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Arctic Ozone Loss and Climate Sensitivity: Updated Three-Dimensional Model Study

M.P. Chipperfield and W. Feng

Institute for Atmospheric Science, School of Earth and Environment, University of Leeds, U.K.

M. Rex

Alfred Wegener Institute for Polar and Marine Research, Potsdam, Germany

We have used a three-dimensional (3D) chemical transport model (CTM) to investigate the variation in Arctic winter-spring chemical ozone loss from 1991-2003, and its observed correlation with low temperatures. The CTM (SLIMCAT) has been updated based on recent field and laboratory results and contains improved treatments of transport, chemistry and denitrification. In contrast to previous model studies, the CTM now gives a good simulation of the observed O_3 loss throughout the years studied. The model reproduces the large column loss of cold winters and also captures the shape of the ozone loss profile. The variation of O_3 loss with interannual variations in temperature is also well reproduced. Hence we show that for the first time a 3D stratospheric model is able to reproduce the past climate sensitivity of Arctic ozone depletion on temperature. This new capability is an essential prerequisite for model predictions of the future of the Arctic ozone layer in a changing climate. With realistic transport, chemistry-climate models with similar chemical modules should be able to reproduce polar chemical ozone depletion over the range of temperatures experienced so far.

1. Introduction

Since the discovery of the Antarctic ozone hole by *Farman et al.* [1985], the depletion of polar stratospheric ozone has been a primary focus of atmospheric research. Through measurements, laboratory work and modelling we have developed a clear qualitative idea of the processes responsible for the seasonal depletion of ozone in the Antarctic and Arctic [e.g. *Solomon*, 1999]. However, when compared to O_3 loss inferred from observations, there has been a persistent underestimation of the large losses in cold winters from a range of models including box/trajectory models [e.g. *Becker et al.* 1998; *Krämer et al.* 2003] and global models [e.g. *Hansen et al.*, 1997; *Goutail et al.*, 1999; *Guirlet et al.* 2000]. As such, our quantitative understanding is incomplete and this places limitations on the confidence of future predictions. This model deficiency has been routinely highlighted in recent international assessments of stratospheric science [*WMO*, 1999, 2003; *EC* 2001].

Recently, *Rex et al.* [2004] (hereafter R2004) published an analysis of the extent of O_3 depletion in the Arctic and demonstrated a remarkable correlation of column O_3 loss with the volume of the atmosphere with temperatures cold enough for the formation of polar stratospheric clouds (V_{psc}). Our basic understanding does suggest that colder Arctic winters will have more PSCs and hence more chemical O_3 loss, but this work showed an unexpectedly compact, almost linear relationship. R2004 also included calculations from one of the few three-dimensional (3D) CTMs able to study this long-timescale problem. This model, similar to other studies, underestimated O_3 loss in the coldest winters and hence severely underestimated the 'climate sensitivity' of the O_3 depletion by a factor 3. These results were another dramatic indication of the failings of models to reproduce the observed Arctic O_3 loss and received alot of attention as they cast doubt on the accuracy of future predictions of the polar stratosphere.

Tilmes et al. [2004] investigated the correlation of accumulated O_3 loss from December to March and V_{psc} using HALOE satellite observations. They confirmed the findings of R2004 of a linear correlation between accumulated O_3 loss and V_{psc}. They also found that if V_{psc} is averaged over the same time period as O_3 loss then solar illumination on the cold parts of the vortex does appear as a factor controlling O_3 loss. They also noted, as did R2004, that O_3 loss increased in response to the the eruption of Mt. Pinatubo in 1991.

The 3D CTM results used in R2004 were produced using our SLIMCAT 3D CTM around 5 years ago and were the first such decadal simulations. They were forced by UKMO analyses [*Swinbank and O'Neill*, 1994] and used a chemistry and polar stratospheric cloud (PSC) scheme essentially the same as that described in *Chipperfield* [1999]. Recently two major international field campaigns in the Arctic (THESEO 2000/SOLVE in winter 1999/2000 and VINTERSOL/SOLVE II in winter 2002/2003) contributed to advances in our understanding of the polar stratosphere. These campaigns, and other studies, have led to the continued improvement of stratospheric models. In this short letter we investigate how results from the current, updated version of the 3D SLIMCAT CTM, which takes account of these advances, improvements in model formulation and improved availability of meteorological analyses, now compares with the observations of R2004.

