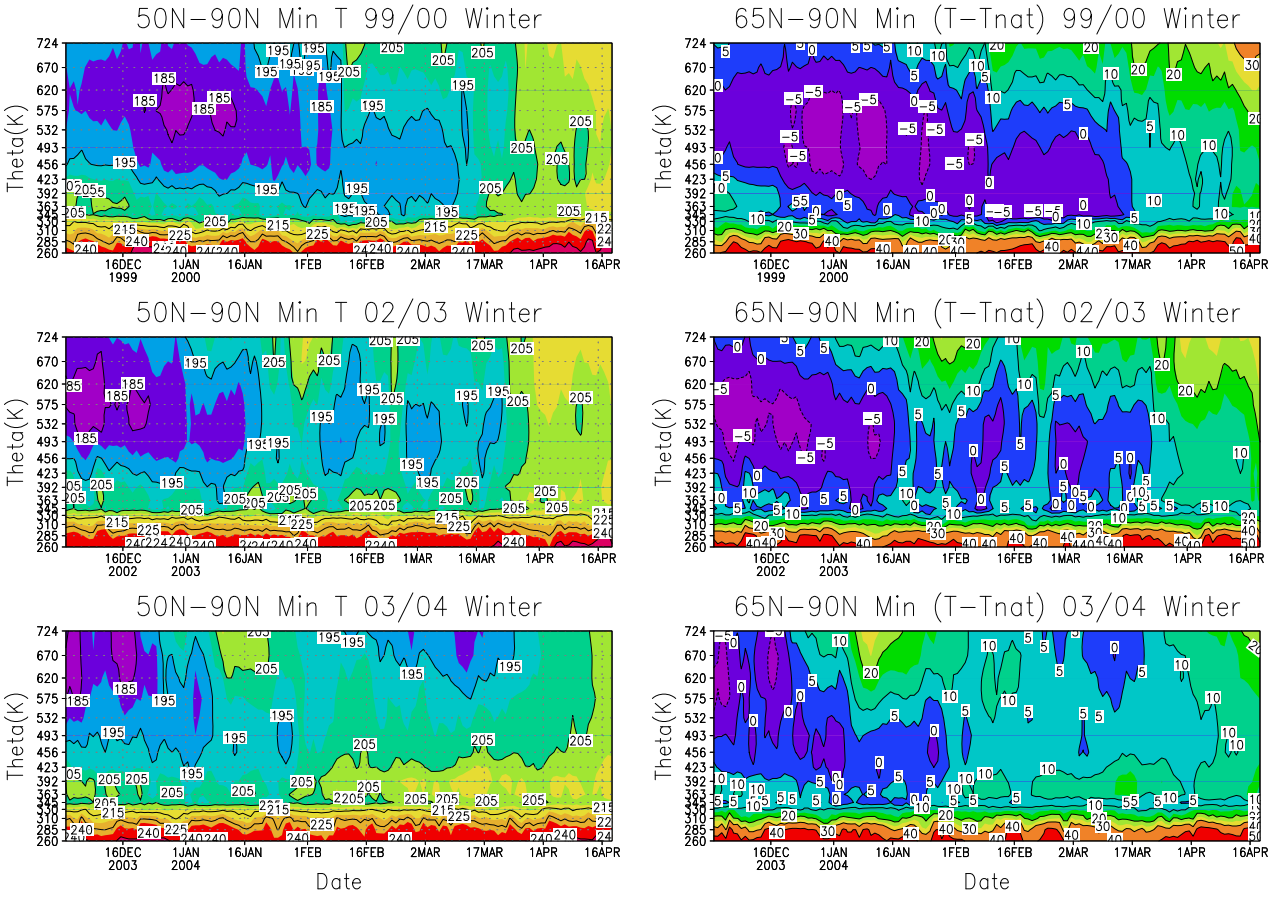

Fig. 1. The evolution of minimum temperature (K) northward of 50° N (left) and minimum difference between temperature and the equilibrium NAT formation temperature (T-T_{NAT}, K) northward of 65° N (right) as a function of time and θ in Arctic winters 1999/2000, 2002/2003, and 2003/2004. T_{NAT} was calculated based on model H_2O and HNO_3 using the expression of Hanson and Mauersberger (1988).

2.2 Experiments

A series of full chemistry model runs were performed to investigate the performance of the updated SLIMCAT model (see Table 1). First, in runs ML and CL, SLIMCAT was initialised on 1 January 1989 and integrated at low horizontal resolution (7.5°×7.5°) for ∼14 years using 6-hourly ECMWF analyses. These runs used ERA40 reanalyses from 1989 to 1999 and then operational analyses from 1 January 2000 onwards. The model used 24 levels from the surface to ∼55 km with a resolution in the lower stratosphere of ∼1.5–2 km. Run ML used the MIDRAD radiation scheme while CL used the CCM radiation scheme. The surface values of tropospheric source gases (CH_4, N_2O, halocarbons) were specified from WMO (2003) and an extra 100 pptv of chlorine and 6 pptv of bromine were assumed to reach the stratosphere from short-lived Cl/Br source gases (see WMO, 2003). The resulting halogen loadings in 2002 are around 3.7 ppbv Cly and 21 pptv Bry.

Output from these low resolution runs were taken and interpolated to a higher horizontal resolution (2.8°×2.8°) to initialise seasonal simulations for winters 1999/2000, 2002/2003 and 2003/2004. These seasonal runs were forced with ECMWF operational analyses throughout (runs CH99, CH02, MH02, CH03 and MH03, see Table 1). As well as providing the current best estimate of SLIMCAT modelled polar O_3 depletion for these winters, these experiments investigate how the model radiation scheme and horizontal resolution affect the calculation of chemistry and transport in the polar winter/spring.

3 Meteorology in the three Arctic winters

The evolution of minimum temperatures is frequently used to provide an overview of the times favouring PSC formation (e.g. Manney et al., 1994; Feng et al., 2005). Figure 1 shows time series of minimum temperatures north of 50° N between 260–724 K (surface to ∼28 km) from 1 December to late April in the years 1999/2000, 2002/2003 and 2003/2004.

