Analysis of residual mean transport in the stratosphere. Part II: Distributions of  $CO_2$  and mean age.

by

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## ABSTRACT:

Distributions of  $CO_2$  and the mean age of stratospheric air are examined using the interactive two-dimensional model described by Schneider et al. (1998). It is shown that the model can explain the distribution of age in the lower stratosphere and reproduce the correlations between  $CO_2$  and  $N_2O$  observed at mid latitudes in the lower stratosphere. Seasonal characteristics of the measured correlations are captured by the model. The model uses externally specified coefficients,  $K_{yy}$ , to account for large scale isentropic mixing in the stratosphere. The sensitivity of calculated distributions of  $CO_2$  to  $K_{yy}$  is be examined by comparing model runs using three different distributions of  $K_{yy}$ . It is shown that the  $CO_2$  measurements require  $K_{yy}$  to exceed a threshold value of approximately  $10^5 \text{m}^2 \text{sec}^{-1}$  for most of the year. Once this threshold is exceeded, the slope of the  $CO_2/N_2O$  correlations depends only weakly on the magnitude and the structure of the distribution of  $K_{yy}$ .

# 1. INTRODUCTION:

The existence of an air mass boundary in the subtropics of the stratosphere has been established by observations of aerosols (Trepte and Hitchman, 1992; McCormic and Veiga, 1993),  $N_2O$  and  $H_2O$  (Randel et al, 1993) and  $NO_y$  and  $O_3$  (Murphy et al, 1993). The isolation of the tropics from the midlatitudes and its implications for distributions of trace species have been described in the conceptual model of a "tropical pipe" (Plumb, 1996).

The subtropical barrier is not, however, an impermeable boundary. The rate of transport between the tropics and the midlatitudes of the stratosphere, sometimes referred to as the "leakiness of the tropical pipe", has an important influence on the distributions of tracers in the lower stratosphere. Transport across the subtropical barrier has been investigated using assimilated winds by Chen et al. (1994) and Waugh (1996). In accordance with the above observations and the analyses of ozone data (Leovy et al., 1985; Manney et al., 1993), the assimilated winds give rise to transport out of the tropics during Rossby wave breaking events which result in filaments being drawn into midlatitudes. While the observational analyses mentioned above and the 3-D model calculations find for the most part transport out of the tropics at stratospheric levels, Minschwaner et al. (1996) have estimated that midlatitude air is entrained into the tropical upwelling region significantly at altitudes above 22 km. This study was based on measurements of N<sub>2</sub>O and CCl3F3.

In Part I of this paper (Schneider et al., Analysis of residual mean transport in the stratosphere. Part I: Model description and comparison with satellite data, submitted to *J. Geophys. Res.*, 1998), we described an interactive two-dimensional (2-D) residual circulation model of the stratosphere and examined distributions of long lived tracers at altitudes for which satellite data are available. The model was shown to produce tropical and midlatitude profiles of long lived tracers in reasonable agreement with observations. In the following, we will analyse exchange of mass between tropics and midlatitudes at stratospheric altitudes by comparing model calculations of mean age and  $CO_2$  to observations.

As discussed in Schneider et al. (1998), the most important factors that determine the rate of transport between tropics and midlatitudes in the model are:

(1) Quasi - horizontal diffusion is acting in mid and high latitudes only. The magnitude of the diffusion coefficients decreases smoothly toward the subtropics. Transport within the tropics and across the subtropics is dominated by advection which is calculated to be consistent - within the framework of the Transformed Eulerian Mean (TEM) theory - with the large scale diffusion specified in the extratropics.

(2) The nonlinear momentum advection terms are included in the dynamics module and have a significant effect on the circulation near the equator and on the exchange of mass across the sub-tropical air mass boundary.

(3) Mechanical damping in the lower stratosphere is assumed to be small. Damping time scales are longer than a season.

We will show that the model reproduces the measured distribution of mean age in the lower stratosphere fairly well and that both the slope and seasonal behavior of measured correlations between  $N_2O$  and  $CO_2$  (Boering et al., 1994) are well simulated by the model.

