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Sensitivity of Global Modeling Initiative CTM predictions of Antarctic ozone zone recovery to GCM and DA generated meteorological fields

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Abstract. We use the Global Modeling Initiative chemistry and transport model to simulate the evolution of stratospheric ozone between 1995 and 2030, using boundary conditions consistent with the recent World Meteorological Organization ozone assessment. We compare the Antarctic ozone recovery predictions of two simulations, one driven by meteorological data from a general circulation model (GCM), the other using the output of a data assimilation system (DAS), to examine the sensitivity of Antarctic ozone recovery predictions to the characteristic dynamical differences between GCM and DAS – generated meteorological data. Although the age of air in the Antarctic lower stratosphere differs by a factor of 2 between the simulations, we find little sensitivity of the 1995 – 2030 Antarctic ozone recovery between 350 K and 650 K to the differing meteorological fields, particularly when the recovery is specified in mixing ratio units. Relative changes are smaller in the DAS – driven simulation compared to the GCM – driven simulation due to a surplus of Antarctic ozone in the DAS – driven simulation which is not consistent with observations. The peak ozone change between 1995 and 2030 in both simulations is ~20% lower than photochemical expectations, indicating that changes in ozone transport at 450 K between 1995 and 2030 constitute a small negative feedback. Total winter/ spring ozone loss during the base year (1995) of both simulations and the rate of ozone loss during August and September is somewhat weaker than observed. This appears to be due to underestimates of Antarctic Cl_y at the 450 K potential temperature level.

1. Introduction

Observations over the last decade indicate that the burden of organic chlorine and bromine-containing compounds in the troposphere peaked during the early 1990's and are now declining [WMO, 2003]. Stratospheric inorganic chlorine concentrations have leveled off, and may also soon be observed to be declining [Rinsland et al., 2003; Waugh et al., 2001]. Stratospheric chlorine and bromine concentrations are expected to decline over the next century provided that the halocarbon emission control measures established by the Montreal Protocol and associated international agreements are followed [WMO, 2003].

Stratospheric ozone concentrations are expected to respond to these chlorine decreases due to the important role of halogens in ozone photochemistry [Solomon, 1999]. The potential response of Antarctic ozone is particularly interesting because of its high sensitivity to elevated halogen levels. In addition to changes in halogen loading, there are other factors which may influence Antarctic ozone such as changes in stratospheric temperature and dynamics due to the buildup of greenhouse gases, the chemical effects of continuing increases in methane (CH_4), nitrous oxide (N_2O), and water vapor, and changing stratospheric sulfate aerosol loading [Shindell et al., 1998]. It is therefore not likely that ozone recovery will be simply related to decreases in chlorine. Accounting for the effects of these various factors challenges the scientific community interested in predicting the response of the stratospheric ozone hole to future decreases in stratospheric chlorine/bromine loading.

The representation of stratospheric transport processes in a model, particularly those which control the high latitudes, will clearly affect its prediction of ozone recovery. However, we lack a quantitative understanding of this dependency and the degree of uncertainty in

recovery predictions caused by the uncertainties in modeling atmospheric transport. The goal of this paper is to improve our understanding of this relationship.

Chemistry and transport models (CTMs) of the stratosphere do not generate their own dynamical fields. Instead they rely on externally supplied meteorological datasets (temperatures, winds, etc.) to drive the CTM chemistry and transport modules. The meteorological datasets are generated either by a free -running general circulation model (GCM) or a data assimilation system (DAS). A DAS optimally combines observational information with GCM output to produce ostensibly more realistic meteorological data than is possible with a free -running GCM [Coy and Swinbank, 1997]. GCM and DAS meteorological datasets have characteristic differences which will be discussed below.

In this paper we intercompare two different simulations of Antarctic ozone recovery made with the Global Modeling Initiative (GMI) 3D Chemistry and Transport model [Rotman et al., 2001]. The only difference between the two simulations is the meteorological data driving the model transport. One is driven by the output of a free -running GCM, and the other by a DAS. All other aspects of the model including even the model advection algorithm and number and location of vertical levels, remain the same. Comparing simulations driven by these two meteorological datasets tests the sensitivity of Antarctic ozone recovery predictions to the characteristic differences between GCM -derived and DAS -derived meteorological datasets.

In section 2 we briefly describe the GMI model. In section 3 we outline the model experiments. In section 4 we show and interpret the results of the simulations. We provide a summary and conclusions in section 5.

2. Model Description

The GMI3DCTM was first described by *Rotman et al.* [2001]. Application of the model to the potential impact of stratospheric aircraft emissions is presented in *Kinnison et al.* [2001]. The model was used to investigate the effect of stratospheric aircraft emissions on the occurrence of polar stratospheric clouds (PSCs) in *Considine et al.* [2000].

The current study utilized two sets of meteorological information (horizontal winds, surface pressures, and temperature distributions) to drive the CTM. These datasets were generated by the Goddard Space Flight Center Data Assimilation Office and the Atmospheric Chemistry and Dynamics Branch. The first was generated from a run of the "Finite Volume" General Circulation Model (FVGCM), a new GCM jointly developed by NASA and the National Center for Atmospheric Research (NCAR). This GCM combines the physical parameterizations of the NCARCCM3 GCM [Kiehl et al., 1998] with the *Lin and Rood* [1997] advection scheme (See http://polar.gsfc.nasa.gov/sci_research/fvdas/index.php) for details. The second set of meteorological data was generated by a recent version of the Goddard Earth Observing System Data Assimilation System (GEOS -DAS) [Schubert et al., 1993], which uses the FVGCM to provide forecasts for the DAS. *Schoeberl et al.* [2003] provide a useful comparison of FVGCM and FVDAS characteristics.

Both sets of meteorological data were interpolated to the same hybrid sigma -pressure coordinate system for this study, following the technique used to create meteorological datasets for the Goddard Space Flight Center 3DCTM [Considine et al., 2003]. The horizontal resolution of the coordinate system is 5° longitude by 4° latitude, with 28 vertical levels. The interface between sigma (terrain -following) and constant pressure horizontal surfaces occurs at 247.46 hPa. The modelex tends up to 43 hPa, about the top of the stratosphere.

The photochemical scheme has been upgraded from *Rotman et al.* [2001] with improvements described in *Douglas et al.*, "Radicals and reservoirs in the GMI chemistry and transport model: Comparisons to measurements," submitted to Journal of Geophysical Research, 2003 (hereafter referred to as Douglas 2003). The PSC parameterization settings were changed from that described in *Considine et al.* [2000]. In this version, Type 1 PSCs were assumed to have an nitric acid trihydrate (NAT) composition instead of supercooled ternary sulfate (STS). This choice results from a desire to better represent the PSC characteristics of the Arctic, where recent observations by *Fahey et al.* [2001] show extensive NAT particle formation and denitrification, and the recent study of *Considine et al.* [2003], which showed that STS aerosols are ineffective denitrifiers of the Arctic polar vortex due to their small size. The WMO 2002 background sulfate aerosol distribution is also included, which catalyzes heterogeneous reactions with the same temperature dependence as STS aerosols. Thus the only Type 1b (liquid) PSC processes which are neglected in this parameterization are the swelling of the PSCs at low temperatures due to HNO₃ uptake, and their sedimentation.

3. Experiment Description

The two simulations using the GCM and DAS meteorological datasets will be referred to as the GMI_GCM and GMI_DAS simulations, respectively, for the remainder of this paper. The source gas boundary conditions used for the two simulations correspond to the MA2 scenario specified in Table 4B-1 of the WMO 2002 ozone assessment [WMO 2003]. The simulations were initialized similarly and spun up for five years at 1995 source gas boundary conditions. The simulations were then integrated forward in time for 36 years, through the year 2030. Halocarbon surface mixing ratios were respecified from Table 4B-2 of the WMO assessment

report. CH₄ and N₂O mixing ratios were respecified as scenario A2 of the International Panel on Climate Change 2001 report, also listed in Table 4B-3 of the WMO assessment.

The background sulfate aerosol distribution was specified using the WMO 2002 assessment aerosol dataset. The aerosols followed observed values from 1995 to 1999. Between 2000 and 2030, the aerosol distribution for 1997 was repeated annually.

One year of meteorological data was used to drive each simulation, and the year was recycled for each year of the simulation. The year of FVGCM data used in the GMI_GCM calculation was taken from a 50-year run of the FVGCM. The particular year was chosen on the basis of its relatively cold Arctic winter, for reasons unrelated to the work presented in this paper.

For both simulations the year of data used extends from July 1, through June 30. The FVDA data corresponds to July 1, 1999 through June 30, 2000. Thus one each year of the 36-year simulation there is a discontinuity between June 30 and July 1 where the rearer relatively large changes in wind fields, etc. The analysis presented below does not appear to be affected by this discontinuity.

The model diagnostics used in the following analysis consists of species mixing ratios written out at 0 Zevery 3 days during 1995 and during 2030. The analysis also uses three -day average reaction rates, again written out at 0 Zevery 3 days.