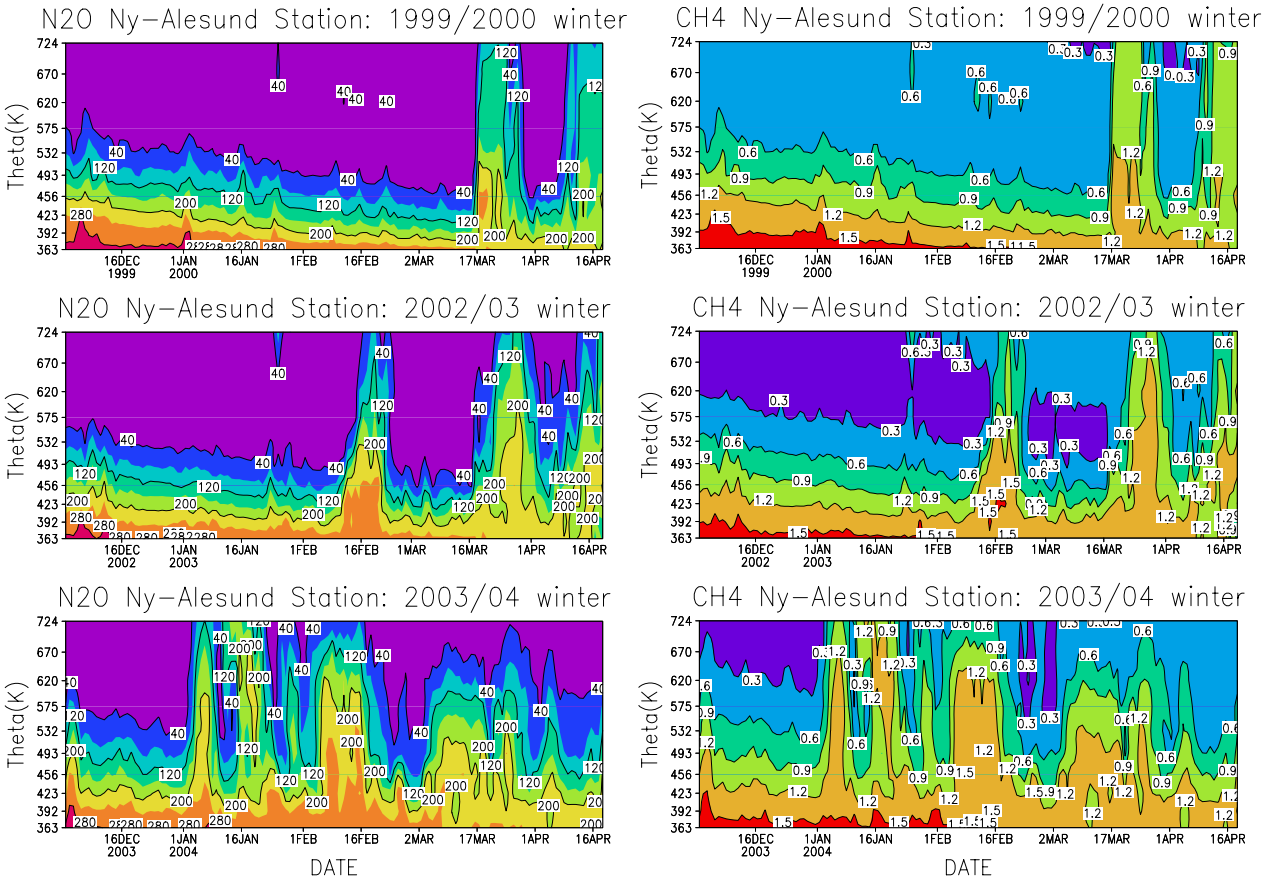

Fig. 2. Model N_2O (ppbv) and CH_4 (ppmv) at Ny-Ålesund station (79° N, 12° E) from SLIMCAT runs CH99, CH02, and CH03 (CCM, high resolution) as a function of time and θ for the three Arctic winters (top: 1999/2000, middle: 2002/2003, bottom: 2003/2004).

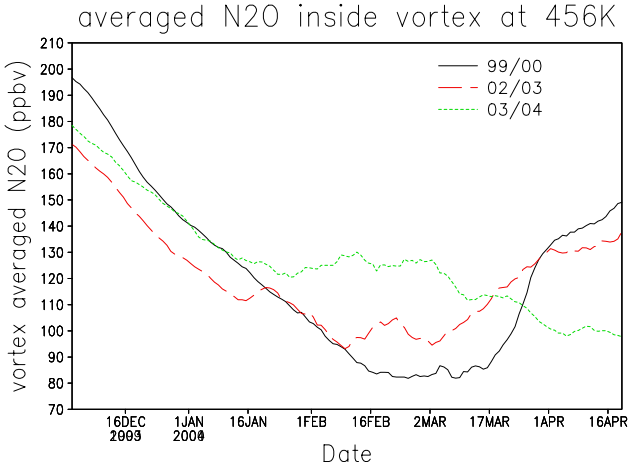

Fig. 3. Time series of the averaged modelled N_2O inside the polar vortex at 456 K from runs CH99, CH02, CH03 for Arctic winters 1999/2000, 2002/2003 and 2003/2004.

In the 1999/2000 winter, low temperatures ($T_{min} \leq 195$ K) occurred persistently above 450 K (∼17 km) and descended with time reaching below 400 K (∼14 km) in January. Minimum temperatures were below 185 K between 532–620 K (∼22–26 km) during the period from late December to mid-January. The final stratospheric sudden warming occurred in late March. The year 2002/2003 can also be classed as an extremely cold early Arctic winter (e.g. Naujokat and Grunow, 2003). Extremely low temperatures occurred from early December until mid-January and the coldest air around 6 December 2002 coincided with the area of an observed O_3 minihole around Scandinavia (e.g. http://www.ozone-sec.ch.cam.ac.uk/EORCU/Reports/wr0203.pdf). Temperatures increased in late January then decreased again in early February. A stratospheric minor warming occurred in mid-February followed by a further cooling in late February and early March. The final warming began in late March. The 2003/2004 Arctic winter was warmer than these other years. Low temperatures ($T \leq 195$ K) occurred from 1 December to early January above 460 K (∼18 km) and there was short cold period in late January. Another interesting point was the

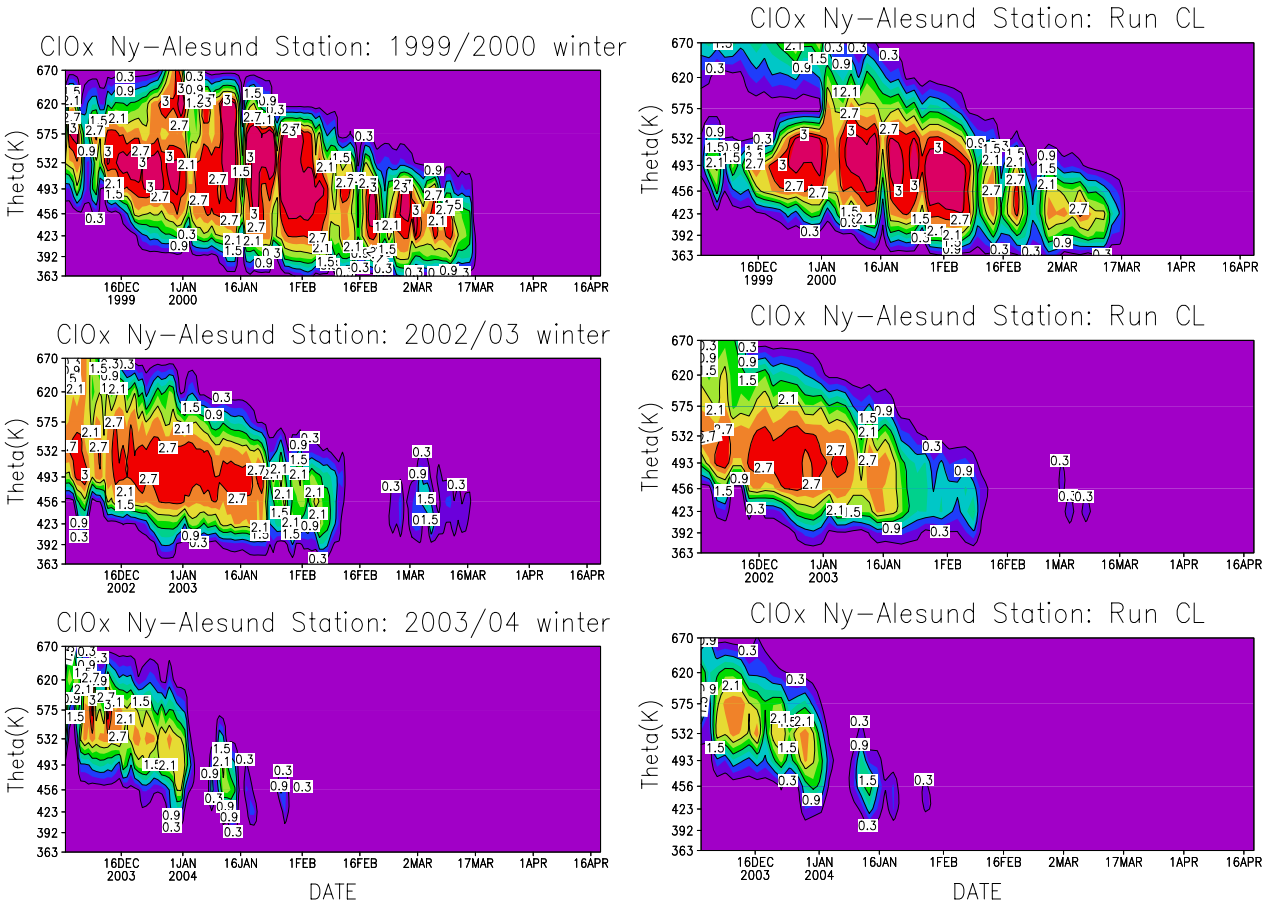

Fig. 4. Modelled ClO_x (=ClO+2Cl_2O_2) (ppbv) at Ny-Ålesund (79° N, 12° E) for the Arctic winters 1999/2000 (top), 2002/2003 (middle) and 2003/2004 (bottom). The left panels show results from the higher resolution runs (top run CH99 (CCM), middle run CH02 (CCM) and bottom run CH03 (CCM)) and the right panels show results from the low resolution multiannual run CL (CCM).

extremely cold period between mid February and mid March at higher altitudes above 620 K (∼26 km). The analyses from the U.K. Met Office (UKMO) (Swinbank and O'Neil, 1994) also captured the abnormal cold period for this winter (Y. Orsollini, personal communication, 2004).