The sensitivity of calculated distributions of  $CO_2$  to values assumed for extratropical Rossby wave mixing coefficients,  $K_{yy}$ , will be examined by comparing model runs using three different specifications of  $K_{yy}$  as described in Schneider et al. (1998). It will be shown that the  $CO_2$  measurements require  $K_{yy}$  to exceed a threshold value of approximately  $10^5 m^2 sec^{-1}$  for most of the year. Once this threshold is exceeded, the slope of the  $CO_2/N_2O$  correlations depends only weakly on  $K_{yy}$ .

In addition, the sensitivity to the treatment of the troposphere in the model and to the meridional coupling provided by gravity wave drag in the lower stratosphere will be examined. In order to reproduce the measurements, the timescale for mechanical damping has to be larger than a season in the altitude range between 10 and 20 km. Calculated distributions of  $CO_2$ depend on the parameterization of the tropospheric heating rates and the matching of these rates with calculated rates for stratospheric heating. The treatment of the tropospheric lower boundary clearly affects the rate of upward mass flux in the lowest layers of the tropical stratosphere and, therefore, the speed at which tropospheric seasonal variations and the annual increase of  $CO_2$  are propagated into the stratosphere. The vertical velocities also determine the rate of adiabatic cooling. In fact, the matching of the heating rates was implemented initially to ensure that the temperature at the tropical tropopause agreed with observations. Correlations of  $CO_2$  and  $N_2O$  are not significantly affected so long as temperatures at the tropical tropopause are within a few degrees of observed values.

Because of the spacing of vertical layers in the model (2 km) no claim is made that the vertical structure of transport in the lower stratosphere is resolved. Parameters like the rate of mechanical dissipation are by necessity bulk values for fairly deep layers. Therefore, the following statements about sensitivities of the model results to parameters are valid only for the resolution used in the current version and are consequently preliminary.

## 2. THE DISTRIBUTION OF MEAN AGE.

The concept of the mean age of stratospheric air has been introduced by Hall and Plumb (1996). It is defined as the time required for a parcel of air to transit from its entry point into the stratosphere, the tropical tropopause, to a given location. The mean age and even more so the age spectra contain important information on the transport of mass in different models. Ages can be derived for the stratosphere by measuring the concentrations of tracers that are long lived and linearly increasing in the troposphere. Mean ages have been derived for the lower stratosphere from observations of SF<sub>6</sub> (Geller et al., 1997) and CO<sub>2</sub> (Boering et al., 1996).

The mean age can also be thought of as the concentration of a tracer with a constant source in the stratosphere and a sink in the troposphere (Boering et al., 1996). This definition of age is equivalent to the transit time, except that it allows for influx of young, tropospheric air through the tropopause in mid and high latitudes. Upward transport in the extratropics can occur through vertical exchange processes in the troposphere that extend into the tropopause region. Therefore, the two definitions of mean age give different results in the vicinity of the extratropical tropopause. The differences diminish rapidly with vertical distance from the tropopause.

Viewed as a tracer with a stratospheric source, the concept of mean age can be used to test the capability of models to simulate the distribution of exhaust products from a planned fleet of stratospheric aircraft. Both age and aircraft  $NO_y$  have stratospheric sources. The source distributions of the two quantities differ spatially and temporally. In addition,  $NO_y$  has a photochemical sink in the upper stratosphere. In the lower stratosphere, however, both tracers should be affected similarly by errors in the rates of transport.

The distribution of mean age, calculated for the low  $K_{yy}$  case as defined in Schneider et al. (1998) is shown in Fig. [1] for the Northern hemisphere Fall and Winter. Starting from the tropical tropopause, age increases rapidly with altitude for a few kilometers. A minimum in the diabatic heating rates is situated near 20 km in the tropical stratosphere. This is sited below the ozone peak. Most of the ultra violet radiation from the sun has been absorbed at this altitude and the flux of upwelling infrared radiation is determined by the temperatures near the cold tropopause. Vertical velocities are, consequently, small in the lower tropical stratosphere.