4. Results

4.1 Dynamical Differences

There are substantial dynamical differences between the two sets of meteorological data used to drive the GMI_GCM and GMI_DAS simulations. One important difference is the strength of the stratospheric residual circulation. A good measure of residual circulation strength is the mean

age of stratospheric air, and how it is distributed in latitude and altitude. The mean age of the air at a particular latitude and altitude can be thought of as the average time for a parcel of air to travel from the ground to that latitude and altitude [Hall and Plumb , 1994; Waugh and Hall , 2002]. Figure 1 shows the vertical and meridional distribution of the mean age of air calculated from simulations using the GCM (Figure 1a) and DAS (Figure 1b) wind fields described above. Figure 1c shows the difference between the two aged distributions, and Figure 1d compares the simulated mean ages with values inferred from NASA ER -2 aircraft observations of CO ₂ [Boering et al. , 1996] . Figure 1a shows that the GCM meteorological data produces same mean age of ~5.5 – 6 years in the upper stratosphere. The DAS -derived upper stratospheric mean age shown in Figure 1b is ~2 years younger. In the upper stratosphere the GCM -derived mean age is in much better agreement with observationally -derived estimates. Figure 1d shows that this is also the case in the lower stratosphere [Hale et al. , 1999; Waugh et al. , 2001] . The mean age differences shown in Figure 1 are typical of GCM -based and DAS -based circulations. In general, residual circulations inferred from analyzed meteorological data are too vigorous, producing young stratospheric air. [Considine et al. , 2003; Schoeberl et al. , 2003; Weaver et al. , 1993].

Distributions of long -lived tracers such as N ₂O and CH ₄ can also provide information about transport differences between the GMI_GCM and GMI_DAS simulations. Figures 2a and 2b show equivalent -latitude averaged N ₂O in September on the 650K and 450K potential temperature surfaces, respectively, as a function of equivalent latitude, from the GMI_GCM and GMI_DAS simulations. The figure also shows equivalent latitude averaged N ₂O distributions derived from Cryogen Limb Array Etalon Spectrometer (CLAES) satellite observations made during September, 1992, at 650K and 450K [Roche et al. , 1996] . A t450K, Airborne Tunable

Laser Absorption Spectrometer (ATLAS) observations made during the September, 1987 flights of the Airborne Antarctic Ozone Experiment (AAOE) and the October, 1994 flights of the Airborne Southern Hemisphere Ozone Experiment (ASHOE) are included as well [Loewenstein et al., 1989; Loewenstein et al., 1990].

The simulated and observed N₂O distributions all exhibit the familiar "rollercoaster" pattern caused by lower stratospheric dynamical processes at both 450 K and 650 K. At 650 K, the gradient separating the tropics from the midlatitudes is substantially weaker in the GMI_DAS simulation than in either the CLAES observations or the GMI_GCM simulation. The gradients separating the midlatitudes from the polar vortex are similar in both simulations at 650 K and are weaker than observed. The weaker gradients indicate that the meridional mixing between the vortex and midlatitudes is too strong relative to the descent rate of air within the vortex in both simulations.

At 450 K the sub-tropical N₂O gradients separating the tropics from the midlatitudes are similar in both simulations and are weaker than are observed. At high latitudes the GMI_GCM N₂O distributions agree better with observations than the GMI_DAS simulation, suggesting a somewhat more isolated and realistic representation of the polar vortex.

Overall, the GMI_GCM simulation more convincingly reproduces observed N₂O distributions. The differences between the simulated N₂O distributions are consistent with the younger age of high-latitude air in the GMI_DAS simulation and further illustrate a characteristic problem with CTM simulations driven by DAS-generated windfields. Further evaluation of the different effects of the transport fields on stratospheric tracer distributions is provided in Strahan, S.E. and A.R. Douglass, "Evaluating the credibility of transport processes

in the Global Modeling Initiative 3D model simulations of ozone recovery," *Journal of Geophysical Research – Atmospheres*, submitted article, 2003 (hereafter referred to as SD2003).

Another important difference to consider is the occurrence of temperatures low enough to provoke formation of PSCs. The occurrence of these low temperatures in the two simulations is shown in Figure 3. Figure 3a shows the size of the Antarctic region below 195 K (the approximate temperature below which PSCs can form) as a function of day of year and potential temperature in the GMI_GCM simulation. The area is expressed as a fraction of the total global area. Figure 3b shows the same plot for the GMI_DAS simulation, and Figure 3c shows the difference between the two. Figures 3a and 3b demonstrate a qualitatively similar morphology, with the coldest temperatures occurring earlier in the year at higher potential temperatures. Figure 3c shows that differences between the two simulations in the occurrence of cold temperatures is not large. In the lower stratosphere (~450 K) the GMI_DAS simulation has a slightly larger cold pool throughout most of the year, although during days ~210 to ~240 the GMI_GCM cold pool is larger by ~10%. The similarity of the occurrence of cold temperatures in the GMI_DAS and GMI_GCM simulations allows for a more specific test of the sensitivity of recovery predictions to dynamical differences between the two simulations.

4.2 1995-2030 Changes in Ozone

Figure 4 shows the change in Antarctic ozone mixing ratios in response to source gas forcing between 1995 and 2030 as a function of day of year and potential temperature in the GMI_GCM (Figure 4a) and GMI_DAS (Figure 4b) simulations. The figures show an average over equivalent latitudes “poleward” of -65° equivalent latitude. We will use such averages throughout this paper to provide a measure of average constituent behavior in the Antarctic. Figures 4a and 4b show similar changes in Antarctic ozone in the two simulations. In the

GMI_GCMsimulation, the peak ozone increase of 0.76 ppm occurs at ~500 K on Julian day 278, in early October. At 450 K, the peak occurs 3 days later and reaches 0.7 ppmv. In the GMI_DAS simulation, the peak ozone increase of 0.70 ppmv also occurs at 500 K on day 278. At 450 K, the peak increase is 0.66 ppmv. Thus, the GMI_GCM peak is ~6% larger than in the GMI_DAS at 450 K and ~8.5% larger at 500 K. The Antarctic ozone recovery predictions appear to display little sensitivity to the substantial differences in the meteorological data which lead to the aged differences shown in Figure 1. Larger differences between the GMI_GCM and GMI_DAS simulations are shown in Figures 5a and 5b, which show the percentage differences in Antarctic ozone between 1995 and 2030 in the GMI_GCM (5a) and GMI_DAS (5b) simulations. In contrast to Figure 4, the peak 1995–2030 change in Antarctic ozone of ~40% occurs at 450 K in the GMI_GCM simulation, while the reisasmallerincreaseof~28%inthe GMI_DAS simulation at 400 K. Peak relative changes occur about the same day of the year, but occur ~50 K and ~100 K lower in potential temperature than the locations of the peak mixing ratio differences in the GMI_GCM and GMI_DAS simulations, respectively.

The relative GMI_GCM peak shown in Figure 5a is ~43% larger than the GMI_DAS peak shown in Figure 5b even though the peak 1995–2000 mixing ratio differences are similar because of differences between the base year (1995) of the simulations. Figure 6 compares Antarctic ozone mixing ratios from the two simulations for the base year on the 450 K (Figure 6a), 550 K (Figure 6b), and 650 K (Figure 6c) potential surfaces. Also included in the figure are observed values from 1992 (red crosses), 1993 (blue crosses), and 1994 (green crosses) version 5 MLS measurements [Froidevaux et al., 1996; Livesey et al., 2003], and 1992 version 9 CLAES observations [Bailey et al., 1996]. Both simulations have the same general morphology, with peak ozone occurring in the winter and minimum during the late spring. However, there is

generally more Antarctic ozone in the GMI_DAS simulation. Figure 6a shows that there is up to ~1.0 ppmv more ozone in the GMI_DAS simulation at 450K than in the GMI_GCM simulation. The higher ozone mixing ratios in the GM_I_DAS simulation produce the smaller relative 1995 - 2030 differences shown in Figure 5b.

The GMI_GCM simulation agrees better with the MLS ozone measurements at 450K and 550K than GMI_DAS. At 650K, both model simulations exhibit an annual cycle which is more pronounced than seen in the observations. Note that the MLS observations at 450K suffer from an instrumental artifact which results in higher ozone mixing ratios during the middle of a yaw cycle than at the beginning or end. This artifact is particularly noticeable during the winter yaw cycle of 1993 in Figure 6a. The cause of the artifact is not well understood [Froidevaux et al., 1996], but the few tenths of a part per million variation does not seriously affect this qualitative comparison. The artifact is known to be smaller in version 5 retrieval than previously [William G. Read, private communication, 2003]. There are significant differences between the 1992 MLS and CLAES observations during the fall and winter yaw cycles. This may be due to the existence of heavy PSCs during this time, which interfere with the CLAES retrieval. At other times of year the CLAES and MLS observations agree well with each other.

Although the 1995 - 2030 increase in ozone is not very sensitive to the dynamical differences between the two simulations, the simulation of the base year is interesting because relative differences are typically compared in assessment exercises, rather than absolute (mixing ratio) differences. Such an approach may introduce substantial intermodel variability into an assessment exercise as a consequence, even in cases where the model responses are similar in absolute terms.

We compare in the next section the evolution of ozone over the course of the winter/spring seasons to determine the manner in which ozone concentrations evolve and are determined by springtime chemical and dynamical influences. This will help explain the low sensitivity of the 1995–2030 increase in ozone to the dynamical differences between the two simulations.

4.3 Evolution of ozone in 1995

We choose to examine in detail the evolution of Antarctic ozone from winter through spring in the base year of the simulations at 450 K potential temperature level, because peak 1995–2030 ozone changes in both simulations occur within 50 K of this surface, with the changes expressed either as a mixing ratio or a percentage difference. We also use July 1 as a reference point to compare specifically the springtime ozone changes. The solid line in Figure 7a shows the change in Antarctic ozone (i.e., the loss of ozone) after July 1 in the GMI_GCM simulation at 450 K. Two other lines are also shown on this plot. The dashed lines show the integrated chemical loss of ozone after July 1, calculated from model chemical rate diagnostic output. The dash-dot lines show the difference between the actual loss of ozone and the integrated chemical loss of ozone, which indicates the influence of transport. The red, blue, and green crosses in Figure 7a show changes in Antarctic ozone mixing ratios after July 1 calculated from ozone observations made in 1992, 1993, and 1994, respectively, by the Microwave Limb Sounder (MLS) instrument.