Figure 1 (right) shows the difference between minimum temperature and the equilibrium NAT formation temperature (T_{NAT}) in the polar region. Negative values of T-T_{NAT} show the possible PSC occurrence as a function of time and altitude. For these three winters, the altitude of possible PSCs gradually descended with time after 1 December. Clearly, PSCs occurred extensively during the 1999/2000 Arctic winter – a large reduction in PSCs occurred only after mid March 2000 due to the stratospheric warming. In 2002/2003 the main period of possible PSCs occurred from December 2002 until mid January 2003. Other occasional PSC formation occurred around 8 February and 1 March 2003. However, much less PSC activity occurred in the winter 2003/2004.

Due to their long chemical lifetimes in the stratosphere nitrous oxide (N_2O) and methane (CH_4) can be used to study stratospheric dynamics. Their time evolution can provide information on the vertical and horizontal transport of air masses. Figure 2 shows the evolution of modelled N_2O and CH_4 (from the higher resolution seasonal simulations) at Ny-Ålesund station (79° N, 12° E) for the three winters 1999/2000, 2002/2003 and 2003/2004. Clearly, in 1999/2000 the vortex remained stable and located over Ny-Ålesund station for most of the winter. The disturbed vortex occurred only after 17 March 2000 due to the final stratospheric warming. In winter 2002/2003, the polar vortex was also stable before mid February but became more disturbed after that due to the minor stratospheric warming. The vortex returned over the station again from late February to mid March 2003 when the temperatures were again low. In contrast to the cold winters of 1999/2000 and 2002/2003, the polar vortex in 2003/2004 was much more disturbed for most of the time after January. Such a warm, disturbed polar vortex is not so conducive to large chemical ozone loss.

Figure 3 shows the time series of averaged N_2O inside the vortex at the 456 K isentropic level where there is strong

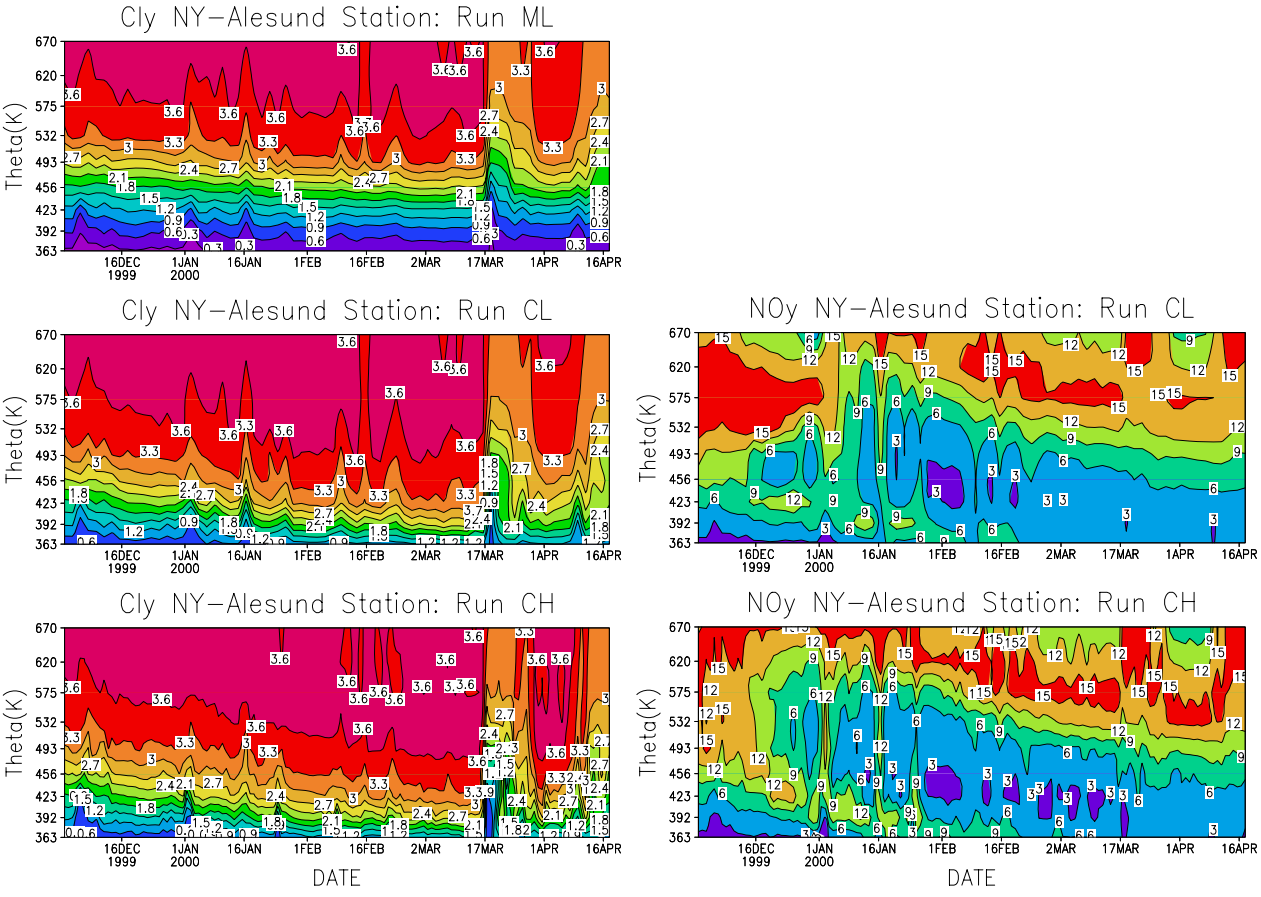

Fig. 5. Modelled Cl_y (ppbv, left) and NO_y (ppbv, right) at Ny-Ålesund (79° N, 12° E) for the Arctic winters 1999/2000 for run ML (MIDRAD, low resolution) (top – Cl_y only), run CL (CCM, low resolution) (middle) and run CH99 (CCM, high resolution) (lower).

vortex edge and large ozone loss for the three winters. Here the vortex is defined as the area enclosed by the 67° N effective latitude (EL) contour. The evolution of this averaged tracer is a combination of both descent and mixing at the vortex edge. Clearly, the descent in the 1999/2000 winter was stronger, and extended over a longer period, than other two winters due to the stable and cold polar vortex. N_2O values increased rapidly after 17 March 2000 due to the stratospheric warming and vortex break-up. For the Arctic winter 2002/2003, the N_2O values inside the vortex decreased rapidly in early December, increased after mid-January, and then decreased again until the vortex elongated and split from 13 to 25 Febrary 2003. There was less descent in the Arctic winter 2003/2004 than the other two winters during the period from mid January to mid March due to its warmer temperature. However, average N_2O continues to decrease from mid March to mid April.