At altitudes above 25 km, heating rates and vertical velocities increase and the vertical gradient in mean age diminishes accordingly. Above 40 km, horizontal zonal mean velocity components increase also. Above 50 km, horizontal velocities reach several meters per second. Speeds of the same order of magnitude are shown by Eluszkiewicz et al. (1996) for the residual circulation diagnosed from atmospheric observations. Strong meridional flow towards the winter pole is required to satisfy the momentum budget needed to decelerate the polar night jets in the mesosphere. As a consequence of the short (in relation to a season) advective timescales coupled with rapid mixing in the extratropics, the whole region above 40 km is characterized by weak gradients in age. In the model, age differences barely exceed a year.

We are not aware of direct measurements of mean age at mesospheric levels in the tropics. Calculated profiles of  $N_2O$  and  $CH_4$ , shown in Schneider et al. (1998) agree reasonably well with observations, lending confidence to the accuracy of calculated mean rates for upwelling in the tropics. Measurements of mean age in the mesosphere at  $60^\circ$  N by Harnish et al. (1996) indicate that the air at this location is twice as old as calculated in the model. If these measurements are correct, important physical processes are not represented in the model. Our current understanding is that the temperature structure of the mesosphere, which is captured by the model, is determined by adiabatic processes. Downwelling over the winter pole results in some of the warmest temperatures in the stratosphere and upwelling in the summer polar regions leads to temperatures that are anomalously cold. To satisfy mass continuity, strong horizontal motions are required, and these would not allow large gradients in age to persist between the tropics and high latitudes.

Once an air parcel has reached the descending branch of the Brewer - Dobson circulation, its age increases with decreasing altitude. However, the parcel is mixed with younger air that has been transported out of the tropical region at lower altitudes and has been distributed across midlatitudes by large scale mixing. The rate of outflow from the tropics varies with altitude. In the model, we obtain moderate outflow at low stratospheric altitudes and strong outflow at altitudes over 30 km with reduced rates in between. This leads to a reversal in the vertical gradient of age in high latitudes. A maximum in age appears in the annual mean at about 25 km in the polar regions. The altitude at which age becomes younger varies seasonally as can be seen in Fig.[1]. Data shown by Harnish et al. (1996) indicate that such a reversal may occur at some altitude. However, we do not intend to read too much into this reversal in the model as the circulation above 30 km is affected by gravity wave drag, effects of which are not well constrained in the region between 30 and 45 km.

The calculated distribution of mean age at 20 km is compared with ages derived from measurements of  $SF_6$  in Fig. [2].  $SF_6$  concentrations were measured by an instrument flown on the ER-2. The ages inferred were provided by J. Elkins (personal communication). The curves display calculated age for all four seasons. Seasonal variations of mean age at this altitude are generally of the order of half a year. The latitudinal gradient of age at low and mid latitudes

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appears to be represented well in the model. The data at high altitudes indicate a large amount of scatter. The version of the model used here does not attempt to simulate the dynamics of polar vortices.

The dependence of calculated mean age on Rossby wave diffusion coefficients in the extratropics is similar to that of other long lived tracers such as N<sub>2</sub>O as described in Schneider et al. (1998). Ages obtained for the low and high  $K_{yy}$  case are compared in Fig.[3]. The distribution at the altitude of 20 km for the high  $K_{yy}$  case is compared to the measurements in Fig.[4]. Increasing extratropical diffusion causes air parcels to undergo more north - south excursions at a given altitude, resulting in an increase of mean age. At the same time, the mean meridional circulation is intensified, resulting in a faster rate of overturning of the stratosphere and younger ages overall. The change in overturning rates is somewhat diminished again by altered ozone distributions. The net effect of the competition between the *slope flattening* and *slope steepening* effect of  $K_{yy}$  (Holton, 1986) is that the air becomes slightly younger for higher diffusion. Differences exceed half a year only in small areas of the high latitude lower stratosphere. The uncertainty of age derived from SF<sub>6</sub> is also about half a year due to difficulties in determining the age which are associated with latitudinal gradients of SF<sub>6</sub> at the tropical tropopause.