2. Model Experiments

We have used the SLIMCAT 3D CTM which is described in detail by *Chipperfield* [1999]. Here we have performed a simulation forced by European Centre for Medium-Range Weather Forecasts (ECMWF) data from 1977-2004 (ERA-40 reanalyses [*Uppala et al.*, 2004] until 1999 and then operational analyses) using a horizontal resolution of 7.5° × 7.5°. The model used 24 hybrid σ-θ levels from the surface to ∼55km (resolution of ∼2 km in the lower stratosphere). In the stratosphere, where the model uses pure θ levels, vertical motion is calculated using a version of the NCAR CCM radiation scheme [see *Feng et al.*, 2005]. The simulation used

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time-dependent source gas boundary conditions from *WMO* [2003] which included an estimated 100 pptv of chlorine and 6 pptv bromine reaching the stratosphere from short-lived halocarbons not explicitly included in the model, though assumed to be released with the same lifetime as CH_3Br. Accordingly the model stratospheric bromine loading around 2000 was around 21 pptv. The model had a simple representation of denitrification due to the sedimentation of large nitric acid trihydrate (NAT) [*Davies et al.*, 2002]. The model time-dependent aerosol loading was specified from a zonal mean dataset compiled from satellite observations (SAGE II in the period of interest here) [*WMO*, 2003]. Photochemical processes were taken from *Sander et al.* [2003], with the exception of the photolysis rate of Cl_2O_2 for which we used *Burkholder et al.* [1990] with a long-wavelength extrapolation to 450 nm as suggested by *Stimpfle et al.* [2004]. For 8 Arctic winters we also performed seasonal model simulations (December-April) at the higher horizontal resolution of 2.8° × 2.8°. These runs were initialised from output of the lower resolution run around December 1 of each year. *Feng et al.* [2005] have shown that for Arctic winter 2002/3 this corresponding seasonal run produces a good simulation of chemical and dynamical tracers.

3. Results

Figure 1 shows the comparison of averaged polar column O_3 depletion versus V_{psc} for the R2004 observations and our 3D CTM results. The model O_3 values have been averaged polewards of 67°N equivalent latitude (75°N in 2000 due to the smaller polar vortex in late March) and between 380 and 550 K. These equivalent latitudes are typical of the edge of the polar vortex in mid-late March deduced in the R2004 analysis of the observations. The model V_{psc} is an average over the same area between 400K and 550K from December 1 to March 31 of each year and was calculated from the model temperature, H_2O and HNO_3 fields. Table 1 gives the values plotted in Figure 1. The observations show a compact, linear correlation with cold winters (e.g. 1996, 2000) giving large column losses of around 100 DU. The figure shows the old 3D CTM simulation which underestimated the depletion in cold winters and overestimated the depletion in warm winters, and as a result underestimated the slope of the correlation (the 'climate sensitivity') by a factor 3. The new, updated model reproduces the observations much better, especially in the cold winters of the mid 1990s. Because of this, the slope of the linear fit to the new model now agrees well with the fit of the observations. The 2.8° resolution simulations result in a slightly larger ozone loss as the higher resolution is better able to maintain the enhanced levels of active chlorine inside the polar vortex. For certain cold winters the model loss at high resolution now exceeds the observations. Clearly there are a variety of factors which affect the model-observation agreement in any winter but it is significant that the model results now span the observations rather than always underestimating them.

Note that there are differences in the V_{psc} values calculated in R2004 and in the model. R2004 calculated V_{psc} using fixed profiles of HNO_3 and H_2O while for the CTM we used the model tracer fields, which vary during a season and interannually. These differences raise the question about the physical meaning of V_{psc} which is used to correlate with chemical O_3 loss. In effect, in R2004 it acts more as a surrogate for average temperature while in the model it is a estimate of NAT occurrence. R2004 also base their estimate of V_{psc} on old operational ECMWF analyses for the 1990s while our new CTM runs use ERA-40 data. An investigation of the interactions between different processes which give rise to this linear correlation is beyond the scope of this letter and is the subject of ongoing work.

