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Simulations of the Trend and Annual Cycle in Stratospheric CO_2

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The distribution and evolution of stratospheric CO_2 in response to the observed annual cycle, interannual variations, and long-term trends in tropospheric CO_2 is simulated with the GISS 23 layer stratospheric general circulation model. Carbon dioxide is a tracer of stratospheric transport which has essentially no local sources or sinks but still displays gradients due to the forcing at the surface. Consequently, observations of stratospheric CO_2, until recently limited to a few flask samples, but now included as high frequency in situ sampling in aircraft campaigns, provide a test of tracer transport in stratospheric simulations independent of model chemistry. In our model, CO_2 enters the stratosphere primarily through the tropical tropopause, where air parcels are effectively labeled in time by their CO_2 values (although not uniquely because of the cycles in tropospheric concentration). Parcels of differing ages are subsequently mixed in the stratosphere. Only when the growth is purely linear can the CO_2 offset in a parcel relative to the troposphere be interpreted as the average time since stratospheric air was last in contact with the troposphere, i.e., the "age" of the stratosphere. Our model is in qualitative agreement with multiyear averages of balloon soundings at northern mid- and high latitudes; the stratosphere at 30 km at mid-latitudes is about 4 years (6 ppm of CO_2) behind the troposphere. We predict significant propagation of the CO_2 annual cycle into the lower stratosphere, an effect which must be accounted for when interpreting observations. While the annual cycle is negligible above the lower stratosphere, interannual oscillations, such as those associated with El Ninos, can propagate well into the middle stratosphere as positive offsets from the linear trend lasting significantly longer than their duration in the troposphere.

INTRODUCTION

Stratospheric circulation controls photochemical losses for many chemically and radiatively important trace gases. For example, the atmospheric residence time of N_2O and the chlorofluorocarbons (CFC's) are limited by the rate at which air motions transport these tracers to the middle and upper stratosphere where they are destroyed. Carbon dioxide provides a unique measure of this stratospheric circulation. It is nonreactive in the stratosphere and thus independent of the stratospheric chemical model. Its concentration in the stratosphere is increasing at a regular rate due mainly to the combustion of fossil fuels [*Keeling et al.*, 1976] and, thus, provides a chronometer for the age of air entering the stratosphere [*Bischof et al.*, 1985; *Schmidt and Khedim*, 1991].

Mid-latitude balloon soundings of CO_2 show decreases from the tropopause to the middle stratosphere that range from 3 to 10 ppm by volume [*Bischof et al.*, 1985; *Schmidt and Khedim*, 1991]. Similarly, aircraft sampling in the lower polar stratosphere during the winter reports CO_2 abundances several ppm less than concurrent tropospheric values [*Heidt et al.*, 1989]. These observations are used here as a fundemental test of tracer transport in the general circulation model of the Goddard Institute for Space Studies (GISS). We use the middle atmospheric version of the GISS GCM (global climate/middle atmosphere model [*Rind et al.*, 1990]) to predict the propagation into the stratosphere of the annual increase and annual cycle of CO_2. The model's simulation quantitatively reproduces some of the large scale features observed for CO_2, thereby providing an independent verification of the tracer transport used in stratospheric chemical modelling. This global three-dimensional simulation of stratospheric CO_2 provides a framework for comparing observations made at different latitudes and during different phases of the annual cycle and for planning new measurements.

The GISS middle atmosphere and chemical transport model is described in section 2. In section 3, we document the specifics and approximations that define the CO_2 calculations. We present results in section 4 for a linearly increasing tropospheric concentration, for annual cycles and for transient pulses. We conclude in section 5, evaluating the role of current and future measurements of CO_2 in understanding stratospheric circulation.

GENERAL CIRCULATION MODEL

Rind et al. [1990] have analyzed the GISS 23-layer stratospheric GCM, and a single year of modeled winds has been archived for use in the Chemical Transport Model (CTM). *Prather et al.* [1987] documented the tropopsheric CTM. The stratospheric version has been applied to meteoric infall [*Prather and J. M. Rodreguez*, 1988], ozone [*Prather et al.*, 1990*b*], and space-shuttle chlorine [*Prather et al.*, 1990*a*]. The CTM adopts the 7.83° latitude by 10° longitude grid of the parent GCM, and uses the lowest 21 layers. The tropospheric layers, 1 to 9, use sigma coordinates, while the stratospheric layers, 10 to 21, lie between fixed pressure levels beginning above 100 mbar.