Although differences in age of half a year may seem insignificant, the change in overturning rates associated with a doubling of extratropical large scale diffusion coefficients results in substantial differences in the vertical velocity at 100 mb as shown in Fig.[5a]. Percent differences for the Northern mid and high latitudes are shown in Fig. [5b]. The changes amount to about 30%, comparable to uncertainties associated with attempts to use observations to determine the flux of mass through the 100 mb level. (Rosenlof, 1995; Eluszkiewicz et al., 1996). Temperature changes associated with the change in overturning rates, averaged between 50 and 100 mb, are between 1 and 2 K in the model. It seems, therefore, that age needs to be known to a fairly high precision in order to reduce current uncertainties in estimates of the mass exchange between stratosphere and troposphere.

Calculated correlations between age and  $N_2O$  are shown in Fig. [6].  $N_2O$  is long lived in the lower stratosphere only. Therefore the relation between the two tracers is not globally compact. The symbols in Fig. [6] indicate correlations for different latitude regions. Compactness is lost with altitude (low  $N_2O$ ) first in the tropics. Correlations between  $40^\circ$  and  $60^\circ$  N remain fairly compact to high altitudes. This pattern is consistent with the observed downward slope of the isolines of  $N_2O$  between the tropics and high latitudes and the distribution of photochemical loss rates for  $N_2O$  in the stratosphere. Ages on  $N_2O$  surfaces derived from measurements of  $CO_2$  has been presented in Boering et al. (1996). The lower envelope of the calculated correlations agrees with these observations to within half a year.

# 3. SIMULATION OF CO<sub>2</sub> IN THE STRATOSPHERE.

Comparison of model generated  $CO_2$  distributions with the measurements in the stratosphere provides a stringent test for the accuracy of the circulation as simulated by the model for the lower stratosphere.  $CO_2$  is controlled by processes at the earth surface. Its concentrations exhibit a long term trend, on which a seasonal signal is superimposed. A maximum is observed in late spring at the mid and high latitudes of the Northern Hemisphere. The seasonal variation observed in the tropics and in the Southern Hemisphere is much smaller.

CO<sub>2</sub> concentrations have been measured with high precision using ER-2 based instruments during the Stratospheric Photochemistry, Aerosols and Dynamics expedition (SPADE) and the Airborne Southern Hemisphere Ozone Experiment and Measurement for Assessing the Effects of Stratospheric Aircraft (ASHOE/MAESA) (Boering et al., 1994). Additional data have been gathered since. In this paper, we will concentrate on the SPADE ASHOE/MAESA measurements in order to analyse the seasonality of mass exchange between tropics and mid latitudes. SPADE data are available for the Fall of 1992 and for the Spring and Fall of 1993. The ASHOE/MAESA data were taken in the Spring and Fall of 1994.

The seasonal signal has been detected at low altitudes in midlatitudes of the stratosphere about four months after it occurs at the surface. This time lag is too short for the signal to have propagated to high altitudes first and then have been advected subsequently downward by the diabatic circulation. After being transported up through the tropical tropopause, some material is advected into midlatitudes at low stratospheric altitudes. Whatever passes through the subtropical barrier is distributed latitudinally by planetary wave mixing on a timescale of weeks to months. At higher altitudes, the seasonal signal is washed out, but the annual increase can be identified.

For model runs, variations of  $CO_2$  at the surface were specified using the NOAA CMDL data (T. Conway, personal communication) from 1980 onward. Integrations were done for 15 years, until the end of 1994. At the start of the model integration, the initial condition for  $CO_2$  was taken as the global average value appropriate for the surface in 1980, constant in latitude and height. It takes at least 5 years of integration for the high latitude distribution of a stratospheric tracer to equilibrate to surface values. Given the annual increase of  $CO_2$ , an integration period longer than 15 years would have been preferable. However, the number of stations

reporting  $CO_2$  before 1980 is small and thus it is difficult to derive monthly mean concentrations as a function of time and latitude.