Figure 1. Scatter plot of vortex-average column O_3 loss ($\Delta O3$) versus V_{psc} inferred from observations [R2004] from 1992 to 2000 (coloured circles). Also shown are average (see text) results from different SLIMCAT 3D model runs. The + marks show the old version of the model for 1992-2000. The small triangles show results from the updated, low resolution (7.5°) model run for 1991 to 2004. The large triangles show results from the higher resolution (2.8°) seasonal runs from 1995 to 2004. Also shown are the linear fits to the observations (red line), old model (green line), new low resolution run (black line) and high resolution run (blue line).

Figure 2 shows profile comparisons from the observations and new model runs for 8 Arctic winters. This shows that the modelled column depletion shown in Figure 1 occurs at the same altitude as the observed loss, giving more confidence in the quality of the new model. For example, in the cold winters of 1993, 1995, 1996, 1997 the model reproduces the observed loss of around 1-1.5 ppmv between 400 and 450 K. For 1996 the profile of O_3 loss from the old CTM run is also included, clearly showing that this run underestimated O_3 loss in the lower stratosphere. Other previous criticism of model simulations has been that they also overestimate the observed ozone loss in warm winters. This can be seen in the column comparisons of Figure 1 for the winters of small observed loss such as 1998 and 1999. However, Figure 2 shows that in terms of the profile, the model reproduces the small O_3 loss between 400 and 475 K well. The column discrepancies arise due to differences at higher altitudes in 1998, while for 1999 there is no data.

Table 1. Chemical ozone loss (ΔO_3 in DU) and V_{psc} values ($\times 10^6$ km^3) in Figure 1.

	R2004 Obs.		Old CTM		New CTM (7.5°)		New CTM (2.8°)	
Year	ΔO_3	V_{psc}	ΔO_3	V_{psc}	ΔO_3	V_{psc}	ΔO_3	V_{psc}
1992	51	8.3	44	6.6	40	7.9		
1993	67	18.0	51	15.1	60	14.9		
1994	50	7.7	55	5.9	47	6.9		
1995	78	25.0	45	21.1	73	21.7	99	27.2
1996	101	35.7	58	32.7	78	31.7	105	39.6
1997	56	16.7	42	17.1	68	17.3	86	23.7
1998	23	4.1	39	5.0	32	4.6	46	6.1
1999	18	1.1	35	1.1	22	0.2	28	0.4
2000[a]	95	38.4	72	36.3	75	24.2	126	39.5
2001					32	7.2	46	12.9
2002					23	1.0	30	1.3
2003	43	13.5			35	12.7	54	14.6
2004					29	1.5	34	1.6

[a] Note that for 2000 the new 2.8° run used ECMWF operational analysis while the 7.5° run used ERA-40.

Figure 2. Profiles of averaged O_3 and O_3 loss (ppmv) for 8 Arctic winters from 1993 - 2000 from observations and the new SLIMCAT runs. The left panels show the averaged profiles from the model (thin black line with + marks - low resolution, thick blue line with diamond marks - high resolution (1995-2000 only)) and R2004 observations (red line with no symbols) for March 25 (solid lines) and January 15 (dashed line). The January profiles have been descended using model heating rates from January to late March for direct comparison with the March profiles. The right panels show the calculated O_3 loss from January to March 25 (March 15 in 1998) from the model and observations (with same labelling). The dotted line in the O_3 loss panel for 1996 shows results from the old SLIMCAT run. The model profiles have been averaged polewards of 67°N equivalent latitude (75°N in March 2000) and the observations are averaged within the polar vortex.