4 Results

4.1 Chlorine activation: Effect of meteorology and model resolution

Chlorine activation plays a key role in the polar ozone loss (e.g. see WMO, 2003). Figure 4 (left) shows the modelled ClO_x (=ClO+2Cl_2O_2) at Ny-Ålesund as a function of time and θ for three years. Here the results are from the $2.8° \times 2.8°$ resolution model using the CCM radiation scheme. For the three winters, chlorine activation occurred from early December and activation gradually descended consistent with the corresponding low temperatures and likely PSC extent (Fig. 1). In the year 2002/2003, much more chlorine activation occurred in early December and even extended down to 400 K around 6 December. A more rapid deactivation followed the minor warming after mid February (not shown). Then, reactivation occurred during the period from late February to mid March 2003 due to the cooling of polar air masses and potential PSC occurrence. The chlorine activation on PSCs lasted longer in the year 1999/2000 due

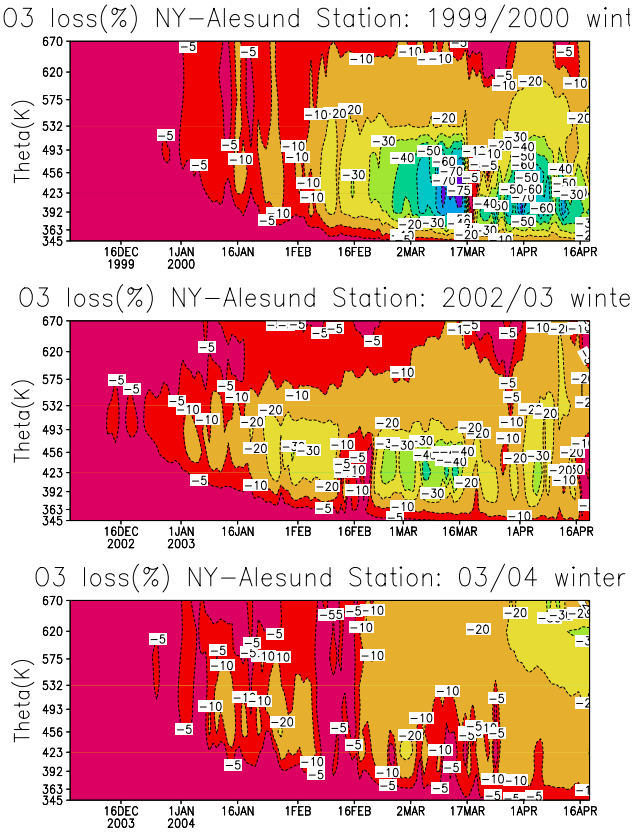

Fig. 6. As Fig. 4 (left), but for % local ozone loss.

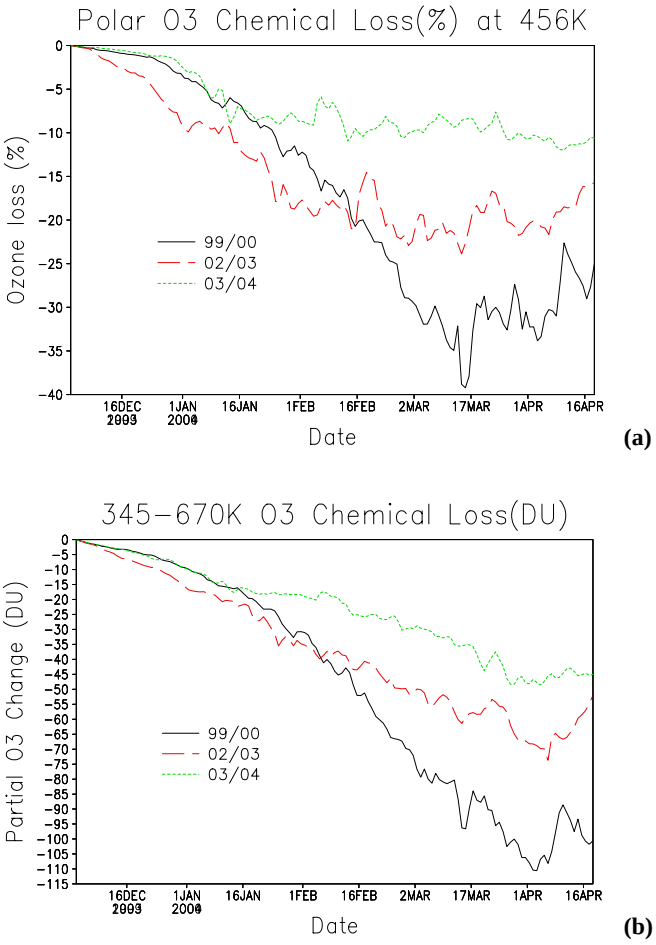

Fig. 7. Time series of averaged chemical ozone loss for **(a)** 456 K (%) and **(b)** 10 to 26 km partial column (DU) between equivalent latitudes 65°–90° N.

to the persisting stable polar vortex and lower temperatures. In contrast, in 2003/2004 the activation occurred over a much shorter period and at higher levels from early December until late January.

The right panels in Fig. 4 show the equivalent plots from the low resolution multiannual run CL. The features are generally similar to the higher resolution runs. The differences in ClO_x in December 1999 near 600 K are due to the different analyses used in the two experiments during this month: run CL used ERA40 analyses until 1 January 2000 while run CH99 used the operational analyses. Late on in this winter (i.e. early March) the ClO_x values in the higher resolution run persist with higher values. Figure 5 shows that the higher concentrations of ClO_x are partly due to higher Cl_y concentrations, caused by stronger descent and less mixing, and partly due to the stronger denitrification found in the higher resolution run.

The different chlorine activation largely determines the different ozone loss. Figure 6 shows the local ozone loss at Ny-Ålesund station for the three modelled winters. Larger ozone depletion occurred in the year of 1999/2000, aided by more denitrification which delayed chlorine deactivation to $ClONO_2$. Ozone loss in the warm 2003/2004 winter is generally much smaller, but abnormally large above 620 K due to the period of very low minimum temperatures above this altitude. In 2002/2003 the local maximum ozone loss at Ny-Ålesund was about 40% of the initial 1 December value before the final warming. Figure 6 also shows that early local ozone loss occurred in December due to more activation on PSCs and extremely low temperatures.

Figure 7 shows time series of averaged chemical ozone loss at 456 K and partial column ozone loss (∼10–26 km) between equivalent latitudes 65°–90° N. The averaged polar O_3 loss is less than the local ozone loss at Ny-Ålesund shown above. At 456 K there is an average ∼25% chemical O_3 loss by the end of March for 2002/2003 and about 10% ozone loss for 2003/2004, with no further ozone loss after mid February 2003. The maximum polar ozone loss in the year 1999/2000 reached about 40% at 456 K. The figure again shows the early chemical loss in December 2002 due to the low temperature and early chlorine activation. The partial column (∼10 to 26 km) ozone chemical loss reached 15 DU by the end of December and about 65 DU by the end of March in the winter 2002/2003. In the year 1999/2000, larger

