3.1   HOW WELL DO CURRENT MODELS REPRESENT CHEMICAL AND PHYSICAL PROCESSES IN THE UPPER TROPOSPHERE AND LOWER STRATOSPHERE?

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1. INTRODUCTION

There remain significant uncertainties in understanding the changes in atmospheric chemical and physical processes affecting ozone and other constituents in the upper troposphere and lower stratosphere (UT/LS), the first few kilometers above and below the tropopause. In recent years, through new insights in atmospheric dynamics coupled with an increase in constituent measurements, the understanding of the UT/LS has evolved greatly. It is no longer adequate to think of the tropopause as being a sharp boundary and transport barrier between the troposphere and stratosphere (Rosenolf, 1995). Wave-induced zonal forces in the extratropical stratosphere induce a global-scale meridional circulation in which mass is pulled upward and poleward in the tropics and pushed downward in the extratropics (e.g., Hoor *et al.*, 2002), transporting air from the troposphere to the stratosphere, and then from the stratospheric "overworld" to the lowermost stratosphere, the "middleworld", where more rapid transport is possible between the stratosphere and troposphere along isentropic surfaces. Logan (1999) suggested that ozone in the lowermost stratosphere builds up during winter in the extratropics as a result of downward transport in the large-scale meridional circulation, and reaches a minimum during autumn as a result of exchange with lower latitudes and the troposphere in spring and summer. During all seasons, there is a transition zone near the tropopause that contains air characteristic of both the troposphere and the stratosphere. The extraction of the net transport still remains uncertain, but may occur either by isentropic transport caused by wave breaking (Holton *et al.*, 1995) or by diabatic cross-isentropic transport associated with synoptic baroclinic disturbances in the subtropical region (Rood *et al.*, 1997).

Given the complexity of the transport and chemical processes in the upper troposphere and lower stratosphere, it is not surprising that the UT/LS is of great interest towards understanding the capabilities to model the processes acting in this region. We are using newest version of the three-dimensional chemical-transport of the global atmosphere, Model for OZone And Related chemical Tracers, called MOZART-3, which has a complete stratosphere and mesosphere as well as troposphere, to try to improve our understanding of the processes that control the chemical composition and structure of the upper troposphere and lower stratosphere, while at the same time trying to understand the capabilities of the model. Comparisons are being made, wherever possible, with available observations, to examine the current theoretical understanding of the UT/LS region. As a result of these studies, we hope to better understand the capability of a state-of-the-art model like MOZART-3 to evaluate the representation of the model in terms of chemical and physical processes in the upper troposphere and lower stratosphere (UT/LS)

2. MODEL DESCRIPTION

The MOZART-3 model is developed from the MOZART-2 model, which was primarily designed for studies of tropospheric processes. Although MOZART-2 extends to 3 hPa (about 35 km), the representation of chemical processes focuses on the troposphere, whereas MOZART-3 is extended to include the entire troposphere, stratosphere and mesosphere. A short description of MOZART-2 and the major differences found in MOZART-3 are given below. A more complete description of MOZART-2 and its capabilities is given in Horowitz *et al.* (2002). The earlier initial version of MOZART is described in Brasseur *et al.* (1998) and Hauglustaine *et al.* (1998).

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Using meteorological fields (global wind, temperature, humidity, cloud fields, convective mass fluxes and diffusion parameters) from the NCAR Whole Atmospheric Community Climate Model (WACCM, up to approximately 140 kilometers), MOZART-3 has a horizontal resolution of about 2.8° latitude by 2.8° longitude, with 64 levels in the vertical. MOZART-3 can also use assimilated meteorological fields, but have not been fully tested. However, we have used National Center for Environmental Prediction (NCEP) reanalyses in studies of PEM-TROPICS data with MOZART-2. The MOZART-2 model includes a mass conserving advection scheme based on the work of Lin and Rood (1996). The chemical scheme includes 140 chemical and photochemical reactions, as well as washout for 10 soluble species. The distributions of more than 50 species are calculated, including O_3, HO_x, NO_x, methane, and a number of non-methane hydrocarbons (NMHC). In addition, the most recent version of MOZART-2 includes a detailed representation of the mass mixing ratio of sulfate, black carbon, and ammonium (Tie et $al.$, 2001) and, more recently, mineral dust aerosol abundance. These aerosols are primarily derived from precursor emissions at the surface. Heterogeneous chemical processes are also included on these aerosol types. Although concerns about mass consistency in the model have been fully resolved in the latest version, the existing MOZART-2 model still tends to overestimate ozone in the upper troposphere at high northern latitudes, probably as a result of too much cross-tropopause transport of ozone although too little vertical resolution in the tropopause region may also contribute (Horowitz et $al.$, 2002). Nonetheless, in general, the model does an excellent job of capturing the characteristics of tropospheric chemistry processes.