The CTM solves the tracer continuity equation by operator splitting with separate calculations of advection, convection, diffusion, and chemical sources and sinks. The single year of circulation statistics from the GCM is recycled for

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multiyear runs, and thus we do not simulate interannual variability associated with changes in circulation. Multiyear sequences of GCM runs [*Rind et al.*, 1990] may be employed in future studies. Advection by the 8-hour average wind fields is computed using the second-order upstream algorithm [*Prather*, 1986] which automatically calculates and stores a quadratic representation of the tracer distribution within each grid box. Vertical convection and horizontal diffusion, based on monthly convection statistics from the GCM, apply only in the troposphere; no diffusion of tracer is included within the stratosphere.

The vertical resolution of the model between 100 mbar and 1 mbar is course, about 5 km per level. We compensate in part for this poor resolution by utilizing the computed second-order moments of the tracer distribution to diagnose concentrations every 2 km in pressure altitude. The diagnostics reported here are taken from 5-day instantaneous samples at this vertical resolution. In the stratosphere, large-scale planetary wave modulation of tracer flux and concentration are the only zonal features, and hence we primarily report here zonally averaged values.

SIMULATION OF CO_2 IN THE STRATOSPHERE

CO_2 Chemistry

The bulk of air entering the stratosphere is believed to carry the chemical signature of the upper tropical troposphere [*Brewer*, 1949; *Holton*, 1990]. Once in the stratosphere, we expect little chemical evolution of CO_2 within a parcel. Carbon monoxide is rapidly oxidized to CO_2 in the lower tropical stratosphere [*Murphy et al.*, 1993] but the initial tropospheric abundance is small relative to CO_2 variations, and will contribute at most 0.1 ppm. Methane, with a concentration of 1.8 ppm near the tropical tropopause, is the dominant hydrocarbon entering the stratosphere, and its oxidation represents a small stratospheric source of CO_2. Methane contributions fall-off gradually in the stratosphere implying contributions to CO_2 abundances away from the tropical tropopause, with largest contributions, about 1 ppm, at the tropical stratopause and in the lower polar winter stratosphere. In the upper mesosphere, above 80 km, CO_2 is photodissociated into CO and O. However, this process is not an irreversible sink for CO_2 because CO is chemically recycled back to CO_2 before the air returns to the stratosphere [*Allen et al.*, 1981]. In the stratosphere, condensation provides a mechanism for the separation and selective removal of species from the gas phase. Condensation, followed by gravitational settling, is believed to be an important process for removing water vapor near the tropical tropopause and removal of water and nitrates from the polar winter stratosphere. Nevertheless, we know of no process incorporating CO_2 into the falling ice crystals.

The calculations presented here have assumed CO_2 is conserved throughout the stratosphere. In later discussion we estimate the relatively small impact of including the CH_4 sources of CO_2.

Tropospheric CO_2 as a Lower Boundary Condition

The biogeochemical cycling of CO_2 is driven by sources and sinks at the surface of the Earth. Studies of tropospheric CO_2 require a documented tropospheric CTM and a detailed discription of surface sources and sinks [*Fung et al.*, 1987; *Heimann et al.*, 1989; *Keeling et al.*, 1989*b*; *Tans et al.*, 1990]. The stratospheric CTM used in these simulations has not been documented for tropospheric tracers as has the nine-layer GISS CTM [*Prather et al.*, 1987; *Jacob and Prather*, 1990; *Fung et al.*, 1987; *Spivakovsky et al.*, 1990]. We believe, however, a complete model of tropospheric CO_2 is unnecessary. Our need for this study is CO_2 at the tropopause as a time-dependent boundary condition. Such observations are obviously not available, while alternatives, such as the use of other model simulations, are problematic.

Our approach is to use the NOAA/GMCC analysis of their CO_2 observations [*Conway et al.*, 1988] as a lower boundary condition on mixing ratios in the CTM. These clean-air, mostly maritime measurements are used to define CO_2 concentrations as functions of latitude and time (biweekly). In our model these concentrations are applied uniformly around each latitude circle and updated monthly. The approximation of zonal symmetry is clearly unrealistic because of the large longitudinal (i.e., continent versus ocean) gradients in sources and sinks. Nevertheless, we expect that the response of the upper troposphere to this surface boundary condition will provide an adequate forcing for the stratospheric simulation. An average annual cycle for surface CO_2 concentrations was derived from 5 years of NOAA/GMCC data [*Conway et al.*, 1988] by removing a linear trend and averaging the five years at monthly resolution. (We use the term "annual cycle" to refer to variation of one year period resulting from the seasonality of plant growth.) As a further simplification, we adopt a fixed, linear rate of CO_2 increase equal to 1.5 ppm/yr [*Keeling et al.*, 1989*a*].