Figure 7a shows that over the course of the winter and spring seasons the GMI_GCM simulation has a maximum loss of ozone of ~1.9 ppmv. The solid line showing the actual ozone loss in the model tracks very closely to the dashed line showing integrated chemical losses through ~day 280 (early October). This indicates that at this potential temperature the evolution of ozone

is dominated by chemical loss and ozone transport plays a minor role - through day 280, Figure 7 suggests that transport has reduced ozone loss by ~9%. Note however that this fact does not mean that transport is unimportant – just that changes in ozone due to the transport of ozone itself are small compared to changes in ozone due to in-situ photochemistry. The in-situ photochemistry may be substantially influenced by transport, for instance of Cl y. After day 280 net ozone loss (solid line) remains steady while the integrated chemical loss term continues to drop by about 0.5 ppm v throughout the end of the year. Ozone is thus being transported into the region during this time to balance the continuing chemical loss. The fact that ozone transport is small during the August/September time frame is consistent with the results of Wu and Desler [2001], which compared ozone loss at 46.5°K observed by MLS with estimates of chemical loss based on MLS observations of ClO during August and September of 1992, 1993, and 1994.

Comparison of the simulated ozone in Figure 7a with the MLS observations shows that the ozone loss is about 20% lower than the average observed ozone loss during 1992, 1993, and 1994. Also, averaged over the UARS springtime yaw cycle, the simulated rate of change of ozone is ~30% smaller than is observed (~28.5 ppbv/day compared to the observed ~41.4 ppbv/day (we average over the yaw cycle to remove the effects of the instrumental artifact discussed above).

Figure 7b shows the same comparison as in Figure 7a for the GMI_DA simulation. The figure shows that there are strong similarities with the GMI_GCM results shown in Figure 7b. The maximum loss of ozone is ~2.2 ppm v about day 330. Although transport causes an initial deviation from the chemical loss in late July (~day 200), simulated ozone loss is virtually identical to the chemical loss by day 280, and approximately parallel to the integrated chemical loss through about day 300 (late October). In contrast to the GMI_GCM results, during the last

two months of the year, model ozone loss exceeds the integrated chemical loss, implying that transport acts to reduce ozone after about day 300 (Note that transport is acting to reduce Antarctic ozone concentrations from day 200 on, but the net effect of transport is near zero through day 300). Kinematic backward trajectory calculations of high equivalent latitude parcels initialized at 450 K indicate that this is due to transport of some air with smaller ozone mixing ratios to the 450 K from below.

As above, the overall loss is somewhat low compared to observations. The ozone loss rate of ~ 26.1 ppbv/day during the August/September time period is also substantially weaker than observed and slightly weaker than in the GMI_GCM simulation. Interestingly, Figure 7b shows that ~ 4 ppbv/day of the loss rate during August/September is due to transport, or about 16%. Without the transport contribution, the chemical loss rate is $\sim 23\%$ smaller than the GMI_GCM loss rate. The total chemical loss is 1.9 ppmv, which is about 18% smaller than the chemical loss in the GMI_GCM simulation.

We next examine in more detail the photochemistry of the two simulations to determine the causes of these differences.

4.4 Photochemistry in 1995

The primary deficiency of the model calculations shown in Figure 7 is the weak ozone loss compared to the MLS observations in the August/September time frame, particularly in the GMI_DAS simulation. Virtually all of the ozone destruction ($>93\%$) at this time in the two simulations is caused by catalytic cycles involving Cl_x and Br_y radical species.

We refer to the sum of the radical ClO and its night time reservoir Cl_x and O_2 as “active chlorine,” and denote it by the symbol Cl_x . Figure 8a shows high-latitude averaged Cl_x and Cl_y on the 450 K surface between days 100 and 350 from both simulations. The figure shows that Cl_y

peaks in August in both simulations with values of ~2.3 ppbv for GMI_GCM and ~1.8 ppbv for GMI_DAS —~22% less. The larger Cl_y mixing ratios in the GMI_GCM simulation result in larger Cl_x mixing ratios for this simulation as well. Figure 8b compares the ratio Cl_x/Cl_y in the two simulations. Peak Cl_x activation exceeds 85% in both simulations. The ratio is similar during August/September, dropping from ~80% to ~60% by mid-September. The similarity of the ratio indicates that the weaker GMI_DAS ozone loss during this time shown in Figure 7b is due to lower Cl_y at 450 K, rather than differences in chlorine activation.

Also shown in Figure 8a is an estimate of Cl_y derived from CLAES N₂O observations made during the Antarctic winter/spring of 1992 using the Cl_y/N₂O relationship derived in Woodbridge et al. (1995). During September (days ~244 – 273), the Cl_y in the GMI_GCM simulation agrees reasonably well with the observational estimates although observationally inferred Cl_y is apparently increasing during this time while the simulated Cl_y is beginning to drop from a winter peak. The Cl_y in the GMI_DAS simulation is low by ~0.5 ppbv. The observational estimates suggest that Cl_y increases until December, which is contrary to the decreasing trend in both simulations. This late -year discrepancy has little impact on simulated ozone loss because the Cl_x/Cl_y ratio falls to very low values by this time due to the return of sunlight and warm temperatures to high latitudes. More detail on the evolution of high-latitude tracer concentrations during the Southern Hemisphere springtime is given in SD2003.

We can also estimate Cl_y from the lower stratospheric high-latitude N₂O observations made during the AAOE and ASHOE expeditions which are shown in Figure 2. Figure 9 shows these observed estimates of Cl_y and simulated Cl_y at 450 K as a function of equivalent latitude. All three of the observational estimates exceed both the GMI_GCM and GMI_DAS simulations throughout the southern hemisphere. At mid-latitudes both simulations are quite a bit lower than

the observational estimates, likely due to the very weak meridional gradient across the subtropics. At high latitudes the GMI_GCM simulated Cl_x is ~15% low, while the GMI_DAS underestimates at high latitudes is ~30%.

Wu and Dessler [2001] estimate from MLS ClO observations vortex-averaged Cl_x mixing ratios of ~2–2.5 ppbv at 465 K during the late August/early September period in 1992, 1993, and 1994. Figure 8a shows that Cl_x in the GMI_GCM simulation is about 15% to 20% lower than this estimate, and Cl_x in the GMI_DAS simulation is about 30% to 35% low. These deficits are consistent with the Cl_y comparison shown above and could account for the weak August/September loss rates seen in the GMI_GCM and GMI_DAS simulations.

Figures 8 and 9 show that the in-situ photochemistry which controls Antarctic lower stratospheric ozone loss is affected by the differences in atmospheric transport which lead to the aged discrepancies shown in Figure 1. Some combination of higher meridional mixing and/or lower vertical descent of air at high latitudes in the GMI_DAS simulation results in lower Cl_y mixing ratios at 450 K than in the GMI_GCM simulation. The result is about 20% lower total chemical ozone loss and a correspondingly smaller chemical ozone loss rate during August/September. Similarly, both Cl_x and Cl_y in the GMI_GCM simulation appear to be about 20% lower than observed values. These differences are relatively modest considering that the age of lower stratospheric Antarctic air differs by about a factor of 2 in the two simulations.

4.5 Chemistry and transport contributions to 1995–2030 ozone change

In section 4.2 above we showed that the peak 1995–2030 increase in Antarctic ozone at 450 K in the GMI_GCM simulation is only ~6% larger than the peak increase in the GMI_DAS simulation, despite the substantial dynamical differences in the meteorological data driving the two simulations discussed in section 4.1. Section 4.3 showed that the dynamical differences

cause~20% differences between the GMI_GCM and GMI_DAS simulations in the springtime chemical ozone loss during the base year of the simulations, due largely to the effects of transport on Cl_y (and by implication, Br_y). To try to determine why the peak 1995 -2030 increases in the two simulations differ by ~6% while springtime ozone loss for the base years differ by ~20%, we now examine the individual chemistry and transport contributions to the 1995 -2030 ozone change.

Figure 10a shows the high latitude winter/spring ozone loss in the GMI_GCM and GMI_DAS simulations at 450K for 1995 (solid lines), 2030 (dashed lines) and the difference between 1995 and 2030 (dashed-dot lines). The figures show that while the winter/spring ozone losses in the GMI_GCM and GMI_DAS simulations differ in timing and magnitude, the 1995-2030 difference in ozone loss in each of these two simulations is very similar, consistent with the differences shown in Figure 4 above. Peak values of the difference are 0.56 ppmv on day 281 in the GMI_GCM simulation and 0.51 ppmv on day 275 in the GMI_DAS simulation. Thus, the GMI_GCM 1995 -2030 peak “recovery” of Antarctic ozone is slightly less than 10% larger than the recovery in the GMI_DAS simulation.

Figure 10b shows the winter/spring chemical loss of ozone in 1995 (solid blue (GCM) and red (DAS) lines) and 2030 (dashed blue and red lines) in the two simulations, in addition to the 1995 -2030 change in winter/spring chemical loss at 450K for the simulations (dashed-dot blue and red lines). On the days of the peak 1995 -2030 change in ozone loss for the two model simulations shown in Figure 10a, the 1995 -2030 change in chemical loss is 0.69 ppmv for the GMI_GCM simulation and 0.66 ppmv for the GMI_DAS simulation. When only the change in chemical loss is considered, the GCM -driven simulation is just 4.5% larger than the DAS -driven simulation.