With the extension into the upper atmosphere, the MOZART-3 model currently includes 90 chemical species and approximately 250 photochemical reactions. Essentially all of the chemical constituents and associated chemical reactions and photochemical reactions important to the stratosphere and mesosphere have been included in MOZART-3. Heterogeneous processes on sulfate aerosols and polar stratospheric clouds (Type 1a, 1b, and 2) are included. Denitrification and dehydration processes are also represented.

3. RESIDUAL CIRCULATION AND MIXING IN THE LOWERMOST STRATOSPHERE

The meridional circulation of the stratosphere is also known as the "Brewer-Dobson circulation" based originally on observations of stratospheric water vapor (Brewer, 1949) and stratospheric ozone (Dobson, 1956). This circulation comprises of upwelling in the tropics and subsidence in the middle and high latitudes in the summer time, whereas the circulation is from the tropics into high latitudes in the winter time (Rosenolf, 1995). Since the stratosphere is characterized by strong stability to vertical displacement, most tracers show horizontal gradients between regions of mean upward motion and downward motion. N_2O serves as a useful tracer of stratospheric transport. Its only sources are in the troposphere, and it is long lived in the stratosphere. It is also the primary stratospheric source of reactive nitrogen NOy, and the observed correlation between NOy and N_2O in the lower stratosphere is well defined and linear.

Figure 1 is one example of the characteristic shape for monthly zonal average distribution of N_2O, as measured by the Cryogenic Limb Array Etalon Spectrometer (CLAES) on the Upper Atmosphere Research Satellite (UARS) (Douglass et $al.$, 1999). The characteristic shape for the long-lived tracers, such as N_2O, is its isopleths bulging upward in the tropics and poleward/downward in the extratropics. Especially in the winter or spring hemisphere, there are strong gradients in the subtropics, relatively flat isopleths in middle latitudes, and strong gradients near 60 degrees in latitude in the winter hemisphere. The model results (Figure 2) show similar features to the observations (Douglass et $al.$, 1999; Strahan et $al.$, 1999). Mean N_2O on pressure levels provide a gross measure of the balance between transport by the mean winds and horizontal mixing. Such as, the maximum N_2O at any pressure level is found in the tropics, and the N_2O decreases with altitude at all latitudes. There is outflow from the tropics at all levels associated with upward transport, and the tracer distribution at middle latitudes depends upon a balance between the mean horizontal and vertical transport as well as mixing across the subtropics. In the Northern Hemisphere (NH), the N_2O contours at middle latitudes are flatter during winter season (e.g., January) than Fall (e.g., September) and the high-latitude contours show evidence of winter decent. The sharpness of the horizontal gradient in the NH winter is produced by the combined influence of the residual circulation and the horizontal mixing driven by planetary wave transport (Douglass et $al.$, 1999). This gradient contrasts with the more

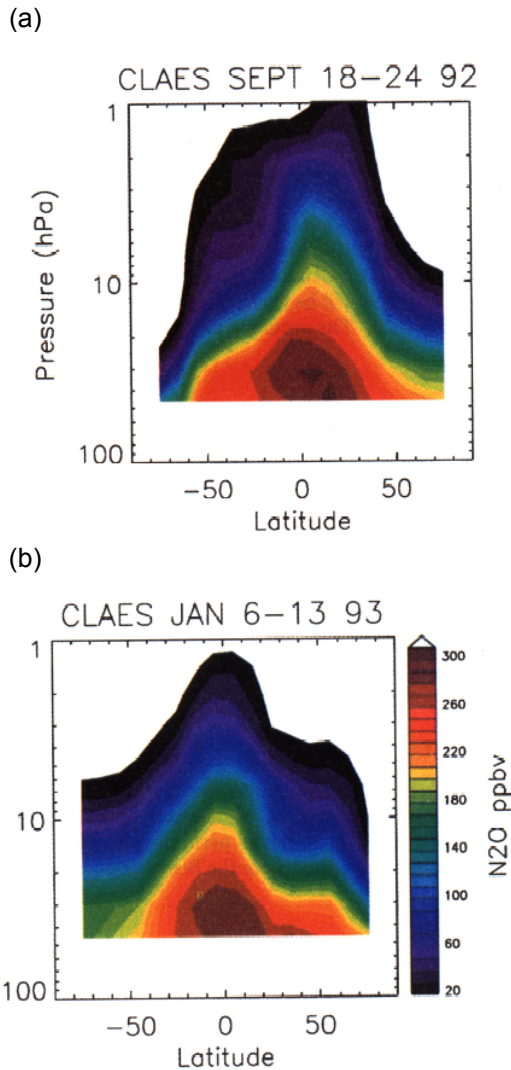constant horizontal gradient seen in the NH autumn.