A brief evaluation of the CTM simulation of the upper troposphere in response to the surface forcing of the annual cycle shows reasonable agreement with previous studies and recent observations. Published studies of tropospheric CO_2 [*Fung et al.*, 1987; *Heimann et al.*, 1989; *Keeling et al.*, 1989*b*; *Tans et al.*, 1990], employing spatially resolved and seasonally varying sources and sinks with three-dimensional models, show a clear attenuation with altitude of the land-ocean zonal asymmetries. From such studies, we expect zonal variations in mean concentration and cycle amplitude of only about 1.0 ppm at 200 mbar [*Heimann et al.*, 1989; *Keeling et al.*, 1989*b*]. Figure 1 shows the peak-to-peak amplitude of the annual cycle as a function of latitude at the surface (the boundary condition), at 10 km in our model, and in the upper troposphere from the observations reported by *Nakazawa et al.* [1991]. In the northern hemisphere, our CTM simulation of the upper troposphere appears adequate. In the southern hemisphere, we see the extension of the northern cycle into the tropical and mid-latitude upper troposphere, although the calculated amplitude is smaller than observed. These patterns in the southern hemisphere are similar to those presented in the more detailed, tropospheric modelling study of *Fung et al.* [1987]. We conclude that the zonal mean forcing is adequate for stratospheric studies.

In this study, we perform separate simulations of the atmospheric response to the linear growth and the annual cycle of CO_2, in both cases recycling one year of GCM winds. The model is initialized with a surface concentration of 336 ppm (circa 1980), and forced at monthly increments with the trend and the annual cycle in the lowest

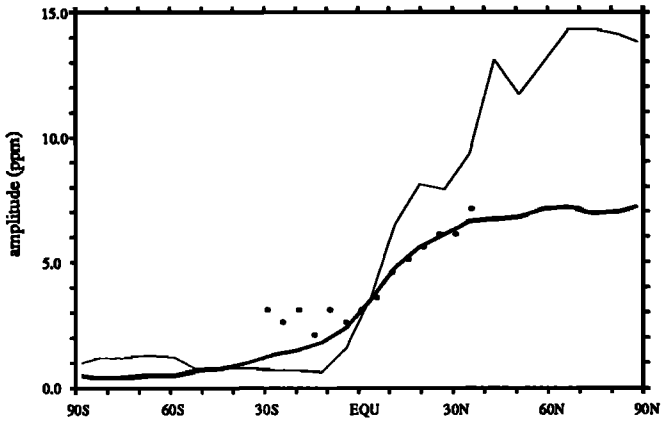

Fig. 1. Peak-to-peak amplitude of the annual cycle in CO_2 concentration at the surface (light line) and at 10 km (heavy line) as a function of latitude. The surface values are assumed, and the response in the upper troposphere is calculated. Points are from the observations of *Nakazawa et al.* [1991]

three levels (approximately the first kilometer of the model atmosphere). We integrate for 10 model years, achieving an approximate steady state in the annual trend and cycle after 5 to 6 years. Tropospheric observations of CO_2, however, show measureable year-to-year variations in the trend and annual cycle associated with the El Nino/Southern Oscillation (both oceanic souce/sink and circulation), with changes in fossil fuel use, and with long-term shifts in biospheric sources and sinks [*Bacastow*, 1976; *Bacastow et al.*, 1985; *Keeling et al.*, 1989*a*]. We might expect stratospheric CO_2 to vary from year to year due to these tropospheric fluctuations in addition to changes in the stratospheric circulation. Our simulation neglects all such interannual variations, but we estimate the effects of irregular tropospheric forcing (with fixed circulation) by examining a third calculation showing the transient response to a single pulse of CO_2.

Results and Discussion

We present results for several stratospheric simulations. Our goal is to access separately the importance to the stratosphere of steady growth, annual cycle, and irregular variations in tropospheric CO_2. This is possible because the calculations defined here are completely linear in CO_2 concentration. We thus simulate the response for these individual forcings separately, considering them in combination afterwards.

Steady Growth

A steady increase of CO_2 at the surface will drive gradients, primarily vertical, throughout most of the atmosphere. The magnitude of these gradients will vary depending on the efficiency of the atmospheric circulation in mixing the tracer. If we force the atmosphere with a regular increase of 1.5 ppm/yr and allow a steady state to be reached (after 6 years), then the CO_2 concentration at each location in the atmosphere is exactly 1.5 ppm greater than its value one year before, a consequence of providing no interannual variation in circulation. Seasonal variations in circulation, however, preclude simple linear growth during the year.

