Diagnosis of the Ozone Budget in the Southern Hemisphere Lower Stratosphere

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Abstract The ozone budget in the southern hemisphere (SH) springtime lower stratosphere is studied using the SLIMCAT 3D chemical transport model (CTM). The model was run with UKMO and ECMWF analyses and both produce O_3 structure and variability that is in reasonable agreement with the observations during APE-GAIA (Airborne Polar Experiment - Geophysica Aircraft in Antarctica) campaign. In particular, the model generally reproduces the location of vortex edge well. The O_3 budget, based on its continuity equation, shows that transport processes play an important role in the lower stratosphere, but the horizontal and vertical transport of O_3 tend to show a large cancellation at mid-latitudes. Consequently, the net ozone change is similar to the chemical loss. The transport and chemistry O_3 change are generally out-of-phase. For the chemical loss, ClO_x and BrO_x cycles dominate in the polar vortex, ClO_x cycles are responsible for about 80% of ozone loss below 400K where vortex air mixes rapidly to mid-latitudes.

INTRODUCTION

Stratospheric ozone loss has been observed in both the Antarctic and Arctic polar vortices and it is likely that this loss contributes to the observed mid-latitude trends [World Meterorological Organization (WMO), 1999]. However, export of air from the polar vortices is expected to be different above/below 400 K. More rapid transport below 400 K may lead to a larger effect on mid-latitudes. Here we examine how Antarctic O_3 loss may affect mid-latitudes by diagnosing the O_3 budget in a 3D CTM. The model O_3 is also compared with 1999 APE-GAIA observations.

APE-GAIA CAMPAIGN

The APE-GAIA campaign took place from 15 September to 15 October 1999 [Carli et al., 2000]. The campaign was aimed at studying the transport and chemistry (and microphysics) in the lowermost stratosphere at the edge of the Antarctic polar vortex during the late winter ozone-loss period. The M55 Geophysica high-altitude research aircraft made 5 flights from Ushuaia, Argentina (54°S, 292°E) southwards towards the Antarctic Pennisula and reached around 70°S. In-situ and remote instruments made measurements from ~10 km to ~20 km, which can be used to investigate the extent of mixing across the vortex boundary as a function of altitude and to define and detect vortex edge and filamentary structure.

SLIMCAT 3D CTM

The SLIMCAT 3-D CTM [Chipperfield, 1999] is an off-line model forced by meteorological analyses for the horizontal winds and temperatures while the vertical advection is diagnosed from

heating rates. The model contains the most important species in the O_x , NO_y , Cl_y , Br_y , HO_x families along with a CH_4 oxidation scheme and long-lived tracers. The model has a detailed gas-phase stratospheric chemistry scheme as well as a treatment of heterogeneous chemistry of liquid and solid aerosols (for more information see *Chipperfield* [1999]). We have run the model forced by UKMO UARS and ECMWF T42L50 analyses with a horizontal resolution of 2.5° \times 3.75° and 2.8° \times 2.8° respectively. In both cases the model was initialized from 10 August 1999, and the integrations end on 12 October 1999.

RESULTS

Figure 1 compares the model O₃ field with both in-situ (ECOC) and remote (SAFIRE and MIPAS) observations. The model captures many features of observed O₃ such as the large variations associated with the vortex edge and change of altitude. The APE-GAIA data shows ozone structure of vortex edge from 14 to 18 km. Weaker latitudinal gradients on the south-bound leg of 21/9 flight (with no obvious vortex edge) indicates stronger stirring and mixing below 400 K, which is captured by the model. The polar vortex edge at 16km and 18km on 23/9 is seen clearly from both in-situ and remote measurements. The remote observations also show the bottom of the strongly contained O₃ loss region and evidence for more mixing below this.

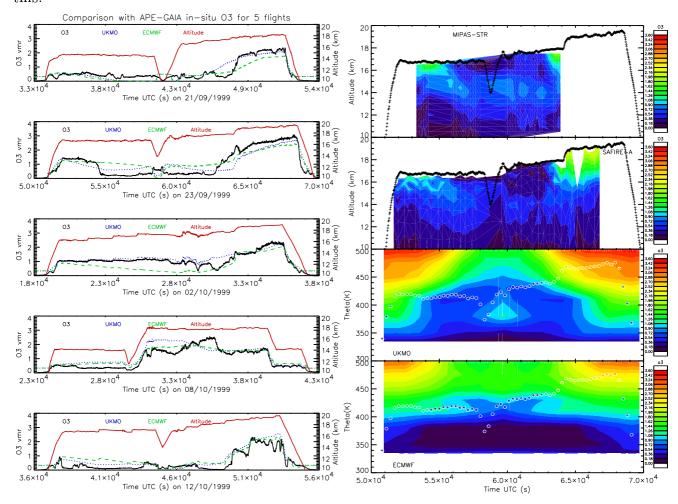


Fig. 1. Comparison of modelled O₃ with (a) ECOC in-situ observations on the 5 scientific APE-GAIA flights; (b) SAFIRE and MIPAS remote observations on 23/9/1999. Results of two model runs (forced by UKMO and ECMWF analyses) are shown.