At the same time the blue and red solid lines in Figure 10b show that the 1995 chemical loss from July 1 through the day of peak recovery is ~ 2.1 ppmv in the GMI_GCM simulation and ~ 1.5 ppmv in the GMI_DAS simulation. So, even though the GCM -driven simulation has a chemical ozone loss that is $\sim 38\%$ larger than the DAS -driven simulation at this time, the change in the chemical ozone loss between 1995 and 2030 is only slightly larger (4.5%) than the DAS -driven simulation. This will be discussed further below.

Figure 10c shows the effects of transport on ozone loss in 1995 (solid lines) and 2030 (dashed lines) for the GMI_GCM (blue) and GMI_DAS (red) simulations. It also shows the 1995-2030 difference (dash-dot lines) for each of the simulations. The 1995 -2030 differences are relatively similar between the two simulations, with the GMI_DAS transport contribution slightly more negative than GMI_GCM. Note that since the 1995 -2030 change in ozone transport is due entirely to changes in ozone gradients in the Antarctic (the wind fields themselves are the same every year), a negative contribution from transport is expected due to weaker ozone gradients in 2030 compared to 1995. On the day of peak ozone recovery (Figure 9), the effect of changes in transport is to reduce the recovery peak by $\sim 19\%$ in the GMI_GCM simulation and $\sim 23\%$ in the GMI_DAS simulation.

Figure 10 shows that the effect of changes in the role of ozone transport between 1995 and 2030 is to create a large difference in the ozone recovery of these two simulations, rather than a smaller difference. In the absence of the transport effects, the difference between the two simulations would simply be that of the 1995 -2030 changes in chemical ozone loss $\sim 4.5\%$. Ozone transport is therefore not obscuring a larger difference between the simulations in the response of Antarctic photochemistry to the 1995 -2030 changes in source gas forcing.

Figures 7a and 7b show that the bulk of the springtime ozone loss in both simulations occurs between days 220 and 280. The decreases in Cl_x and Cl_y from 1995 to 2030 that occur during this time period will determine how ozone changes due to photochemistry between 1995 and 2030. Figure 11 shows these decreases in Cl_x and Cl_y from 1995 to 2030 for both the GMI_GCM and GMI_DAS simulations. Figure 11 shows that the 1995–2030 change in Cl_y in the GMI_DAS simulation is about 10% smaller than the corresponding change in the GMI_GCM simulation. This is about half of the amount by which 1995 Cl_y in the GMI_DAS simulation differs from the GMI_GCM simulation. Figure 11 also shows that the 1995–2030 change in Cl_x in the GMI_DAS simulation is about the same as the corresponding change in the GMI_GCM simulation. This implies that in the GMI_DAS simulation the Cl_x/Cl_y ratio has decreased between 1995 and 2030, making the overall decline in Cl_x about the same as in the GMI_GCM simulation. Although it is not clear exactly why the Cl_x/Cl_y ratio decreases in the GMI_DAS simulation more than in the GMI_GCM simulation, this behavior explains the similarity of the 1995–2030 change in chemical ozone loss in the two simulations.

5. Summary and Conclusions

We have used the GMI 3DCTM to simulate ozone recovery over the 1995–2030 time period using two different representations of atmospheric transport processes. We have compared Antarctic ozone recovery predictions generated in these two simulations to evaluate the sensitivity of the recovery predictions to the characteristic differences between GCM and DAS meteorological datasets.

The simulation driven by the GCM meteorological fields realistically captures the age of air distribution of the stratosphere, while the age of stratospheric air in the DAS -driven simulation is up to 2.5 years younger than an observed. The GCM -driven simulation also has a

more realistic representation of long lived trace species, as demonstrated by the comparison of simulated and observed N₂O. Both simulations have similar cold pool regions in the polar southern hemisphere, a fact which makes the intercomparison of the simulations more direct test of dynamical sensitivity than would otherwise be the case.

Despite the substantial dynamical differences in the two simulations, the 1995 - 2030 response of ozone mixing ratios between 350 K and 650 K at Antarctic latitudes to changes in trace gas forcing is very similar. This indicates that the dynamical differences between the simulations do not strongly influence the 1995 - 2030 ozone change. One cannot therefore argue that a DAS - generated meteorological dataset, or any set which underestimates stratospheric age, is automatically inappropriate for making ozone recovery calculations. Differences between long-lived tracers in the Antarctic lower stratosphere provide a better indicator of the response to changes in source gas boundary conditions.

The relative response of Antarctic ozone to source gas changes between 1995 and 2030 shows larger differences between the two meteorological datasets and is stronger in the simulation driven by GCM output due to lower Antarctic ozone concentrations in the base year of the simulation. The utility of relative ozone change as a measure of recovery therefore depends on how well the current year is simulated.

The lower ozone in the GMI_GCM simulation is in better agreement with observations. In the absence of other factors which could affect recovery, such as changes in stratospheric temperatures or water vapor, the recovery prediction the GMI_GCM simulation would be more credible as a result.

Antarctic winter/spring ozone loss at 450 K in both simulations is driven primarily by photochemistry through the end of October, and ozone transport to or from the vortex at 450 K

playsonlyasmallandsimilarroleduringthistime.Thisresultisconsistentwithobserved behavior.Totalozonelossinbothsimulationsagreesreasonablywellwithobservationsinboth simulations, but the rate of ozone loss during the August/September time period is weak. The weak loss appears to be explained by low Cl_x at high latitudes, which is due in large part to low Cl_y.

These results have implications for the types of model intercomparisons used in the WMO assessment reports. These assessments typically present model results in terms of relative rather than absolute differences from some base year, for instance 1980 (see for example Figure 4-42 of WMO [2003]). If absolute differences were used instead the intermodel variability seen in such figures may be significantly reduced.

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Figure Captions

Figure 1: Mean age of air in the GMICM as a function of latitude and pressure. Top left -mean age using FVGCM meteorological data. Top right -mean age using FVDAS meteorological data. Bottom left -mean age difference, FVGCM -FVDAS. Bottom right: Meridional dependence at 56 hPa of mean age in the FVGCM (solid line) and FVDAS (dashed line) simulations, compared with mean age estimates made using aircraft CO₂ observations (asterisks show the mean age from observation s, and diamonds show the uncertainty range).

Figure 2: Observed and simulated N₂O on the a. 650 K and b. 450 K potential temperature surfaces as a function of equivalent latitude. Solid blue line -N₂O from GMI_GCM simulation, averaged over the September 1 – September 18 time period. Solid red line -N₂O from GMI_DAS simulation, averaged over the September 1 – September 18 time period. Magenta asterisks with error bars: equivalent -latitude averaged N₂O from the CLAES instrument, averaged from September 1 to September 18. Error bars show ± 1 standard deviation of all observed values constituting the average. Green diamonds – equivalent latitude averages of ALIAS N₂O observations made during September, 1987 as part of the AAOE. Black diamonds – equivalent latitude averages of ALIAS N₂O observations made during October, 1994 as part of ASHOE.

Figure 3: Cold pool areas in the GMI_GCM and GMI_DAS simulations. a. Cold pool area in the GMI_GCM simulation as a function of day of year and potential temperature. The area below 195 K in the extratropical southern hemisphere is expressed as a percentage of the total surface area of the Earth. b. Same as in a, but for the GMI_DAS simulation. c. Difference in area

between the GMI_DAS and GMI_GCM simulations. A positive value means a larger pool in the GMI_DAS simulation and hence, a larger region of temperatures below 195K.

Figure 4: Change in Antarctic ozone (ozone averaged over equivalent latitudes poleward of 65° S) between 1995 and 2030 as a function of day of year and potential temperature, in mixing ratio units. a. Difference between Antarctic ozone in 2030 and 1995 in the GMI_GCM simulation. b. Difference between Antarctic ozone in 2030 and 1995 in the GMI_DAS simulation.

Figure 5: Change in Antarctic ozone (ozone averaged over equivalent latitudes poleward of 65° S) between 1995 and 2030 as a function of day of year and potential temperature, in percent. a. Difference between Antarctic ozone in 2030 and 1995 in the GMI_GCM simulation. b. Difference between Antarctic ozone in 2030 and 1995 in the GMI_DAS simulation.

Figure 6: Antarctic ozone mixing ratio in parts per million by volume as a function of day of year during 1995 at a. 450K, b. 550K, and c. 650K in the GMI_GCM (solid line) and GMI_DAS simulations (dashed line) simulations. Also shown are averages of observed ozone values for equivalent latitudes poleward of 65° S on each potential temperature surface from the MLS instrument in 1992 (red crosses), 1993 (blue crosses), and 1994 (green crosses), and the CLAES instrument (magenta crosses).

Figure 7: Antarctic ozone loss in the a. GMI_GCM and b. GMI_DAS simulations for 1995 at 450K. Ozone loss is defined as the change in ozone since July 1. Solid line: simulated ozone

loss. Dashed line: integrated chemical ozone loss (see text for explanation). Dash-dot line: simulated loss minus integrated chemical loss, which can be interpreted as the effects of transport. Red, blue, and green crosses: vortex averaged ozone loss calculated from 1992, 1993, and 1994 MLS observations, respectively.