The oversimplified treatment of the troposphere in our 2-D model presents a problem for simulating tracers with gradients at the surface. Vertical diffusion coefficients are specified in the model troposphere to guarantee a timescale for vertical exchange of about one month for the troposphere. Therefore, existing latitudinal surface gradients near the equator are maintained throughout the troposphere in the model. The gradients can be reduced by adding large horizon-tal diffusion in the troposphere. However, it is a difficult and not very rewarding exercise to adjust these parameters in such a manner that, for example, observed surface gradients of SF<sub>6</sub> (Elkins et al., 1996) can be reproduced when specifying SF<sub>6</sub> emissions in the model and that gradients of CO<sub>2</sub> in the upper troposphere agree with observations (Nakazawa et al., 1991) at the same time.

Differences between the seasonal maximum and minimum of  $CO_2$  at the surface and in the upper tropopause are shown for two years in Fig. [7]. The structure of the peak to peak gradients seen by Nakazawa et el. (1991) is indicated in the figure. Because the boundary condition for  $CO_2$  is set at the surface in the model, concentrations at the tropopause are, in general, different from actual measurements at that altitude. Therefore, direct comparisons of the stratospheric  $CO_2$  signal with measurements are not always possible. However, correlations between  $CO_2$  and  $N_2O$  at midlatitudes, discussed in the next section, are not significantly affected by the ambiguity in specifying  $CO_2$  at the lower boundary of the stratosphere. This was verified by making a control run with  $CO_2$  specified at the surface independent of latitude. The concentration was taken as the average of measurements at Mouna Loa and Samoa.

The coarse vertical resolution of the model (2 km) is cause for concern about the accuracy of the numerical solution for the upward propagation of the seasonal cycle of  $CO_2$  through the lowermost tropical stratosphere. Age increases by several years within the first few kilometers above the tropical tropopause. A finite amount of vertical diffusion will, therefore, critically affect the attenuation of seasonally varying tracer signals with height. The propagation of time dependent tracer signals is discussed in detail in Kogan-LeFlore et al. (1998). It is shown in this paper that an adequate numerical solution is obtained in the model with the 2 km vertical resolution in conjunction with the profile specified for the vertical diffusion coefficients.

A time sequence for calculated  $CO_2$  concentrations is shown in Fig. [8]. The first panel shows concentrations for late spring in the Northern Hemisphere, corresponding to the seasonal maximum at the surface. About four months later, tropospheric values have decreased and the high concentrations in the lower tropical stratosphere can be seen to extend into mid latitudes at low levels. During the course of a year, the concentrations increase by about 1 ppm (the annual rate of increase) throughout the model domain. However, the increase is not uniform.

The seasonal behavior of  $CO_2$  and age is illustrated further in Fig. [9]. The figure shows time series for the two quantities in the tropics and for mid and high latitudes at altitudes of 17, 21, 31 and 41 km. At the lowest altitude in the tropics, the seasonal variation of  $CO_2$  shows no correlation with age and is determined mainly by variations imposed at the lower boundary. At higher altitudes in the tropics,  $CO_2$  increases more or less steadily and age remains constant. Large seasonal variations can be seen at 31 km at mid latitudes. Here, fluctuations of  $CO_2$  and age are anticorrelated and caused by seasonally varying rates of outflow of younger,  $CO_2$ enriched air from the tropics. Low levels at midlatitudes show a combination of seasonality induced by changing rates of tropical outflow and changes in the boundary condition imposed for  $CO_2$ . The seasonality at low altitudes is not symmetric with respect to the equator because the minima and maxima of  $CO_2$  are not centered at the equator in the model.

### 4. CORRELATIONS OF N<sub>2</sub>O AND CO<sub>2</sub>.

Some aspects of the transport of  $CO_2$  into the mid and high latitudes of the lower stratosphere can be checked against observations. Correlations of  $CO_2$  and  $N_2O$  have been measured at ER-2 flight altitudes (20 km and below) for different seasons (Boering et al. 1994).  $CO_2$  and  $N_2O$  are both long lived tracers ( $N_2O$  only for the lower stratosphere) with tropospheric sources. Concentrations of long lived tracers with similar source regions are proportional to each other in steady state (see the discussion in Plumb and Ko, 1992). However, the temporal variations of  $CO_2$  occur on a timescale that is much faster than the increase in  $N_2O$  at the surface and also smaller than the average age of stratospheric air. Therefore, the slope of the correlations changes with time and season and is sensitive to the rate of transport of  $CO_2$  through the subtropical barrier.