The significant quantitative improvements in the model calculations shown in Figure 1, and confirmed by Figure 2, is the result of a number of complementary factors which all make a contribution. Some of these will apply to all models and some are specific to our CTM. The new simulation includes updates to ClO/Cl_2O_2 kinetics which result in faster loss due to one of the principal polar O_3 loss cycles, though this effect is expected to be comparatively small. The model also now has a treatment of denitrification by large NAT particles which allows the model to denitrify during the cold years of e.g. 1995, 1996 and 1997 which did not happen using the previous ice-based scheme [*Chipperfield*, 1999; *Davies et al.* 2002]. *Davies et al.* [2002] showed that in the cold winter of 1999/2000 this increased modelled O_3 loss by about 30%, which is probably an upper limit for this effect. The transport in our CTM has been improved by the change from the former MIDRAD radiation scheme to the CCM scheme now that SLIMCAT extends down to the surface. This change in radiation scheme results in stronger modelled polar winter descent and more inorganic chlorine (Cl_y) in the lower stratosphere available for activation (see *Feng et al.*, [2005]). This model-specific change is likely to be a significant factor in the improvement, and is supported by the changed profile of O_3 loss in the lower stratosphere in 1996 in Figure 2. We can also note that the source gas scenario used to force the new model runs has an extra 100 pptv of stratospheric Cl_y assumed to come from shorter lived Cl source gases [*WMO*, 2003]. This will increase polar lower stratosphere Cl_y by ∼3%. The computational cost of the model runs precludes a step-by-step inclusion of all of these changes in a series of long model runs.

4. Summary

We have examined how new multiannual simulations of an updated version of the SLIMCAT 3D CTM reproduces the long-term variations in Arctic ozone loss. The model updates include improvements to the transport, chemistry and treatment of PSCs which all contribute to improving the simulation of Arctic O_3 loss. Here, using the updated model, we present the 3D model calculations which reproduce the observed winter/spring Arctic chemical ozone depletion throughout the 1990s, including the shape of the loss profile and its year-to-year variability.

The new model results presented here have a number of important implications. They are the first calculations which demonstrate that a global 3-D chemical model can capture the observed extent of Arctic O_3 depletion over a wide range of meteorological conditions. This resolves a long-standing discrepancy in model studies of Arctic ozone depletion. These results therefore demonstrate that the extensive work on this issue over the past few years from observations, laboratory studies and modelling have led to significant improvements in our overall understanding as summarised in a detailed 3D model. For the first time the model is able to capture the full extent of Arctic ozone depletion, the interannual variability, and its sensitivity to temperature changes. Hence, these simulations show that, if the relevant detailed chemical/physical processes are included in chemistry-climate models, accurate predictions of Arctic ozone loss is now possible for all conditions so far sampled by observations. As such this is a much more positive outlook than has been synthesised based on previously published work [e.g. *Ball*, 2004].