The annually averaged, zonal mean concentration of CO_2, plotted in Figure 2 as a (negative) offset from the tropical tropopause, shows the essential features of how tracers are mixed in the stratosphere. The shape of the CO_2 surfaces at constant mixing ratio (isopleths) is similar to that of other long-lived tracers such as N_2O and CH_4 [*Jones and Pyle*, 1984]. Gradients of N_2O and CH_4 are driven by photochemical loss which becomes more rapid at high altitudes, whereas the structure of CO_2 is determined by the 1.5 ppm/yr increase in air entering the stratosphere. This common stratospheric tracer structure may be explained qualitatively by a Brewer-Dobson cell [*Brewer*, 1949] combined with rapid latitudinal mixing [*Mahlman*, 1985; *Holton*, 1986]. Ascent occurs in the tropics; poleward motion, in mid-latitudes; and descent at high latitudes. As a result, a CO_2 isopleth lies slightly above a given potential temperature surface at low latitudes and below the same surface at high latitudes. Within the winter polar vortex, descent is strong and exchange with mid-latitudes is inhibited, and thus the tracer isopleths descend through a large range of potential temperature. This polar feature is evident in our model in the middle stratosphere (although somewhat washed out by the annual average), but is significantly underestimated below 20 km.

Schmidt and Khedim [1991] have analyzed balloon soundings of CO_2 at mid- and high latitudes which they made from 1982 to 1990. For our purposes, from their original data we form a composite profile as a function of pressure altitude by adjusting CO_2 concentrations to the year 1989 using an assumed trend of 1.5 ppm/yr. Figure 3 displays these profiles and their variability (± 1 standard deviation) versus pressure altitude at 44°N and 68°N. Samples from the three mid-latitude soundings in 1989 and 1990 are systematically more than a standard deviation above the composite. We apply a single additional offset to all three bringing them near the composite; this additional offset reduces the composite standard deviation by about 30%, but does not significantly affect the profile shape.

To compare with the composite profile of balloon soundings, we calculate a weighted average by sampling model profiles at 0°W according to the dates of the balloon launches and shift the absolute value of each profile, employing the 1.5 ppm/yr trend, to match the observed 1989

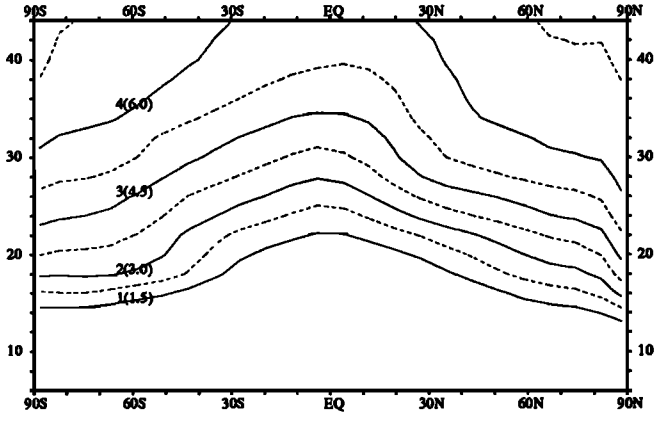

Fig. 2. Annually averaged, zonal-mean concentration of CO_2 as a function of latitude and pressure altitude in response to a forced linear growth rate of 1.5 ppm per year. Contours show the concentration as a negative offset from the tropical tropopause. Contours are labelled as ppm and in "years" (1.5 ppm = 1 year). In this paper pressure altitude is defined as $z = 16 \log (1000/p)$, with z in km and p in mbar.