Calculated  $CO_2/N_2O$  correlations are compared with the SPADE observations in Figs. [10] and [11] for the low  $K_{yy}$  case. Correlations are shown for the Fall of 1992, the spring of 1993 and the Fall of 1993. The seasonal behaviour of the correlations in the Northern Hemisphere follows the observed changes in slopes fairly well. The appearance of the seasonal minimum of  $CO_2$  in Fall at low altitudes is evident in the data and captured by the model. The data also show that the annual increase of  $CO_2$  appears at low values of  $N_2O$ , or higher altitudes, between fall and spring (Fig. [10]). The correlations remain almost unchanged between spring and fall (Fig.

[11]). The seasonal variations of the correlations are a composite of the seasonal behavior of  $CO_2$ , shown in Fig. [9], and the seasonal changes in the isolines of  $N_2O$ .

Correlations of  $CO_2$  and  $N_2O$ , calculated with the model, are compared with the ASHOE/MEASA data in Figs. [12] and [13]. The figures show correlations for the Southern Hemisphere, tropics and Northern Hemisphere for spring (Fig. [12a]) and fall (Fig. [12b]) of 1994. Agreement with the Southern Hemisphere data is good for all seasons. In the Northern Hemisphere, concentrations calculated for  $CO_2$  are about 1 ppm higher than the observed values. This applies for all seasons of the SPADE and ASHOE/MAESA campaigns.

In order to examine the sensitivity of calculated  $CO_2/N_2O$  correlations to the choice of  $K_{yy}$ , runs were made using the large and constant  $K_{yy}$  distributions, discussed in Schneider et al. (1998). Fig. [13a] compares correlations calculated for the large  $K_{yy}$  case, i.e. coefficients multiplied by a factor two compared to the run discussed above, for the Fall of 1992 and the Spring of 1993 in the Northern Hemisphere. The change in slope is minor compared to the small  $K_{yy}$  case shown in Fig. [10]. It is not possible to call the agreement with observations better or worse for these cases, especially when we recall that important processes such as the Quasi Biennial Oscillation are not included in the model. Doubling  $K_{yy}$  has an effect on the circulation, both at midlatitudes and in the tropics by continuity. Upward motions in the tropics and the overturning rates in the lower stratosphere increase by approximately 20% on average. However, the change in the circulation pattern affects  $N_2O$  and  $CO_2$  similarly. Lines of constant mixing ratio for both species remain more or less parallel.

In order to examine the influence of seasonal variations in  $K_{yy}$ , results from the constant  $K_{yy}$  case are compared with observations in Fig. [13b]. The change in the slope of the correlations during winter is comparable to that in the previous cases. It follows that the seasonality induced by the time varying flow across the subtropics is the dominant contribution to the change of the slope of the correlation curves.

If  $K_{yy}$  is reduced drastically, the annual increase in CO<sub>2</sub> is not transported fast enough to mid and high latitudes in the lower stratosphere. The result for very small diffusion in the Southern hemisphere,  $K_{yy}=10^4 \text{m}^2 \text{sec}^{-1}$ , is shown in Fig. [14] together with the ASHOE/MAESA data for spring. These model results are clearly unacceptable. The value of  $K_{yy}$  needed to obtain agreement with the measurements is approximately  $10^5 \text{m}^2 \text{sec}^{-1}$ . Once diffusive transport in the winter hemisphere exceeds this threshold, correlations between CO<sub>2</sub> and N<sub>2</sub>O are relatively insensitive to  $K_{yy}$ . Frictional damping in the lower stratosphere is a sensitive parameter in the model. For the runs discussed so far, frictional damping time scales exceed a season in the altitude region between 10 and 30 km, reaching a maximum of 300 days at 20 km. We have not allowed for any latitudinal structure in Rayleigh friction anywhere in the model because of the lack of information about gravity wave spectra and gravity wave breaking in the atmosphere.

If friction is applied independent of latitude in the lower stratosphere, mass exchange between tropics and midlatitudes is drastically altered. Meridional coupling provided by friction induces a large horizontal flow component, resulting in an overestimate for  $CO_2$  at the mid and high latitudes of the lower stratosphere and in lower concentrations at higher altitudes.