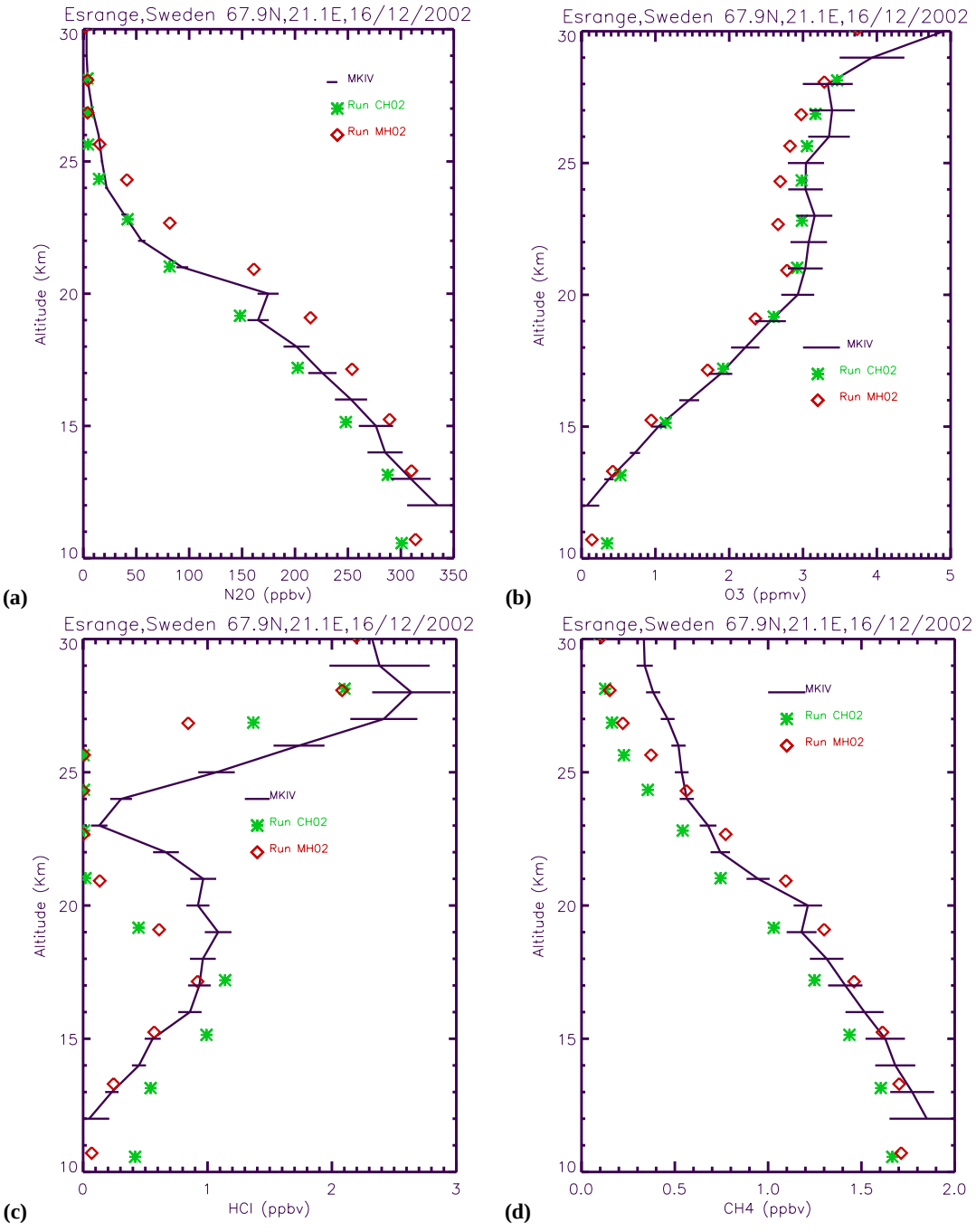

Fig. 8. MkIV balloon observations of **(a)** N_2O (ppbv), **(b)** O_3 (ppmv), **(c)** HCl (ppbv) and **(d)** CH_4 (ppmv) at Esrange (68° N, 21° E) on 16 December 2002 along with results from SLIMCAT model runs CH02 (CCM, high resolution) and MH02 (MIDRAD, high resolution).

chemical ozone loss occurred and reached about 100 DU by the end of March. While less chemical ozone loss occurred in the warm and more disturbed winter 2003/2004, the partial column ozone loss between 345 K–670 K is about 40 DU by the end of March 2004.

4.2 High latitude descent: Comparison with data for 2002/2003 and 1999/2000

The differences in the model results for runs ML and CL (for more details see Sects. 4.3 and 4.4), which differ only in the scheme used to diagnose vertical transport, clearly indicate the need to test these runs against tracer observations. Some balloon and aircraft observations are available for the

winter 2002/2003 and many more are available for winter 1999/2000.

Figure 8 shows profiles observed by the MkIV balloon instrument (Toon, 1991) on 16 December 2002 along with SLIMCAT output from the higher resolution simulations for 2002/2003 winter (run CH02 and run MH02). The profiles of long-lived species N_2O and CH_4 can be used to verify the modelled tracer transport. Downward transport is strongly underestimated by the SLIMCAT run using MIDRAD (run MH02) based on the N_2O profile comparison. The model run using the CCM radiation scheme (run CH02) gives more descent than run MH02. However, the modelled CH_4 from run CH02 has lower values than the observations, which may be caused by the lower model tropospheric CH_4. The surface values of tropospheric source gases (CH_4=∼1.76 ppmv, N_2O=∼316 ppbv) were specified from WMO (2003), and this determines the origin of the CH_4:N_2O correlation. With this boundary condition the model cannot reproduce both the observed CH_4 and N_2O simultaneously. Simulations using the more realistic CCM radiation scheme for diagnosing diabatic descent (run CH02) more accurately reproduces the observed ozone profile from the MkIV balloon than the MIDRAD scheme (run MH02).

Comparisons with in-situ aircraft observations during 2002/2003 also show similar results. Figure 9 shows an example of one M55 Geophysica flight for 26 January 2003. The Geophysica aircraft flew at between ∼18 km and ∼20 km, corresponding to ∼450 K and ∼500 K. The aircraft dived to ∼14 km when it reached northward of 75° N. The operational ECMWF temperatures used in the model compare well with observations. The transport, in particular the descent of air masses, is well reproduce by the model using CCM radiation scheme (run CH02) when compared with the observed N_2O measured on board the M55 by the HAGAR instrument (Volk et al., 2000; Riediger et al., 2000). Similar results from other flights comparisons (not shown here) also indicate that the model run using MIDRAD radiation scheme (run MH02) underestimates the descent of air masses. Reasonable diabatic descent also gives a better simulation of ozone. Figure 9 also shows that ∼0.5–0.7 ppmv ozone loss occurred at 18 km and ∼0.6 ppmv loss at 20 km from run CH02. Less ozone loss is diagnosed from run MH02; different vertical descent of air masses in the polar vortex can significantly affect vertical transport of ozone.

Figure 10 shows the evolution of the averaged N_2O inside the vortex for 2002/2003 Arctic winter/spring using MIDRAD, the CCM radiation scheme and their relative difference. Obviously, the structures of modelled N_2O inside the polar vortex are very similar when using the CCM and MIDRAD radiation schemes (e.g. the sharp gradient in modelled N_2O after mid Februrary due to the split of vortex and intrusion of midlatitude air). The relative modelled N_2O difference between using CCM (run CH02) and MIDRAD (run MH02) radiation schemes can be used to show when and where the two runs diverge over the course of the winter.