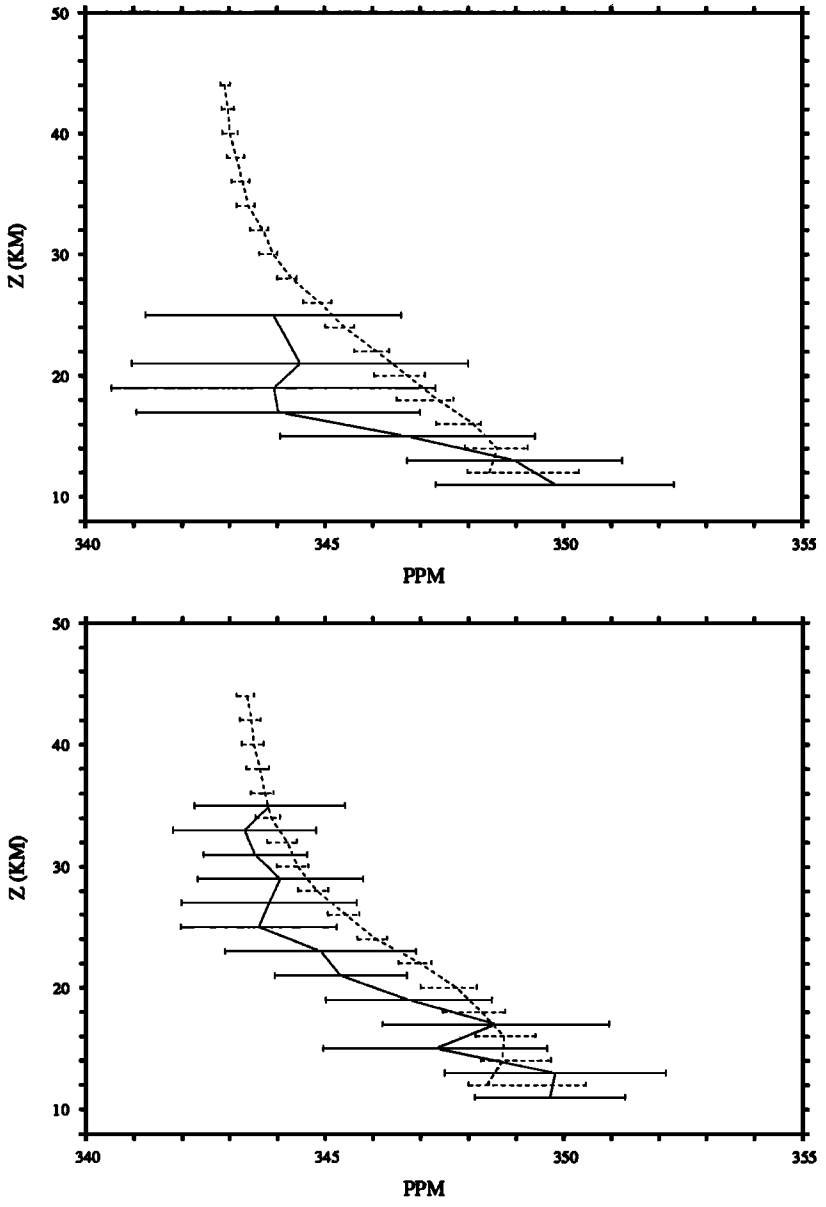

Fig. 3. CO_2 concentration (ppm) versus pressure altitude at 44°N and 68°N. The solid lines are composite profiles from a series of balloon soundings [*Schmidt and Khedim*, 1991] and the dashed lines are model profiles whose averaging has been weighted to match the balloon launch dates. For both observed and modelled profiles, the variation shown is ± 1 standard deviation.

global mean value. These model profiles are shown at 44°N and 68°N in Figure 3, along with their standard deviations (computed with respect to a mean not weighted by the balloon launch dates). The choice of longitude is arbitrary because of the zonally uniform forcing. We plot model profiles at a particular longitude rather than zonal means so that propagating zonal features will be reflected in the standard deviations. We may then more meaningfully compare model and observed variability.

Gross features of the model and observations are in agreement. Both show a rapid fall-off of CO_2 with height in the lower stratosphere and a zone of weaker vertical gradient in the middle and upper stratosphere. This zone is lower at 68°N than 44°N in both model and observation. The appearance of a region of weak vertical CO_2 gradient can be understood in terms of the time scale for effective vertical transport: when the time scale for transport over a scale height is much shorter than a year, the corresponding CO_2 gradient will be much less than 1.5 ppm. On the other hand, no such region is observed for N_2O below the stratopause [*Jones and Pyle*, 1984; *Schmidt et al.*, 1991], presumably because the time scale for photodissociation decreases rapidly with altitude, becoming less than 2 months at 45 km, and thus remains shorter than the vertical transport time scale. The total fall-off in CO_2 concentration from the upper troposphere to the upper stratosphere is similar in both model and observation, about 6 ppm. The obvious discrepancy between model and measurements in Figure 3 could be resolved if the model CO_2 profile were displaced down by about 5 km. Some of this error is caused by a model tropopause that is too high, as has been noted in O_3 studies [*Prather et al.*, 1990*b*] and $C^{14}O_2$ studies [*Remsberg and Prather*, 1992]. Additional problems in this simulation may be due to the model's poor vertical resolution in the

lower stratosphere; only three levels lie between 200 mbar and 22 mbar (11 km and 27 km).