Figure 8:a. Timeseries of Antarctic Cl_{y} (solid lines) and Cl_{x} (dashed lines) in the GMI_GCM and GMI_DAS simulations during 1995 on the 450K potential temperature surface. Cl_{x} is defined as $\text{ClO} + 2 * \text{Cl}_{\text{y}} \text{O}_2$. Blue lines show GMI_GCM results and red lines show GMI_DAS results. Crosses are observationally-based estimates of Antarctic Cl_{y} from CLAES N_2O observations as discussed in text. b. Ratio of Antarctic Cl_{x} to Cl_{y} in the two simulations, as a percentage of Cl_{y} .

Figure 9: Equivalent $\text{\textit{l}}$ -latitude averaged Cl_{y} comparisons. Magenta asterisks with error bars indicate Cl_{y} inferred from CLAES N_2O observations from September, 1992. Black crosses show Cl_{y} inferred from ATLAS N_2O observations made on NASA ER-2 flights in October, 1994, as part of the ASHOE mission. Green crosses show Cl_{y} inferred from ATLAS N_2O observations made on NASA ER-2 flights in September, 1987, as part of the AAOE mission.

Figure 10: Comparison of ozone loss at 450K in 1995, 2030, and their difference. a. Shows ozone loss after July 1 during 1995 in GMI_GCM (solid blue line) and GMI_DAS (solid red line) simulations. Dashed blue and red lines are the corresponding losses for the year 2030. Dashed-dot lines show 2030 - 1995 difference for GMI_GCM (blue) and GMI_DAS (red). b. Shows chemically integrated ozone loss after July 1 during 1995 (solid lines), 2030 (dashed

lines) and the 2030 - 1995 difference (dashed -dot lines), for GMI_GCM (blue) and GMI_DAS (red). c. Shows net effect of transport after July 1 during 1995 (solid lines), 2030 (dashed lines) and the 2030 - 1995 difference (dashed -dot lines), for GMI_GCM (blue) and GMI_DAS (red).

Figure 11: Change in Antarctic Cl_y and Cl_x between 1995 and 2030 at 450K in the GMI_GCM and GMI_DAS simulations. Solid blue (red) line corresponds to 1995 - 2030 change in time series of Cl_y averaged at equivalent latitudes above 65°S in the GMI_GCM (GMI_DAS) simulation. Dashed blue (red) line corresponds to 1995 - 2030 change in time series of Cl_x averaged at equivalent latitudes above 65°S in the GMI_GCM (GMI_DAS) simulation.