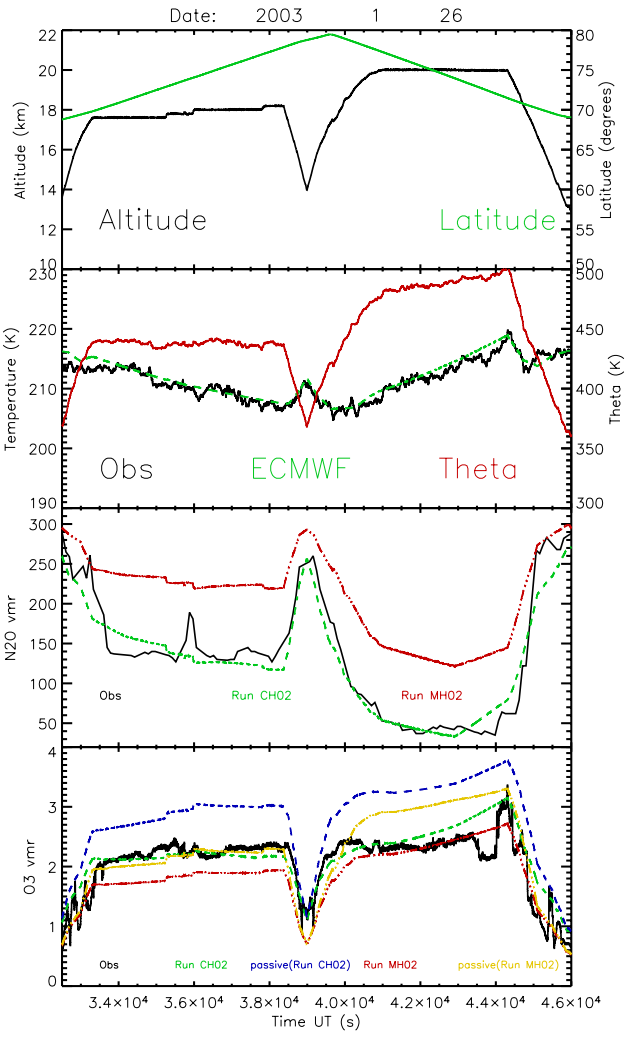

Fig. 9. Observations from the M55 Geophysica flight of 26 January 2003 compared with SLIMCAT model runs. **(a)** Latitude and altitude of flight track. **(b)** Observed and model (ECMWF) temperature and calculated θ. **(c)** Observed HAGAR N_2O (ppbv) (solid black line) and model results from runs CH02 (CCM, high resolution) and MH02 (MIDRAD, high resolution). **(d)** Observed FOZAN O_3 (ppmv) (solid black line) and model O_3 and passive O_3 (ppmv) from runs CH02 (CCM, high resolution) and MH02 (MIDRAD, high resolution).

Clearly, there was difference between ∼425 K and ∼620 K after five days run, and the maximum difference reached about 80% relative to modelled N_2O value from run CH02. This also shows that overall the model run using the CCM radiation scheme (run CH02) gives more descent than run MH02 in the lower stratosphere.

Figure 11 compares the higher resolution model with the CCM radiation scheme with tracer data from winter 1999/2000. The data comes from the ER-2 flight of 11 March 2000 where the aircraft flew inside the polar vortex near the start and at the end of the flight (Konopka et al., 2004). The model reproduces the magnitude of observed N_2O and CH_4

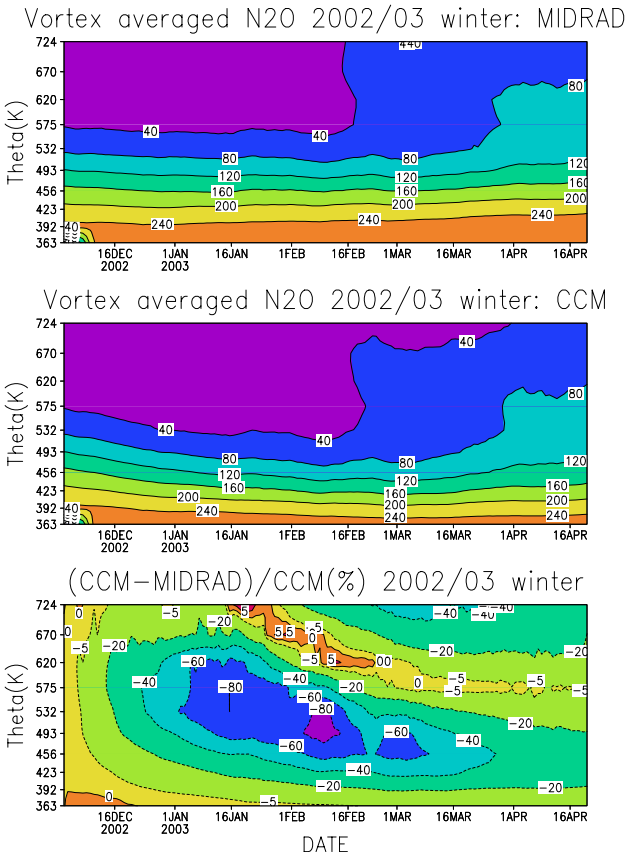

Fig. 10. Evolution of the averaged modelled N_2O (ppbv) inside the vortex and the relative difference (%) between the runs CH02 and MH02 for Arctic 2002/2003 winter/spring: top run MH02, middle run CH02 and bottom relative difference.

inside the vortex, as well as the gradient at the vortex edge. This supports the conclusion that this model setup gives a realistic representation of transport in the lower stratosphere.

4.3 Winter 1999/2000: comparison with ozone sondes

As a number of updates have been made to the SLIMCAT model we also compare it with O_3 sonde observations in 1999/2000. Previous studies with SLIMCAT have shown a good agreement between the model and the observed large depletion in the lower stratosphere in this cold winter (Sinnhuber et al., 2000). These older runs used the purely θ-coordinate model and were forced by UKMO analyses. As discussed by Davies et al. (2002), although these simulations produce a realistic extent of chlorine activation (based on comparisons with in-situ aircraft data) this was partly due to denitrification caused by ice sedimentation which was facilitated by the erroneously cold UKMO temperatures for this winter. Similar simulations with ECMWF analyses for 1999/2000 did not give such large O_3 loss and the UKMO-forced model underestimated O_3 loss in previous cold winters (e.g. 1996/1997).

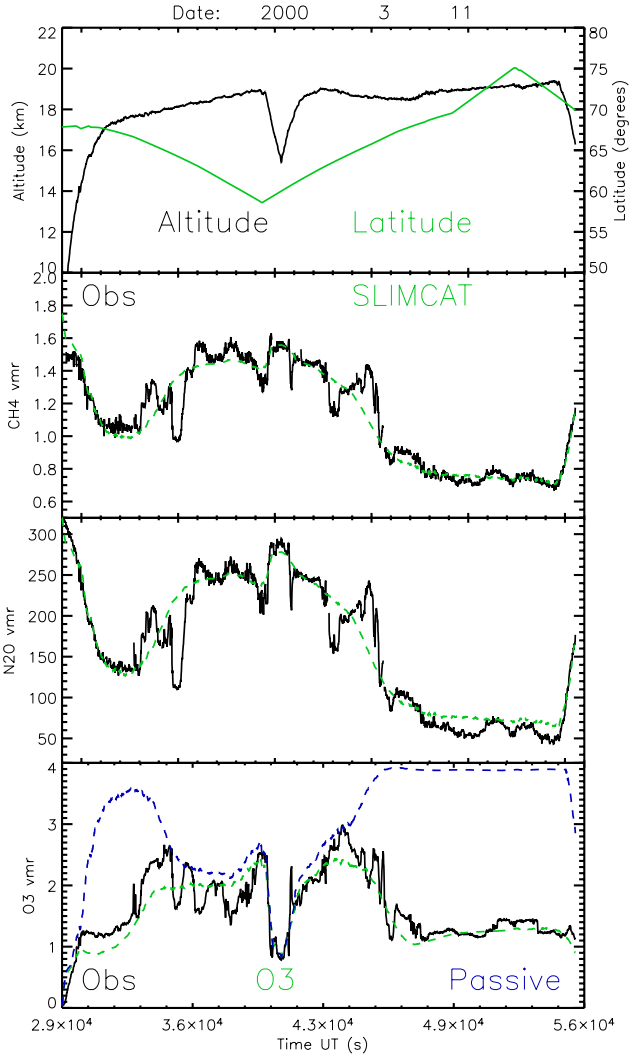

Fig. 11. Observations from the ER-2 flight of 11 March 2000 compared with SLIMCAT model run CH99 (CCM, high resolution). **(a)** Latitude and altitude of flight track. **(b)** Observed and modelled CH_4 (ppmv). **(c)** Observed and modelled N_2O (ppbv). **(d)** Observed and model O_3 (ppmv) along with passive O_3 (blue).