#### 5. SUMMARY AND DISCUSSION.

Measurements of age and  $CO_2$  provide stringent constraints on the flow of air across the subtropical region at stratospheric altitudes. We have shown that a 2-D residual circulation model gives a good representation of the mean age of air in the lower stratosphere and can reproduce the slope and seasonal variations observed in correlations between  $CO_2$  and  $N_2O$  in the mid latitudes of the lower stratosphere.

In order to obtain agreement with observations, mechanical damping must be small in the altitude region between 10 and 30 km. As long as the time scale for frictional damping exceeds a season for most of this range, the effect of the friction on the meridional velocities does not impact calculated tracer distributions significantly. Given the rather coarse vertical resolution of the model, no claims are made that the vertical structure of the lower stratosphere is resolved and that the effects of gravity wave breaking have been treated in a physically meaningful way.

The model results are robust as far as assumptions for  $K_{yy}$  are concerned. A threshold value for  $K_{yy}$  in the lower stratosphere of approximately  $(0.5-1.0)*10^5 \text{m}^2 \text{sc}^{-1}$  has to be applied to ensure that sufficient quantities of young air are transported to mid and high latitudes after having been advected across the subtropical barrier. Once this threshold value is exceeded, the sensitivity of the results to the assumptions for  $K_{yy}$  is small. Within the constraints of currently available data, the eddy mixing coefficients can be varied by at least a factor of two. In addition, the temporal and spatial structure of  $K_{yy}$  does appear to play a determining role for the performance of the model. The relative insensitivity to the magnitude of the assumed Rossby wave diffusion coefficients can be understood by considering the dual role of  $K_{yy}$  in regulating the diffusive transport of tracers and in providing a forcing term for the residual mean circulation (Holton, 1986).

Changing the wave driving of the circulation by a factor of two results in changes of the order of half a year in the calculated mean age of air. Temperature changes are of the order of 1 to 2 K in the lower stratosphere. However, the rate of the stratospheric overturning changes by about 30%. Currently available data on age and long lived tracers cannot be used to reduce this uncertainty in vertical velocities at the 100 mb level. The uncertainty is of the same order as was found in attempts to diagnose the residual circulation by calculating heating rates from available data (e.g. Eluszkiewicz et al., 1996; Rosenloff et al., 1996, Appenzeller et al., 1996).

An inherent limitation of a 2-D model is that the tropospheric lower boundary cannot be treated in a self consistent manner. Vertical velocities and vertical diffusion at the tropopause level determine the flux of material from the troposphere to the stratosphere. The distribution of a tracer with constant surface concentrations and a sink in the stratosphere will eventually equilibrate and errors in the treatment of the tropopause will affect the concentration profile to some extent. Tracers whose surface concentrations change on a time scale shorter than the mean age of the stratosphere are much more sensitive to the representation of dynamics in the region near the tropopause.

In our model, parameterized heating rates are used to generate a distribution of temperatures and wind velocities in the troposphere. The parameterization of tropospheric heating rates and the transition to calculated stratospheric heating rates above the tropopause was initially adjusted to yield tropical tropopause temperatures close to observations. Since the parameterization consists of a relaxation to specified equilibrium temperatures and a zonal mean momentum dissipation to ensure finite heating rates (Cunnold et al., 1975), the tropospheric temperatures and the Hadley circulation are not fixed in the model. To some extent, they depend on stratospheric conditions. Increased Rossby wave diffusion, for example, causes more "suction" out of the tropics and, therefore, increased adiabatic cooling and reduced temperatures at the tropical tropopause.