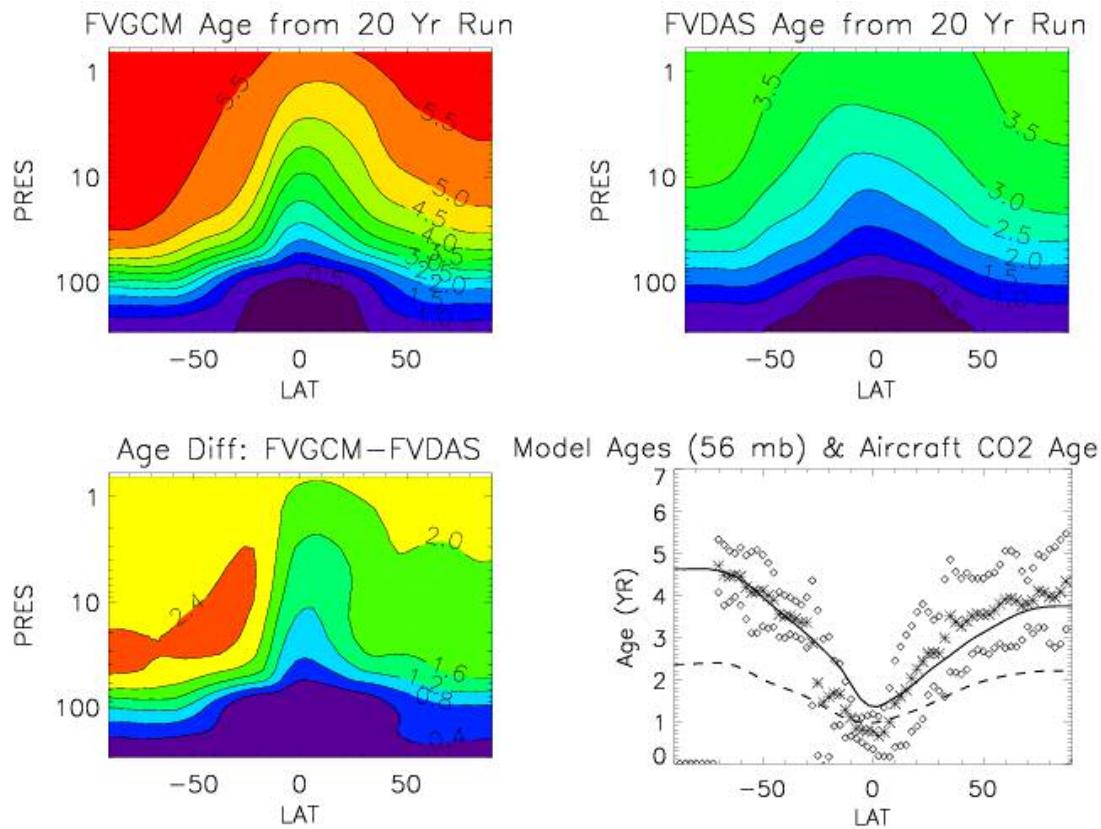


Figure1

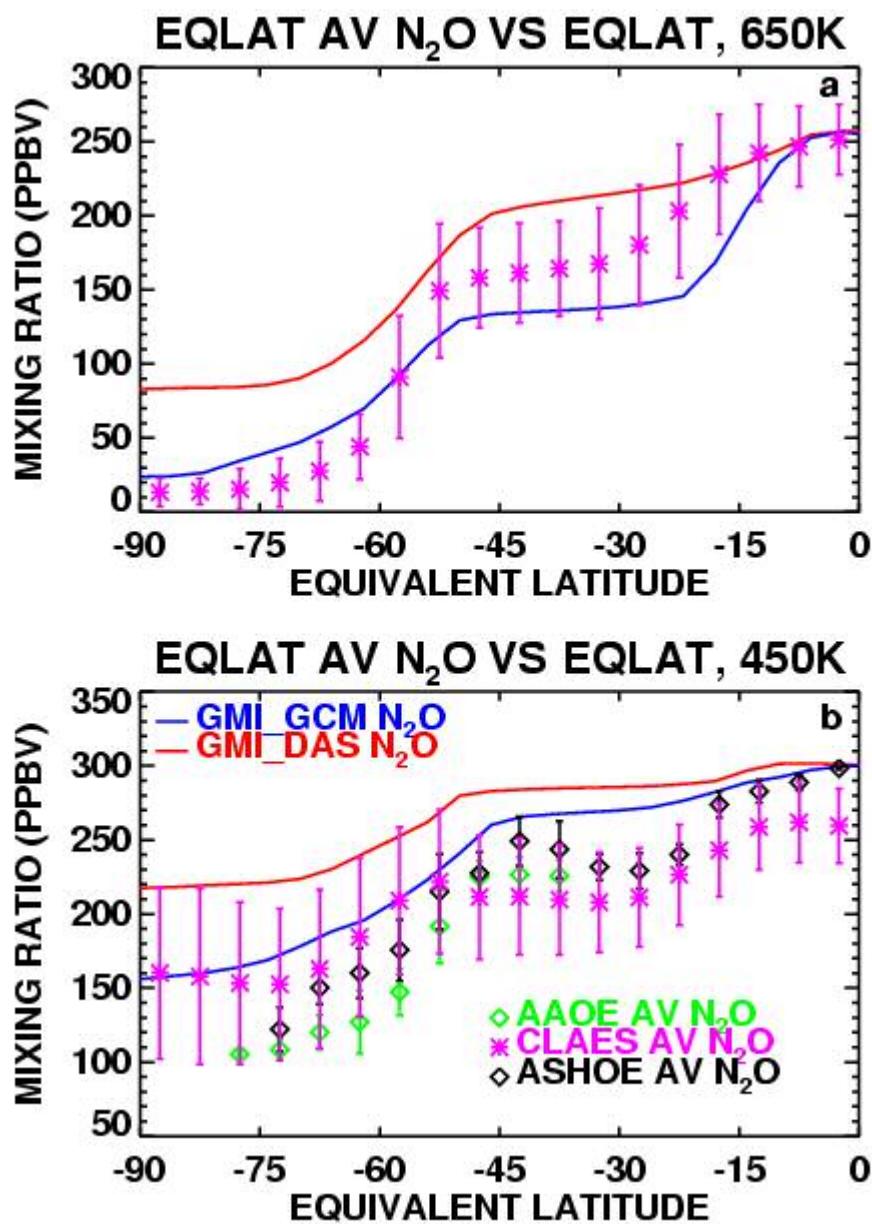


Figure2

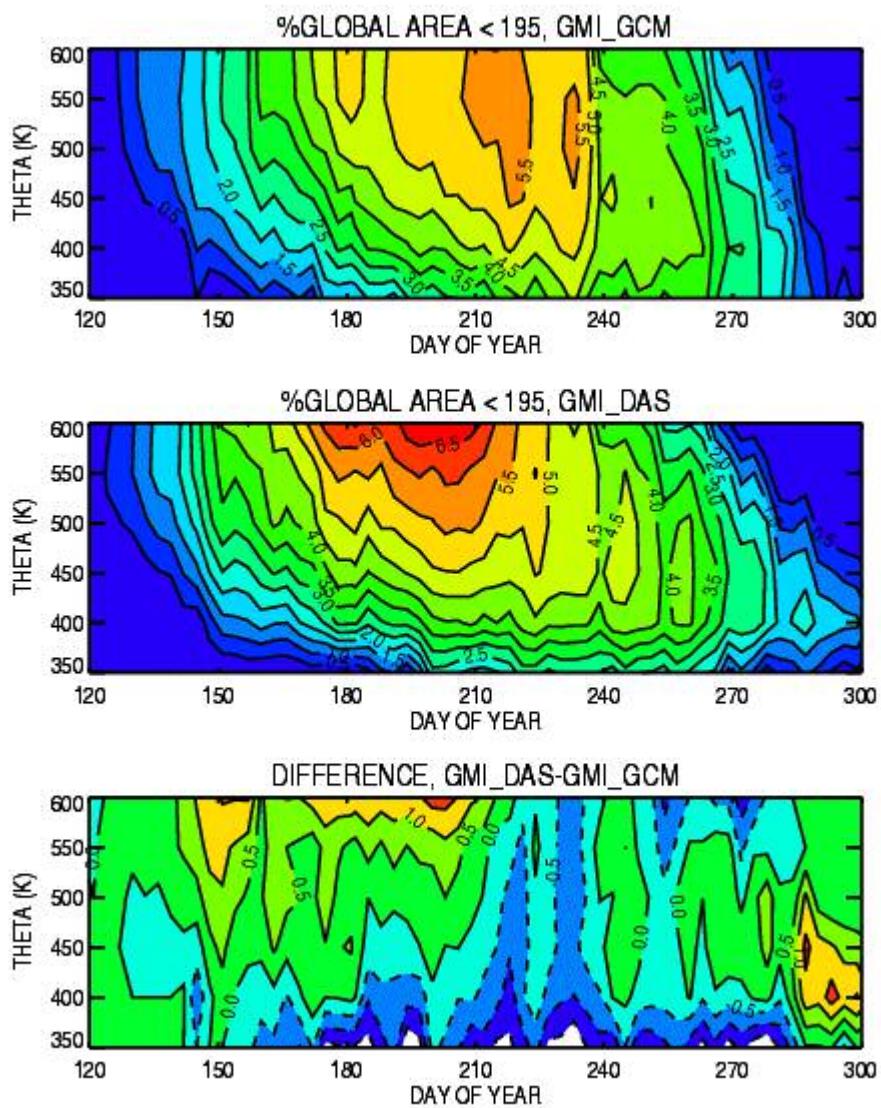


Figure3

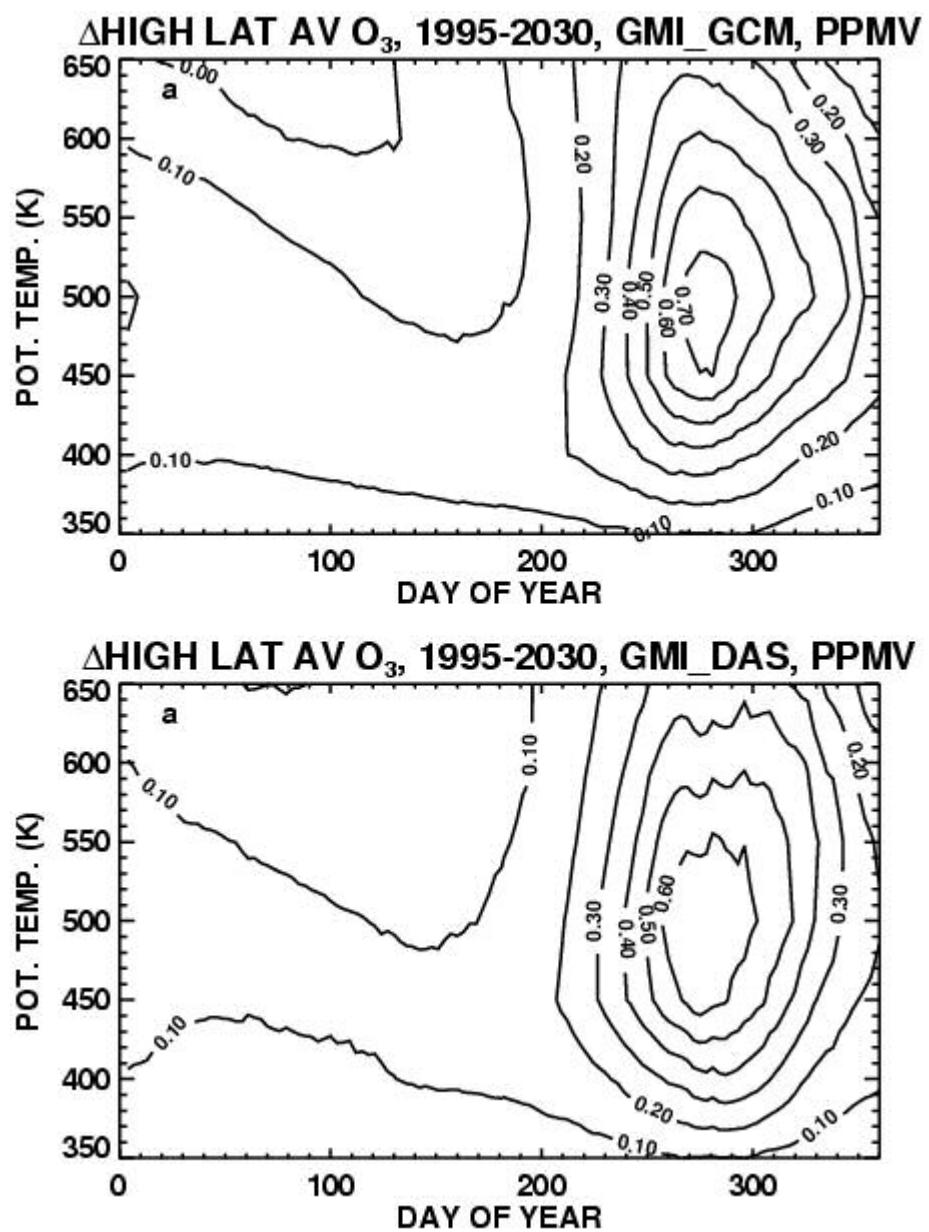


Figure4