Figure 12 compares O_3 sonde observations at Ny-Ålesund with model runs for 1999/2000 (see Table 1). Clearly, the low resolution, multiannual run starting in 1989 using the MIDRAD radiation scheme (run ML) underestimates the observed O_3 at the start of the winter (e.g. by ~0.5 ppmv at 460 K and 425 K in December). Run ML gives a poor simulation for 1999/2000 Arctic winter. While the old (pure θ) version of SLIMCAT, with a lower boundary at 330 K was able to use MIDRAD successfully, the new model as used here extends down to the surface although MIDRAD was developed for use in the middle atmosphere only. MIDRAD cannot be expected to accurately calculate the upward radiative fluxes from the troposphere.

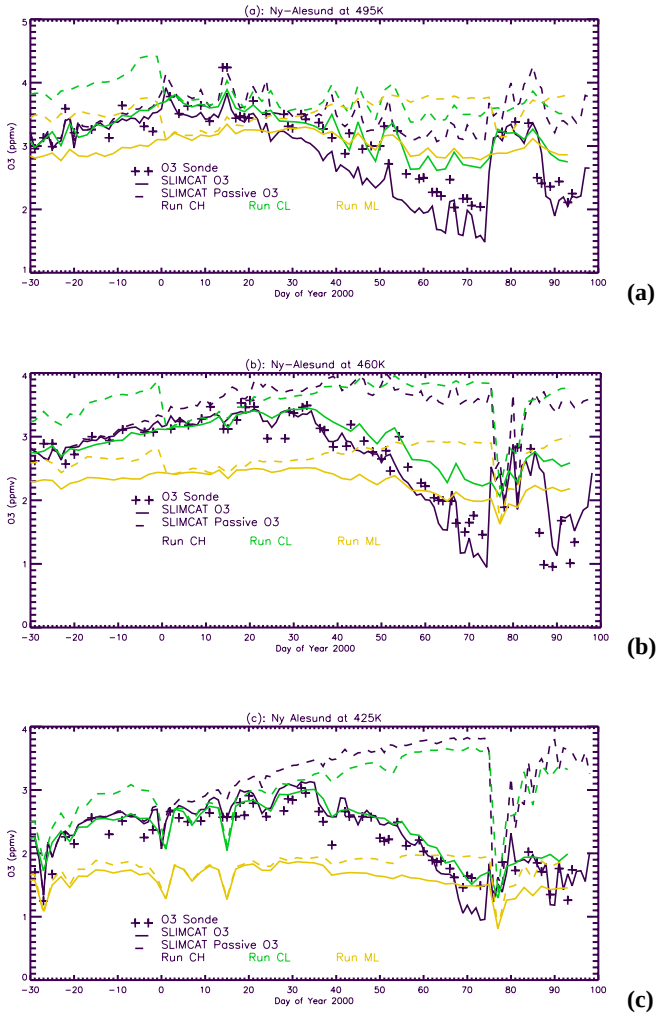

Fig. 12. Comparison of O_3 sonde observations (+ marks) at Ny-Ålesund for 1999/2000 with results for SLIMCAT runs ML (MIDRAD, low resolution), CL (CCM, low resolution) and CH99 (CCM, high resolution) for θ levels **(a)** 495 K, (**b)** 460 K and **(c)** 425 K. The dashed lines indicate the passive model O_3 tracer.

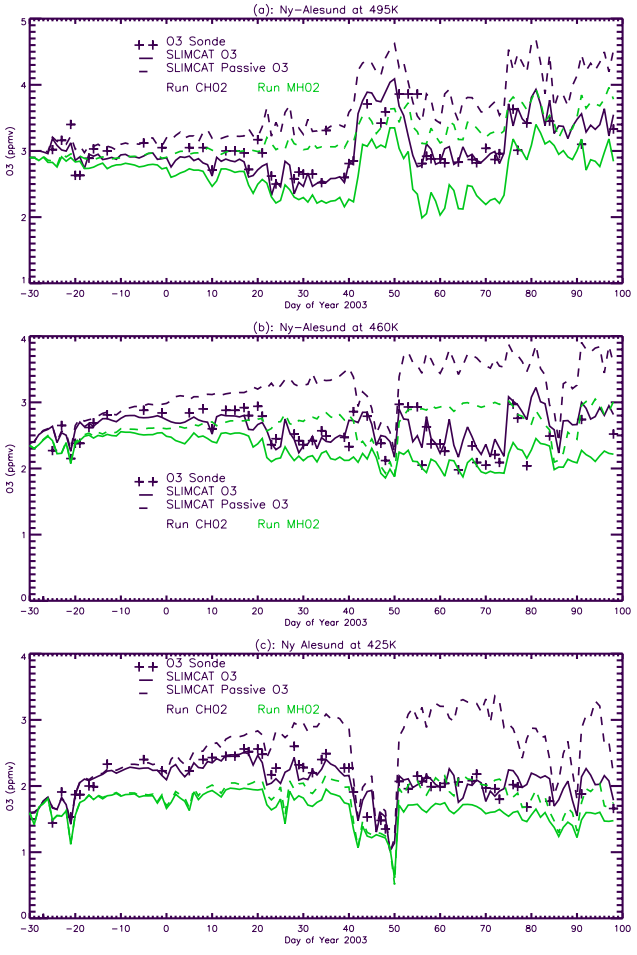

Fig. 13. Comparison between O_3 sonde observations (+) at Ny-Ålesund and results from the 2.8°×2.8° resolution SLIMCAT runs CH02 (CCM, high resolution) and MH02 (MIDRAD, high resolution) for winter 2002/2003 (top: 495 K, middle: 460 K, bottom: 425 K).

Figure 12 also shows results using the CCM radiation scheme. The O_3 simulation by the new version of SLIMCAT using the CCM radiation scheme (run CL) clearly gives better results than run ML. The calculated ozone in run CL reproduces the observations in early December 1999 (after ten years of spin up) and successfully reproduces the large observed decrease in ozone at 425 K. However, this low resolution multiannual simulation overestimates the observed O_3 above 450 K after late February. The figure also shows the model passive ozone tracer. This is reset equal to the chemically integrated O_3 in early winter (1 January for run ML and run CL, see Table 1) and then advected passively without chemistry. At any point and time after that the difference between this passive O_3 and the model's chemically integrated O_3 tracer is the net chemical O_3 loss. Run ML using MIDRAD calculates less polar ozone loss than run CL using CCM radiation scheme.

A further significant improvement is made by the higher resolution (2.8°×2.8°) seasonal simulation using the CCM radiation scheme (run CH99). The initialisation from run CL is good and the higher resolution run captures the decrease up to day 70 better and also has lower O_3 when the vortex comes back over the station around day 90. Overall, run CH99, (forced by ECMWF analyses) successfully reproduces the observed ozone in the lower stratosphere for the cold Arctic winter 1999/2000 (see also Sects. 4.1 and 4.2).