In addition to a detailed disagreement in shape, the model profiles show much less variability in the middle stratosphere than the observations. In the lower stratosphere the contribution to the model variability from the annual cycle in tropospheric forcing is large, while above 24 km it is negligible. The model variability in the middle stratosphere, the order of ±0.4 ppm, is due mostly to the effects of propagating planetary waves but partly to annual cycles in meridional circulation. The standard deviations of the observations are 5 to 8 times larger than the model, suggesting a dominant role for interannual variability which not simulated here.

It is tempting to use the lag in CO_2, defined as the instantaneous offset between the stratospheric concentration and that at the tropical tropopause, to specify the time elapsed since stratospheric air was last in contact with the troposphere. For example, the 6.0 ppm offset at the tropical stratopause (see Figure 2) could be interpreted as an age of 4 years, given a growth of 1.5 ppm/yr. However, a single parcel of this stratospheric air cannot be equated to a single parcel of tropospheric air that entered the stratosphere 4 years ago; rather it is an ensemble of air of many different ages which have been irreversibly mixed and whose average CO_2 offset is 6.0 ppm. The distribution of ages in this ensemble, which is determined by the details of the stratospheric transport, is identical to the distribution of CO_2 values only if tropospheric CO_2 varies linearly. Therefore, only to the extent that tropospheric CO_2 varies linearly can the average age of stratospheric air be inferred from CO_2 measurements. Similarly, this inferred age may only be used to predict the concentration of another rapidly growing trace gas without stratospheric source or sink if the growth of that gas is also linear.

The Annual Cycle

Forcing an annual cycle of CO_2 at the earth's surface generates a corresponding cycle in the upper tropical troposphere which then propagates into the stratosphere. The surface amplitude of the cycle is much larger than the annual CO_2 increase, and thus the distribution of CO_2 in the lower stratosphere may be dominated by annual oscillations rather than the steady growth. In addition, the cycle may obscure the determination of mean ages of stratospheric air based on measurements of CO_2.

In the upper tropical troposphere, we predict an annual cycle with a peak amplitude mear 12°N and with a phase similar to that of the large amplitudes in the lower troposphere at northern mid-latitudes. The amplitude (defined hereafter as peak-to-peak) of the CO_2 cycle is strongly attenuated with altitude as shown in Figure 4 . In the tropical troposphere at 16 km the amplitude decreases from 4.1 ppm at 12°N to 2.3 ppm at 12°S. This north-south difference diminishes into the stratosphere. The amplitude in the tropics falls to less than 1.5 ppm above 20 km and less than 0.5 ppm above 25 km. At extratropical northern latitudes there is a strong drop in amplitude between the troposphere and the stratosphere. Stratospheric amplitudes become progressively smaller away from the tropics and diminish with altitude. Corresponding latitudes in both hemispheres have similar amplitudes above 20 km, but in the lower stratosphere are greater in the north.

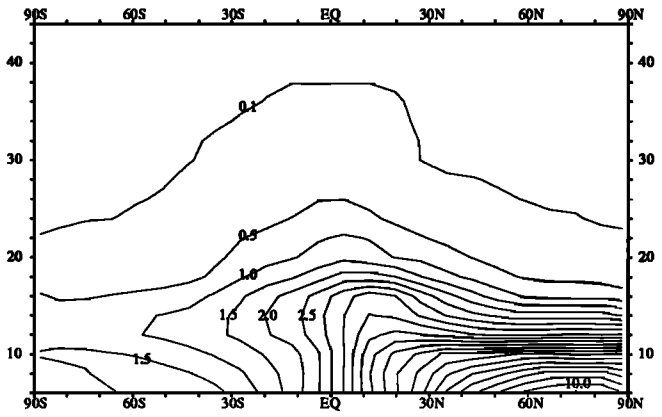

Fig. 4. Peak-to-peak amplitude of the annual cycle in CO_2 (ppm) contoured as a function of latitude and pressure altitude. The calculated amplitude results from the forcing of the observed annual cycle at the surface.

Figure 5 shows the phase of the CO_2 cycle versus latitude and month at the surface (the assumed forcing), at 16 km (emphasizing the upper tropical troposphere), and at stratospheric levels of 20 and 24 km. As the signal propagates vertically into the stratosphere, the phase increases steadily, shifting approximately 150° between the tropical tropopause (maximum in June) and 24 km (maximum in December). Similarly, at 24 km the phase of maximum shifts an additional 90° from equator to polar latitudes (maximum in March). The amplitude attenuates rapidly away from the tropics. At certain altitudes and phases the cycles are asymmetric between northern and southern mid-latitudes. A major cause of these differences appears to be the more vigorous latitudinal mixing during the winter season, but may also include the latitudinal asymmetry in the forcing at the tropics.