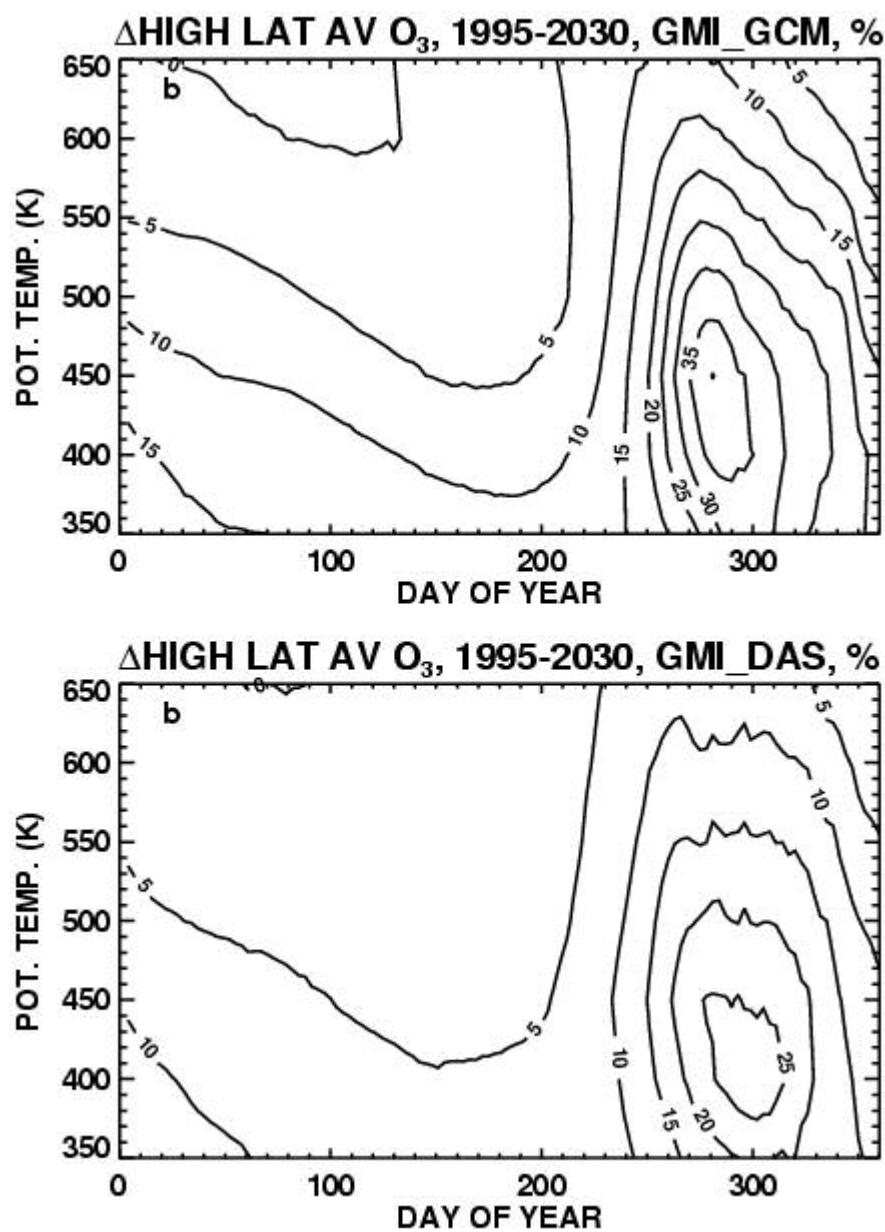


Figure5

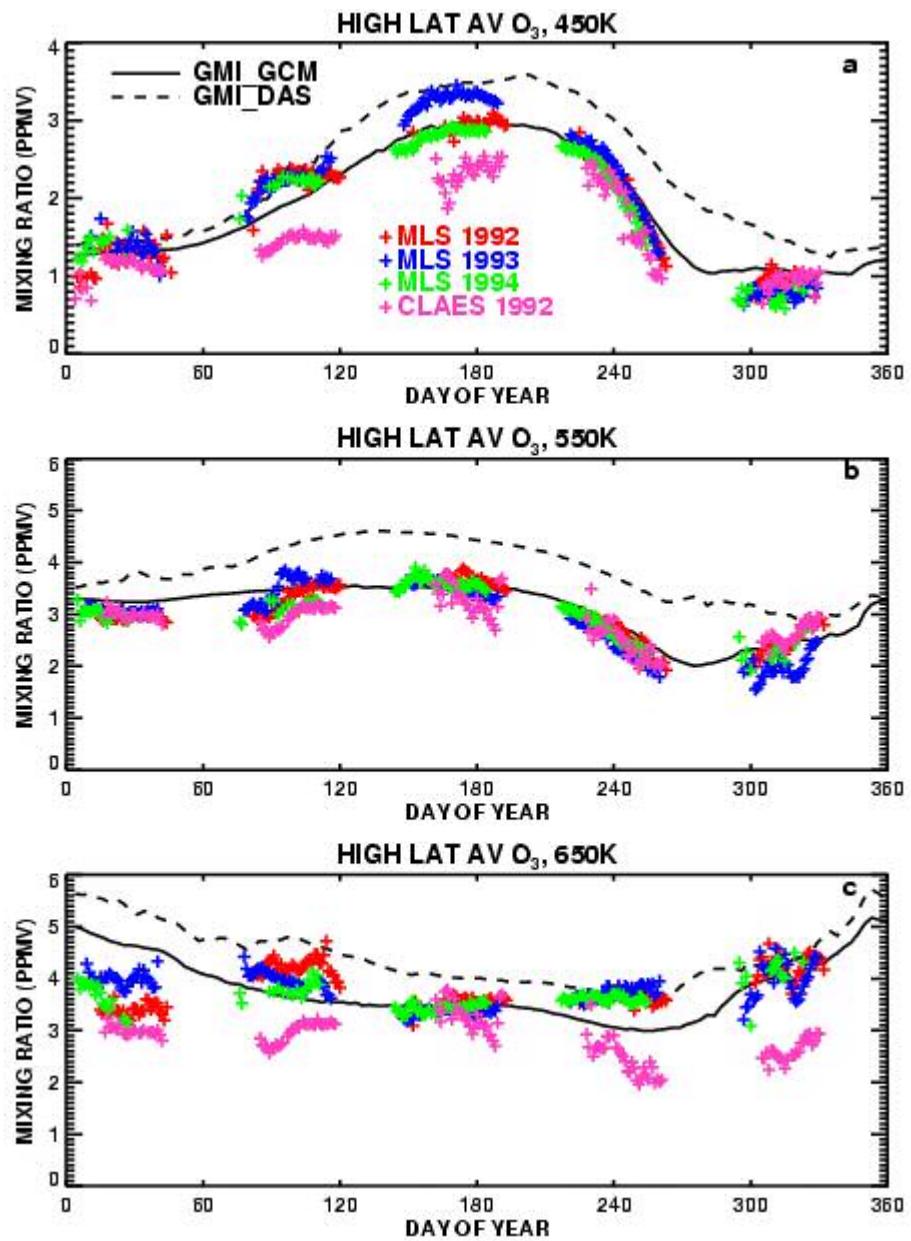


Figure6

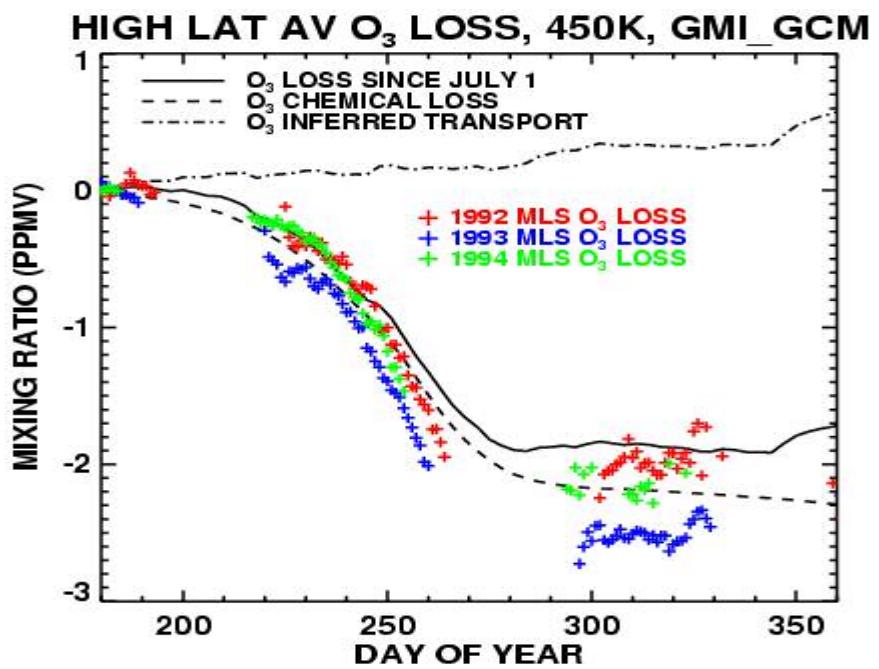


Figure 7a

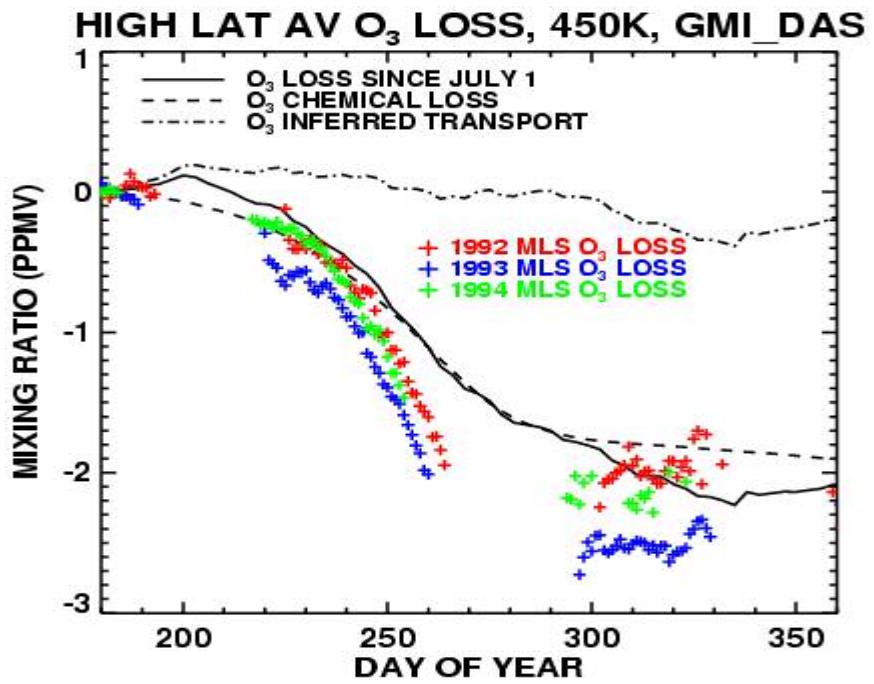


Figure 7b