(a)

(b)

Figure 1. Averaged N$_2$O distribution based on CLAES observations from (a) September 18 to 24, 1992 and (b) from January 6 to 13, 1993 (right) (Douglass *et al*., 1999)

A sense of the importance of the transport processes can be gained by considering vertical profiles from comparison between model and observations. Strahan *et al*. (1999) used the measurements of the ATLAS instrument on the NASA ER-2 aircraft between August 1988 and September 1997 to construct a climatology for N$_2$O from the upper troposphere to the lower stratosphere. The vertical range for the observation is between 360 K to 530 K. The data reveal seasonal dependence, such as lower mixing ratios of N$_2$O in the middle latitude during
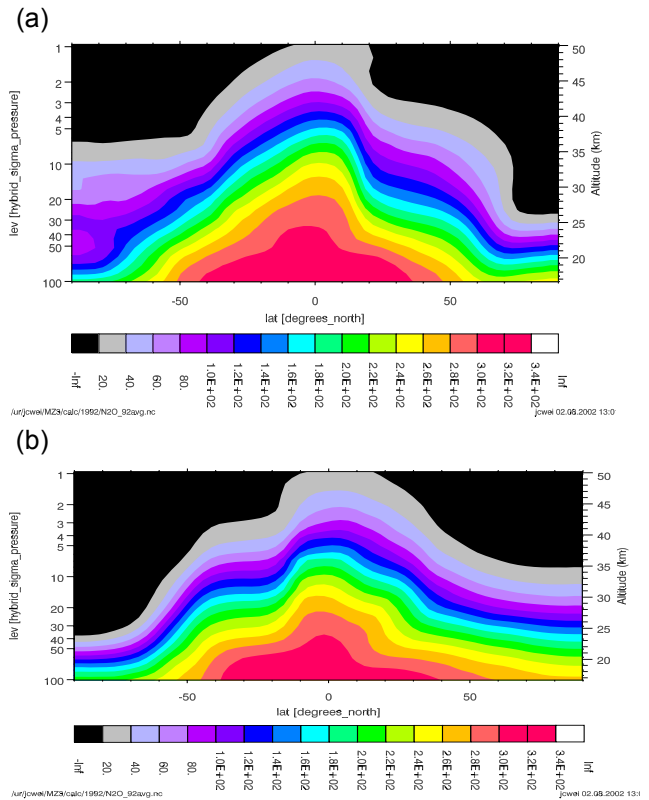
the winter than in the fall (one example flight is shown in Figure 3).

(a)

(b)

Figure 2. Zonal average N$_2$O distribution calculated from MOZART 3 for (a) January and (b) September.
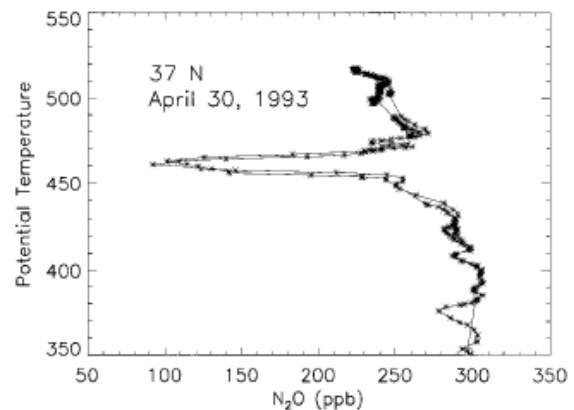

Figure 3. Vertical profile of N$_2$O near 37 N on April 30, 1993 during the Stratospheric Photochemistry Aerosols and Dynamics Expedition (SPADE) (Strahan *et al*., 1999)

In this study, we will first evaluate the ability of MOZART 3 in reproducing realistic transport in the

upper troposphere and lower stratosphere by comparing with long-term observed satellite data. In order to examine the residual circulation and the horizontal mixing in this region more carefully, we will evaluate the vertical profiles from the model calculation for N_2O interpolated on to insentropic surface for the range from the upper troposphere into the lower stratosphere (e.g., 360 K to 530 K). This comparison particularly evaluates the effectiveness of transport processes across the lower boundary the stratosphere.