One might expect that an annual cycle forced at the surface with a mean value of zero would produce cycles in CO_2 concentration throughout the atmosphere with zero means. There may, however, be some correlations between the phase of the annual cycle and the annual variations in the circulation, causing systematic offsets in the annually averaged concentration. Indeed, the tropospheric circulation results in a systematic offset of about -0.15 ppm CO_2 in the upper tropical troposphere (compared to an annual range of 3 ppm at 16 km at the equator). Of primary concern here, if annual fluctuation of CO_2 in the upper tropical troposphere are correlated with the flux of air crossing the tropopause, then the annually averaged contribution to stratospheric CO_2 will be additionally offset from the upper tropospheric average. In this model, we see no evidence of significant annual correlations with air entering the stratosphere: net offsets in the stratosphere range from -0.13 ppm to -0.17 ppm.

If not accounted for, the annual cycle can cause significant errors in the inferred age of stratospheric air derived from CO_2 measurements. Comparing Figure 2 and Figure 4, we see the isopleths in the stratosphere of constant annual cycle amplitude look similar to those for a mean offset. The annual amplitude decays with distance from the tropopause at the same time that the net offset increases. Therefore, the potential error, defined here as the ratio of annual amplitude to mean offset, decreases from about 100% at a mean offset of 1 ppm to about 25% at 2 ppm offset to less than 5% for

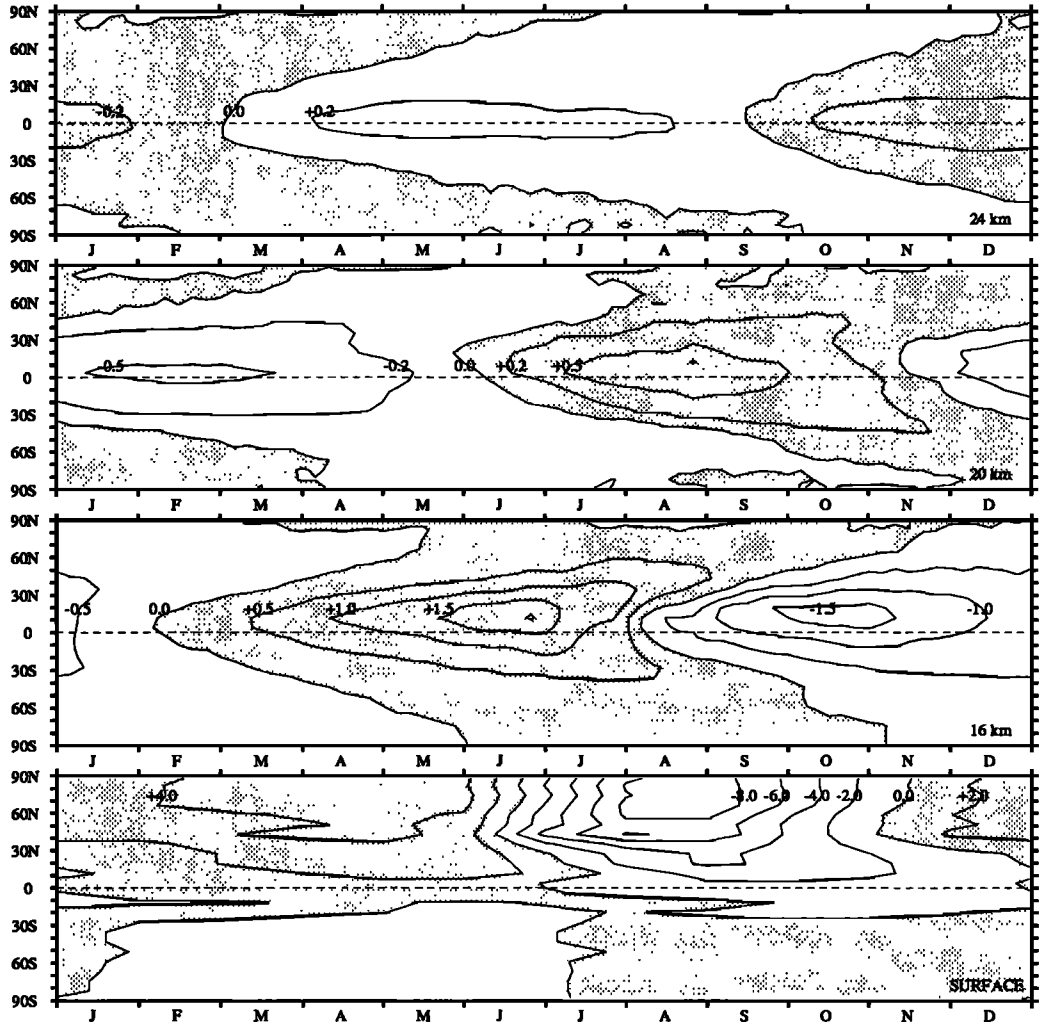

Fig. 5. Predicted annual cycle in CO_2 at pressure altitudes of (a) 24 km, (b) 20 km, (c) 16 km, and (d) the surface. The contour levels are labelled in ppm and indicate deviations from the annual mean. The shaded areas are positive CO_2 anomalies.