We have made several runs in which the parameterization of tropospheric heating rates was modified, resulting in changes of temperatures at the tropopause level and corresponding changes in vertical velocities and meridional outflow at the bottom of the tropical pipe. Large modifications that induced temperature changes at the tropical tropopause by 4 to 5 K significantly altered the rate at which the annual increase of CO<sub>2</sub> propagated into the stratosphere. Resulting CO<sub>2</sub>/N<sub>2</sub>O correlations were clearly ruled out by the observations. Circulations obtained with smaller modifications resulting in a change of less than 2 K at the equatorial temperature minimum did not affect computed correlation curves significantly. Tropospheric

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parameters clearly play an important role in constraining the exchange of mass between troposphere and stratosphere in a 2-D model. For the particular form of the parameterization used in the present model, results for tracer correlations in the lower stratosphere appear to be robust under small parameter variations.

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#### FIGURE CAPTIONS.

- Fig. [1]: Calculated distributions of mean age for October (a) and January (b). Units are years.
- Fig. [2]: Latitudinal distribution of mean age at 20 km for 4 seasons. The diamonds represent ages determined from measurements of  $SF_6$  (Geller et al., 1996).
- Fig. [3]: Calculated mean age for January for the low  $K_{yy}$  case (solid lines) and the large  $K_{yy}$  case (dotted lines).
- Fig. [4]: As Fig. [2], but for the large  $K_{yy}$  case.
- Fig. [5]: (a) Calculated vertical velocity at 100 mb for the low K<sub>yy</sub> case (solid line) and the large K<sub>yy</sub> case (dotted line). (b) Percent difference between vertical velocities, shown in (5a) for Northern mid and high latitudes.
- Fig. [6]: Calculated correlations between  $N_2O$  and mean age. Points have been sampled between the altitudes of 10 km and 40 km. Plus signs are points obtained for the latitude region between  $-10^\circ$  and  $10^\circ$ , asterisks are for  $10^\circ$  to  $20^\circ$ , diamonds for  $20^\circ$  to  $40^\circ$  and triangles for  $40^\circ$  to  $60^\circ$ .
- Fig. [7]: Peak to peak differences in CO<sub>2</sub> concentrations during the year 1993. The solid line represents values at the surface, the dotted line is the seasonal amplitude at 300 mb. Measurements by Nakazawa et al., 1991) are indicated by asterisks. Units are ppm.
- Fig. [8]: Calculated concentrations of CO<sub>2</sub> for the year 1993. Panels show the distribution every two months, starting from late spring. The Julian day is indicated in the panels. Units are ppm.
- Fig. [9]: 12 months time series, starting in late spring, of  $CO_2$  and age for points in the model domain as indicated on the panels.
- Fig. [10]: Correlations between  $CO_2$  and  $N_2O$ . Shaded areas indicate data gathered during the SPADE campaign. The dark shading are measurements from the Fall of 1992, the lighter shading are data from the Spring of 1993. Model results for the Fall of 1992 are indicated by open circles. Triangles are model results for the Spring of 1993.
- Fig. [11]: As in Fig. 10 but for the Fall of 1993. Dark shaded areas indicates the measurements, model results are shown by open circles. The data for the Spring of 1993 (light shading) are shown for reference.
- Fig. [12]: (a) Correlations between CO<sub>2</sub> and N<sub>2</sub>O for the Southern hemisphere in the Spring of 1994. Shaded areas are data from the ASHOE/MAESA campaign, model results are represented by the open circles. (b) as in (a) but for the Fall of 1994.

- Fig. [13]: (a) Correlations between CO<sub>2</sub> and N<sub>2</sub>O for the Northern Hemisphere midlatitudes for the Fall of 1992 and the Spring of 1993, calculated for the large K<sub>yy</sub> case. Averages of the SPADE data, shown in Fig. 10 are indicated by the solid line for the Fall of 1992 and the dotted line for the Spring of 1993. (b) as in (a), but model results from the constant K<sub>yy</sub> case.
- Fig. [14]: (a) Correlations between CO<sub>2</sub> and N<sub>2</sub>O obtained for extremely small K<sub>yy</sub> in the Southern hemisphere for the Spring of 1994 (squares). The average of the data, shown in Fig. [12a] is indicated by the solid line.

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Figure 1



Figure 2



Figure 3



Figure 4



Figure 5a



Figure 5b



Figure 6



Figure 7



Figure 8



Figure 9



Figure 10



Figure 11



Figure 12a



Figure 12b



Figure 13a



Figure 13b



Figure 14