4.4 Arctic O_3 loss in 2002/2003 and 2003/2004: comparison with sondes

Figure 13 shows a comparison of the 2.8°×2.8° resolution SLIMCAT simulations with O_3 sondes at Ny-Ålesund for

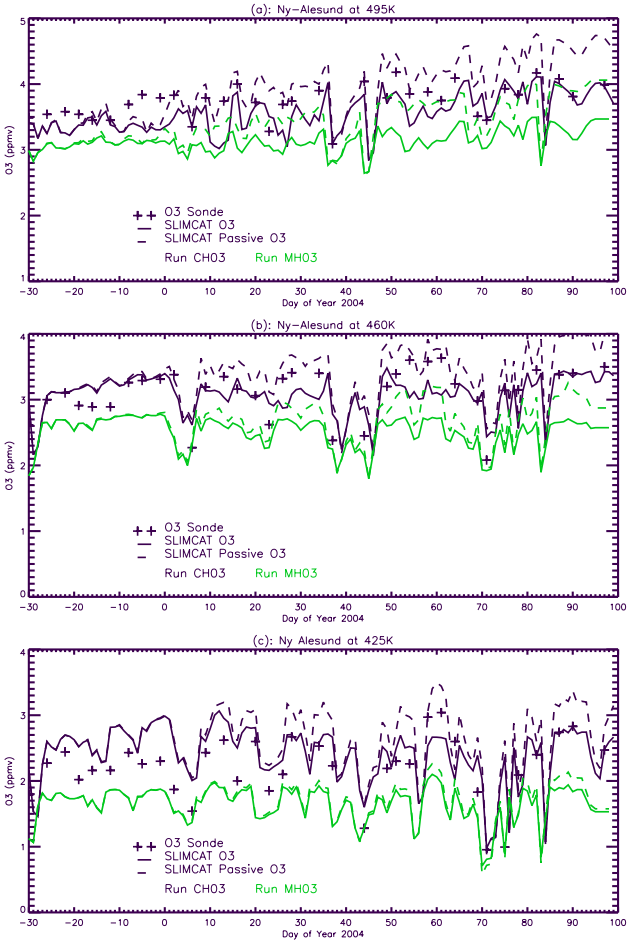

Fig. 14. Same as Fig. 13 but for Arctic winter 2003/2004. Here SLIMCAT runs CH03 (CCM, high resolution) and MH03 (MIDRAD, high resolution).

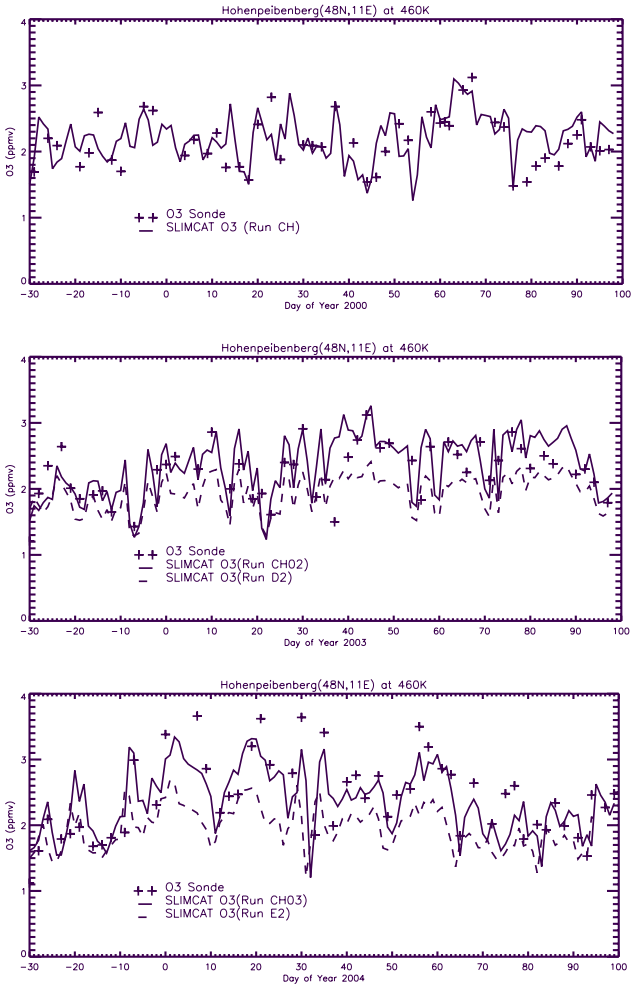

Fig. 15. Comparison of ozone sonde observations (+) at 460 K at Hohenpeißenberg (48° N, 11° E) with SLIMCAT 2.8°×2.8° resolution simulations for three Arctic winters (top: 1999/2000, middle: 2002/2003, bottom: 2003/2004). The solid lines use the CCM radiation scheme and the dashed lines use the MIDRAD radiation scheme.

Arctic winter 2002/2003. The results from the low resolution simulations (run ML and CL) are not shown here as the results are similar to Fig. 12. For this year there is only a small difference (<0.1 ppmv) in the early December ozone value between runs CH02 and MH02 (initialised from runs ML and CL, respectively) after 13 years run. However, the winter/spring evolution of ozone using different radiation schemes is again very different.

The run using the CCM radiation scheme (run CH02) is in good agreement with ozone observations for winter 2002/2003. The observed sudden change in ozone around mid February (day 45) and mid March (day 75) in the lower stratosphere is due to the stratospheric sudden warming and more disturbed polar vortex (see Figs. 1 and 2). The large observed decrease in ozone around mid February below 460 K (∼18 km) is due to the vortex moving away from Ny-Ålesund and is characteristic of mid-latitude air.

The higher resolution run CH02 still shows large loss (∼1.1 ppmv) in the lower stratosphere in the winter 2002/2003. Interestingly, earlier ozone loss in December 2002 also can be seen here if compared with 1999/2000 Arctic winter. Figure 8 shows that the Mk IV balloon instrument detected evidence for Cl activation on the flight of 16 December 2002 around 23 km. This supports the model results of activation and O_3 loss, although in the model the HCl removal extends over a deeper region.

Figure 14 shows the equivalent comparison of ozone in the 2003/2004 Arctic winter. The observed O_3 levels in 2003/2004 are larger than the other colder winters discussed here (i.e. 1999/2000 and 2002/2003 in Figs. 12 and 13). Overall, the higher resolution SLIMCAT simulation reproduces the observed O_3 levels in the lower stratosphere well for the warm and disturbed winter 2003/2004, but there is noticeable discrepancy around the period when the vortex

becomes more weak and disturbed (days 55 and 70). Model results show about 0.6 ppmv ozone loss at 460 K between 1 December and the end of March.

4.5 Comparison with midlatitude O_3 sonde data

For the 3 winter studies we show a brief comparison with ozone observations at mid-latitudes. Figure 15 shows comparison of O_3 between sonde observations and high-resolution simulations at Hohenpeißenberg (48° N, 11° E) for the three winters (see Table 1). The observed O_3 at middle latitudes is clearly larger in 2003/2004 than for the other two years. Significant changes in ozone were observed at Hohenpeißenberg in the three years at 460 K. However, the model shows that these are dynamical effects – the difference between the model O_3 and the passive O_3 (not shown) is still very small in this period.

5 Conclusions

We have used the recently updated SLIMCAT 3-D off-line CTM to study Arctic ozone loss in winter 2002/2003 and compare it with the very cold winter of 1999/2000 and the warm, disturbed winter 2003/2004. We have tested different radiation schemes in the model and performed experiments at different resolutions. These different radiation schemes and resolutions result in different tracer transport and polar ozone loss. For the new version of SLIMCAT used here, which extends down to the surface, the more detailed CCM radiation scheme produces more accurate tracer transport in the cold winters 1999/2000 and 2002/2003 and produces a better simulation in mid-latitude region. The higher resolution model gives more reasonable transport and mixing than the lower resolution.

The CTM results show that very early chemical ozone loss occurred in December 2002 due to extremely low temperatures and early chlorine activation in the lower stratosphere. Thus, chemical loss in this winter started earlier than in the other two winters studied here. In 2002/2003 the local polar ozone loss in the lower stratosphere was ∼40% before the stratospheric final warming. Larger ozone loss occurred in the cold year 1999/2000 which had a persistently cold and stable vortex during most of the winter. For this winter the current model, at a resolution of $2.8° \times 2.8°$, can reproduce the observed loss of over 70% locally. In the warm and more disturbed winter 2003/2004 the chemical O_3 loss was generally much smaller, except above 620 K where large losses occurred due to the period of very low minimum temperatures at these altitudes.

Overall, the best version of the updated model presented here gives a realistic representation of O_3 and inferred O_3 loss for a selection of winters. This is an advance over earlier versions of our model and other published studies, and shows that our ability to reproduce polar ozone loss is becoming more quantitative.

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