4. TRACER-TRACER RELATIONSHIP IN EXAMINING TROPOSPHERE-STRATOSPHERE INTERACTION

The so-called "middle world" is right above the tropopause in the extratropoics bounded between isentropic surface at 380 K and PV at 2 PVU (potential vorticity unit) (Holton *et al.*, 1995). However, the tropopause is not really a rigid barrier in the atmosphere. Hoor *et al.* (2002) used tracers like O_3-CO correlation to investigate seasonal variation of cross-tropopause exchange in the extratropics and concluded that during the winter time the tropospheric influence only can reach from local tropopause up to potential temperature at 330 K (Figure 4a) and is most probably caused by isentropic cross-tropopause transport at the polar front. When polar vortex breaks during late winter or early spring, airmass may transport downward from higher latitudal stratosphere to lowermost stratosphere due to diabatic transport, may even transport cross-tropopause to troposphere due to isentropic transport and depends on the strength of polar vortex During the summer time (Figure 4b), tropospheric influence is more apparent at potential temperature at 360 K.

Plumb and Ko (1992) proposed that if the tracers are in "slope equilibrium", i.e., if quasi-horizontal mixing along isentropes is much faster than the residual advection (which ensures that isentropic gradients are weak) and if both are much fast than chemical processes, then we should expect to see points on scatter plots of long-lived tracers form "smooth compact curves or lines". Therefore, with different chemistry mechanism in different long-lived tracers, the correlation in different tracer pairs should have its own curvature or line in the relationship. This "slope equilibrium" concept has been extensively examined and is now being applied in the tracer-tracer correlation.
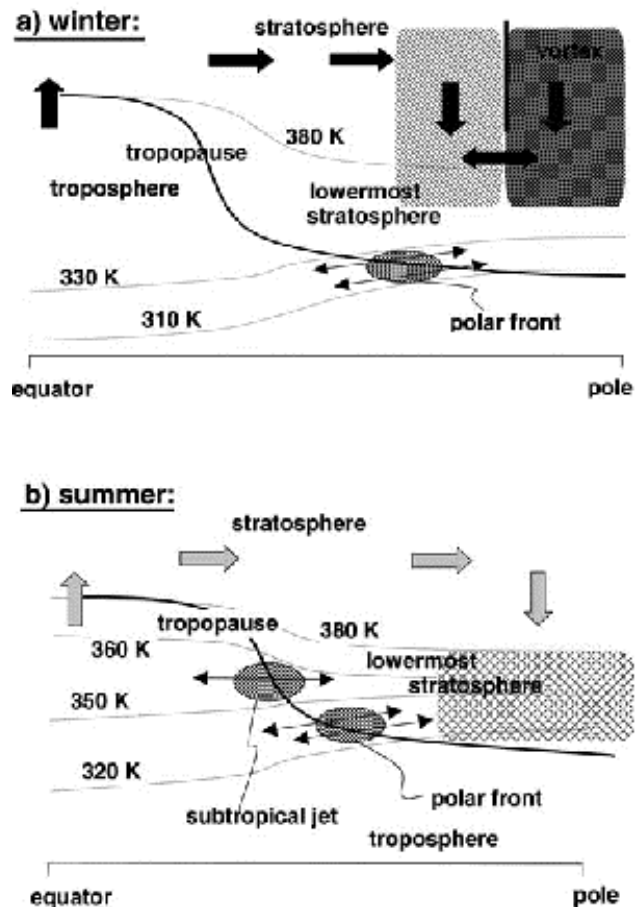

Figure 4. Diagram of seasonal transport into the lowermost stratosphere derived from STREAM measurements (Hoor *et al.*, 2002).

Correlations between stratospheric abundances of O_3 and N_2O or O_3 and CH_4 have proven to be an invaluable tool for distinguishing and making comparisons between ozone-depleted Antarctic or Arctic vortex air masses and those, either inside or outside the vortex, that are depleted in ozone (Bregman *et al.*, 1995, 1997, 2000, 2001; Michelsen *et al.*, 1998). CO-O_3 correlations have been used to examine the seasonal variations in the mixing layer in the lowermost stratosphere (Hoor *et al*, 2002).