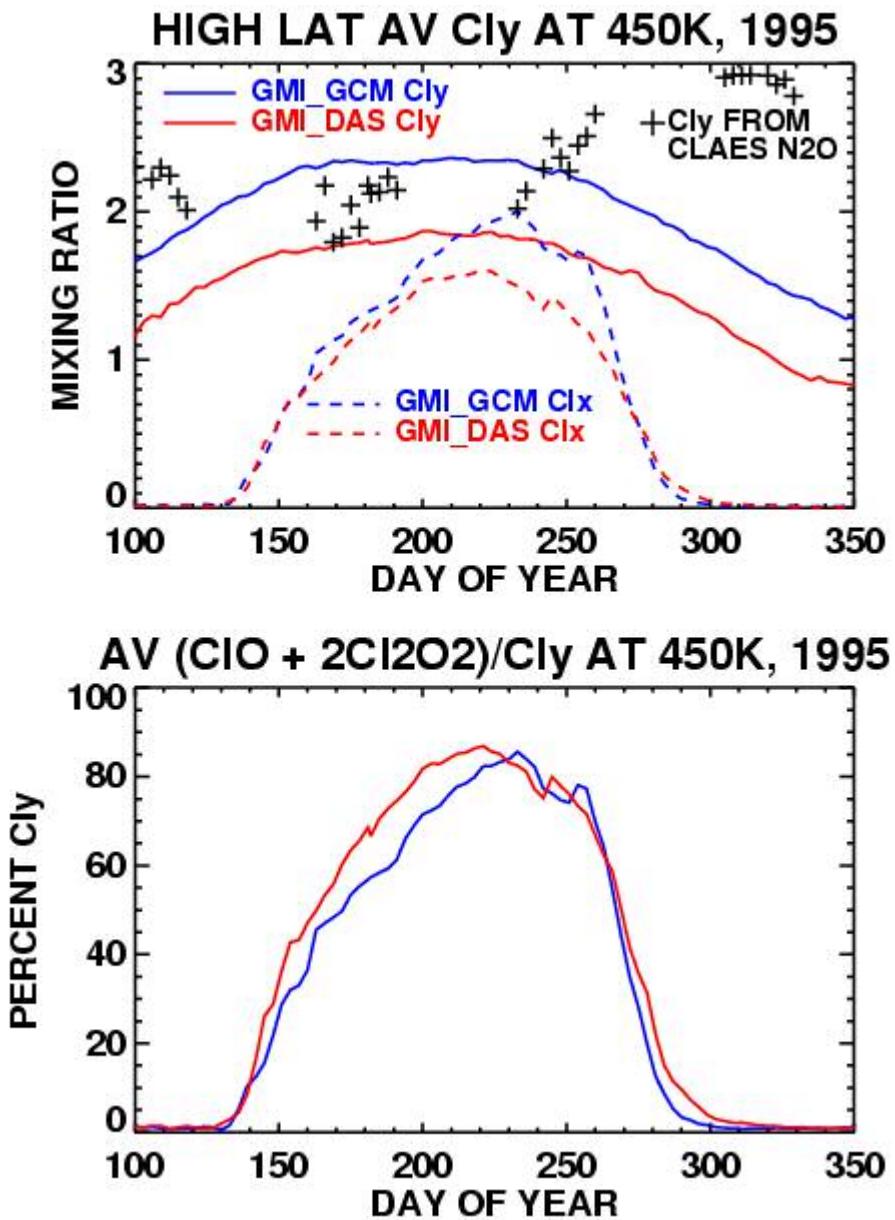


Figure8

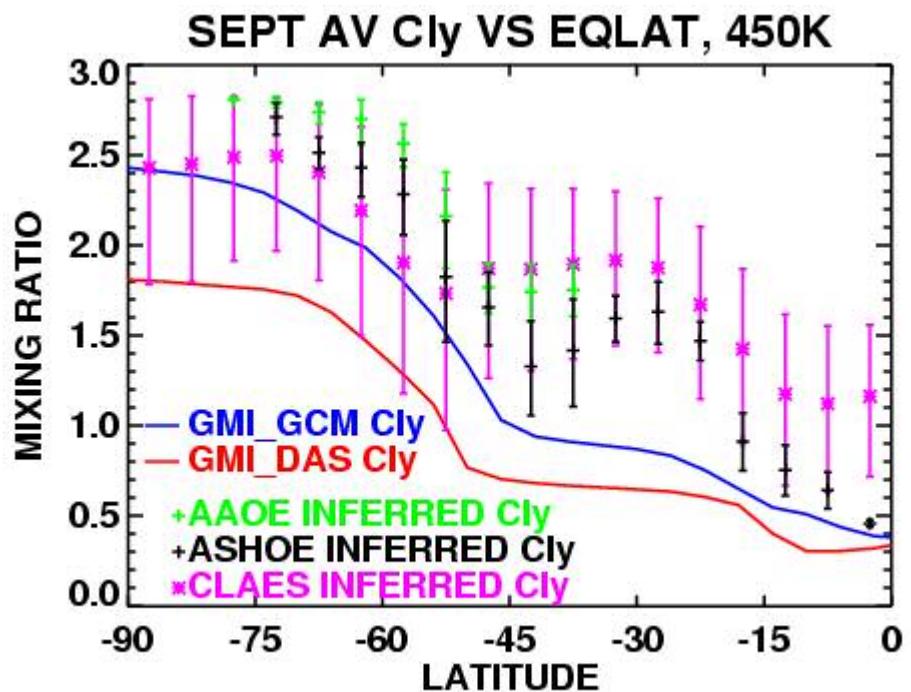


Figure9

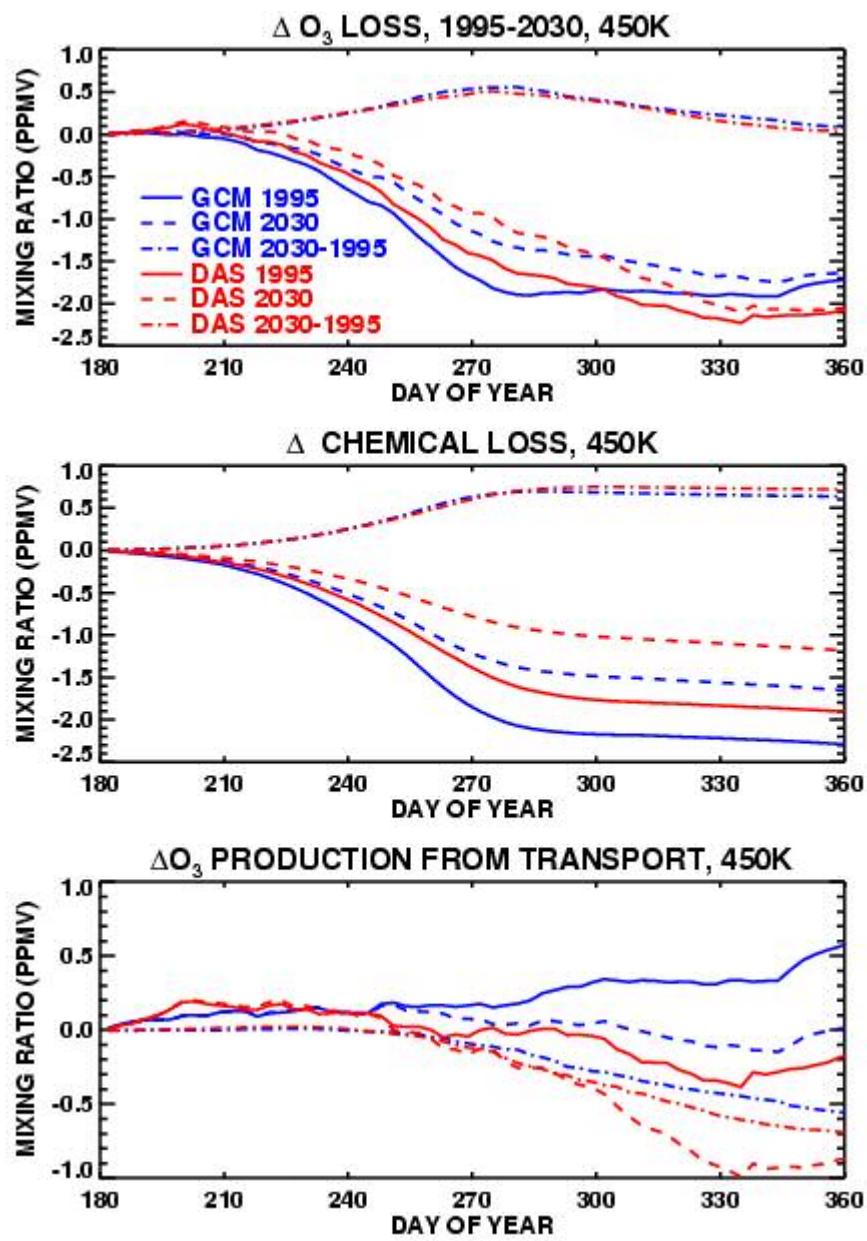


Figure 10

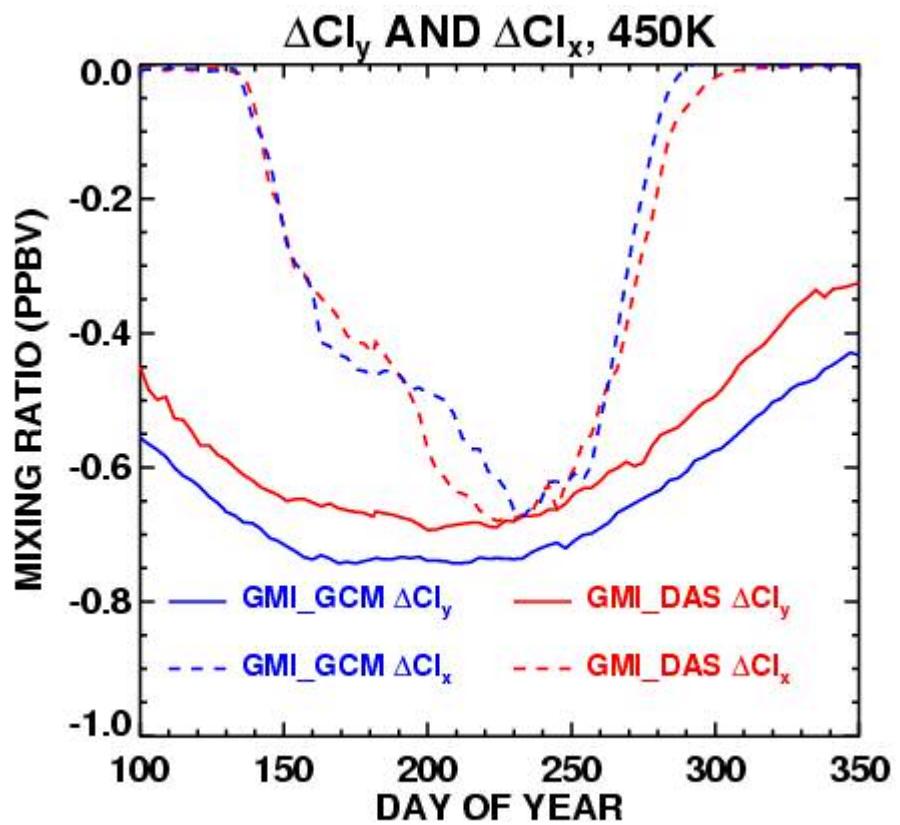


Figure11