Given the overall reasonable agreement in Figure 1, we have diagnosed the O_3 budget and analysed it to determine the relative importance of individual terms in the O_3 continuity equation.

The continuity equation can be written as:

$$\frac{\partial [O_3]}{\partial t} = P - L - \nabla \cdot [\vec{V}O_3] \tag{1}$$

where P is the photochemical production rate of ozone, and L is the chemical loss rate. The details of chemical contributions to changes in ozone by individual reaction cycles can be found in $[Millard\ et\ al.,\ 2001]$. $\nabla\cdot[\vec{V}O_3]$ is the net transport of ozone out of the volume, \vec{V} is the flow velocity.

Figure 2 shows time-averaged tendency of zonal mean O_3 over the simulated period and the individual terms for horizontal/vertical transport and chemical loss. Negative values for the horizontal transport in the polar vortex and mid-latitudes, show that there is a net transport of O_3 out of this region. The maximum effect of vertical transport on O_3 occurs at mid-latitudes with the downward transport of O_3 from the higher stratosphere. The transport processes play important role in the O_3 change, but the horizontal and vertical transport tend to balance at mid-latitudes. The ozone chemical production rate (not shown) is small during this period in the mid-high latitude in the lower stratosphere. As expected chemical loss is fastest in the polar vortex region. The diagnosed transport terms are similar between the two analyses, though the ECMWF seems cause more chemical ozone loss (-55ppbv/day) than UKMO (-50ppbv/day). ClO_x cycles are responsible for over 75% O_3 loss in the polar vortex, while BrO_x cycles are responsible for about 20% O_3 loss (we have divided the BrO + ClO cycles between the two families). In fact, ClO_x , BrO_x , HO_x and NO_x cycles are all important at midlatitudes in certain regions.

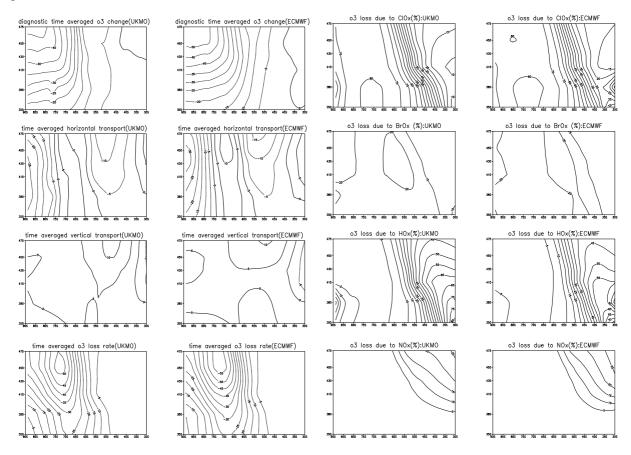


Fig. 2. Zonal mean diagnostics of the ozone budget in the SH lower stratosphere (averaged from 10/8/99 - 12/10/99) from 2 3D CTM runs with UKMO and ECMWF analyses. (a) net O_3 change and individual terms (ppbv/day) (left) and (b) Contribution (%) of various catalytic cycles to the total chemical loss rate of O_3 (right).

For a better comparison of the terms, Figure 3 shows the net O₃ change over the course of the model runs at two altitudes 450 K, where there is a strong vortex edge, and below this at 380 K. Figure 3 shows that transport and chemical O₃ change are generally out-of-phase. Horizontal and vertical transport tend to show a large cancellation at mid-latitudes.

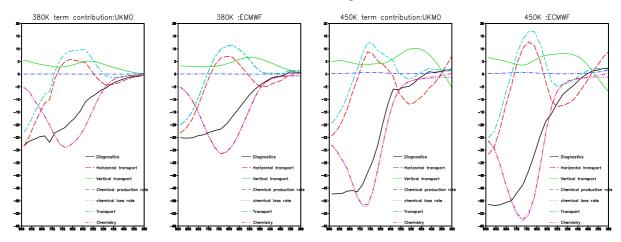


Fig. 3. Net O₃ change (ppbv/day) due to its individual chemical/dynamical contributions from two 3D CTM runs (UKMO and ECMWF forcing) for SH mid-high latitudes at 380K (left) and 450K (right).

SUMMARY

For SLIMCAT simulations of the Antarctic winter and spring in 1999, comparisons between the model O_3 and APE-GAIA observations show encouraging agreement in the lower stratosphere. Analysis of the model ozone budget shows that transport processes play an important role in the lower stratosphere, but the horizontal and vertical transport tend to balance at midlatitudes. Here, net O_3 change is similar to chemical loss. However, the balance between these terms may vary interannually and between the hemispheres - and these different cases need to be examined further. Moreover, the diagnostics presented here are based on a zonally averaged framework, which is not ideal for studying the detailed ozone budget structure in the mid-high latitude lower stratosphere.

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