Existing studies of the correlations between 3-D model calculated O_3, N_2O, NOy, and CO in the lowermost stratosphere when compared with available observations indicate that the best agreement between models and observations is found during winter and the worst comparison occurs during summer, where most models underestimate the mean ozone concentrations (partly due to inaccurate description of the relatively small-scale transport processes, mainly

associated with convective activity and partly by incomplete ozone chemistry in the parameterized scheme) (Logan *et al*., 1999). Reasons for these discrepancies may be due to the input meteorological data, the model formulation of O_3 transport downward from the stratosphere, including the way O_3 is prescribed at the top boundary, and poor vertical and/or horizontal resolution around the tropopause.

CO-O_3 scatter plot can be used to identify a mixing layer in the UT/LS and quantify the isentropic range of tropospheric influences. The mixing ratios of O_3 and CO exhibit strong gradients at the tropopause, with high CO (low O_3) values in the troposphere and low CO (high O_3) in the stratosphere. Therefore, the idealized relationship between O_3 and CO can be expected to display an L shape (Fischer *et al*., 2000). In the vicinity of tropopause region, this correlation is highly nonlinear due to the different chemical compositions of the stratosphere and troposphere. Mixing between the troposphere and the stratosphere will result in linear mixing lines connecting both stratospheric and tropospheric airmasses.
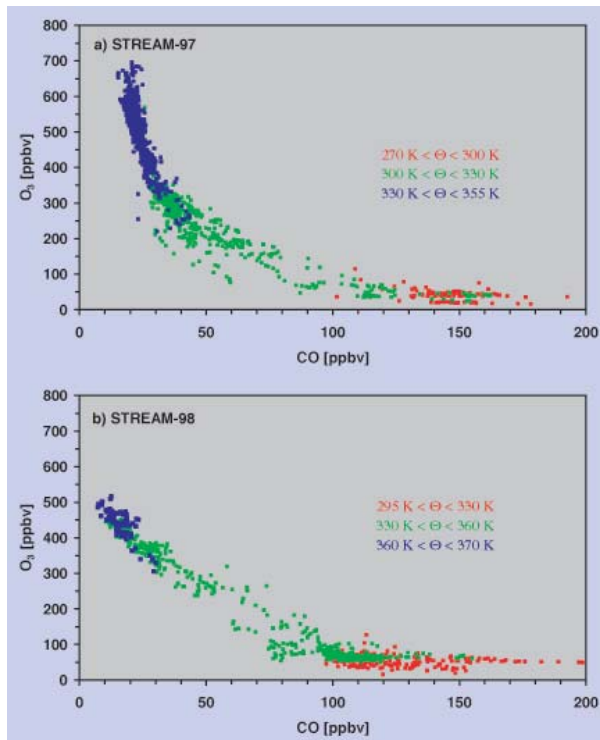

Figure 5. Correlation in CO-O_3 from STREAM 1997 March (top) and 1998 July (bottom) measurements. Points are grouped according to different potential temperature. (Hoor *et al*., 2002).

Figure 5 shows scatter plots of O_3-CO obtained during two STREAM (Stratosphere TRoposphere Experiments by Aircraft Measurements) campaigns from Kiruna, Sweeden in early March 1997 (top) and Timmins, Canada in July 1998 (bottom) (Hoor *et al*., 2002). Cross-tropopause mixing can be identified by deviations from the L-shape for CO mixing ratios between 30 and 100 ppb (green dots). During the summer time, the mixing lines seems to have stronger tropospheric air influences into this region (bottom, between 330 K and 360 K), whereas during the winter time the mixing line seems to extend to the highest O_3 mixing ratios (top, stratospheric air influences between 300 K and 360 K). We will be presenting comparisons with such datasets.

5. CONCLUSIONS

Although more specific analyses will be presented at the AMS meeting, this limited discussion has shown that the MOZART-3 model has generally a good representation of simulating transport in the upper troposphere and lower stratosphere. However, there are still some discrepancies in the model and measurements especially in the UT/LS. In order to fully understand the processes by which the stratosphere interacts with the troposphere or the other way around, we will conduct a series of analyses by comparing model calculations and available observations. A number of these will be presented at the meeting.

6. ACKNOWLEDGEMENT

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