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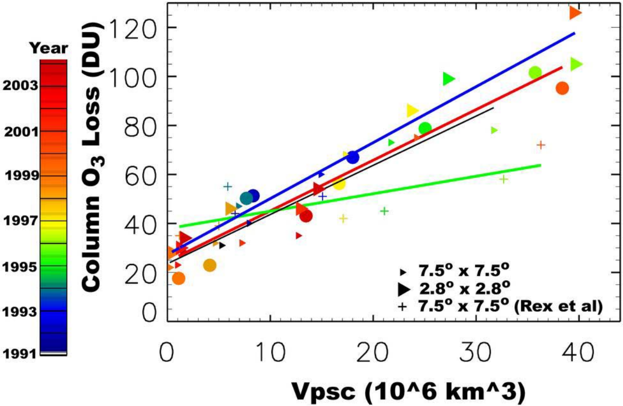
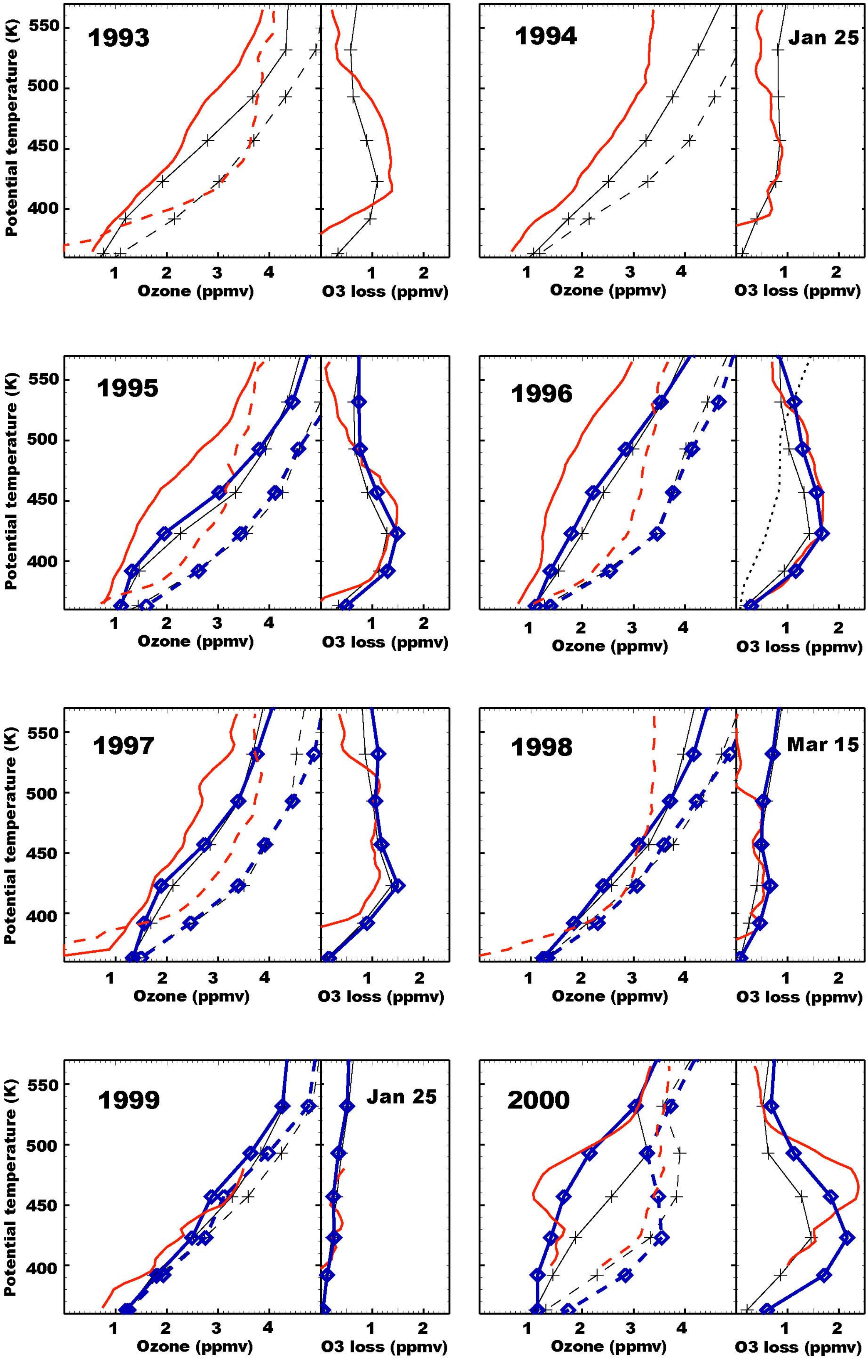
M.P. Chipperfield, W. Feng, School of Earth and Environment, University of Leeds, Woodhouse Lane, Leeds, LS2 9JT, UK. (e-mail: martyn@env.leeds.ac.uk, fengwh@env.leeds.ac.uk)

M. Rex, Alfred Wegener Institute for Polar and Marine Research, Potsdam, Germany. (e-mail: mrex@awi-potsdam.de)


Year
2003
2001
1999
1997
1995
1993
1991
Column O3 Loss (DU)
0
20
40
60
80
100
120
0
10
20
30
40
Vpsc (10^6 km^3)
▸ 7.5° x 7.5°
▶ 2.8° x 2.8°
+ 7.5° x 7.5° (Rex et al)


1993
1994
Jan 25
1995
1996
1997
1998
Mar 15
1999
Jan 25
2000
Potential temperature (K)
550
500
450
400
Ozone (ppmv)
1
2
3
4
O3 loss (ppmv)
1
2