mean offsets greater than 4 ppm. This simple relationship may underestimate the potential error at high northern latitudes. Time series of the predicted CO_2 concentrations for four locations typical of ER-2 flights in the northern hemisphere are shown in Figure 6 . The induced annual cycle is not sinusoidal, and the phase clearly shifts between 16 km and 20 km. The amplitude is largest at 35°N (e.g., the Stratospheric Photochemistry, Aerosols and Dynamics Expidition (SPADE) mission), but still significant at 65°N (e.g., the Airborne Arctic Stratospheric Expedition (AASE) mission).

The modeled annual cycle in the middle stratosphere (40 km) lags the tropical tropopause by about 1.5 years (see Figure 7), a result which at first appears to contradict the 4 year mean age of the stratosphere deduced from the constant growth simulation. The constant phase trajectories in Figure 7 (represented by crosses and circles) connecting the tropopause to the upper stratosphere, however, cannot be interpreted simply as the advective transport (i.e., piped flow) of the complete annual cycle of CO_2 into the stratosphere. Rather, these phase lines are just the front of such a wave. As discussed above, the air at 40 km does not have a single age since it left the troposphere; it is a mix of air with different ages. The propagation of the annual cycle follows the fraction of air that reaches 40 km first. That this is a small fraction of the air is clear by the small amplitude, less than 0.1 ppm. The remainder of the air at 40 km averages over several years, and the annual cycles cancel. This explanation is supported by the simulation of a single pulse described below.

Irregular Variations

Irregular variations in the surface forcing of CO_2, such as those associated with the El Nino/Southern Oscillation (ENSO) [*Bacastow*, 1976], will have repercussions in the stratosphere. We examine how a pulse of CO_2 would propagate into the stratosphere. The model is initialized with a uniform value of CO_2 north of 32°N in the lowest kilometer of the atmosphere; there are no subsequent sources or sinks. We choose the initial value so that the atmosphere ultimately reaches a uniform 1.0 ppm.

The CO_2 signal appears within the first month in the upper troposphere at northern mid-latitudes, reaches a maximum value of 1.1 ppm at 6 months, and then declines as CO_2 is distributed throughout the rest of the northern hemisphere (troposphere) and into the south. After one year, the troposphere is fairly well mixed, the level in the mid-latitude

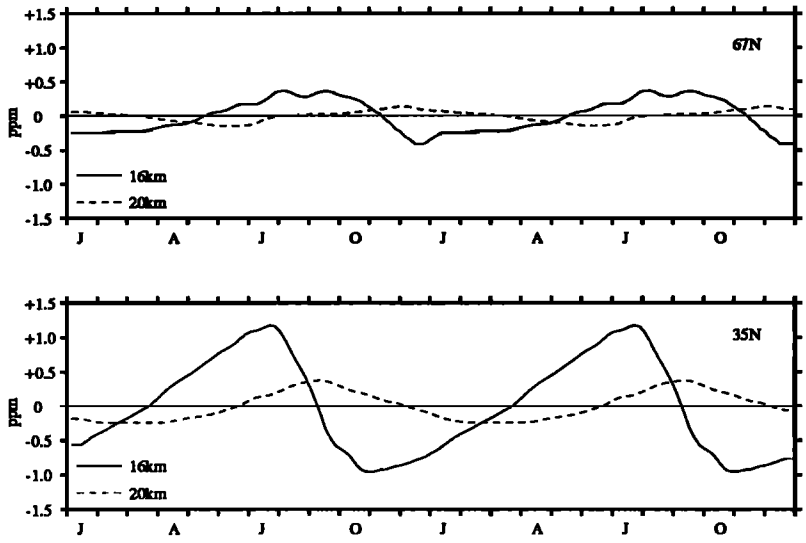

Fig. 6. Time series of predicted zonal mean CO_2 concentrations (ppm) at 35°N and 67°N for both 16 km and 20 km. Calculations include only the variations driven by the annual cycle at the surface.

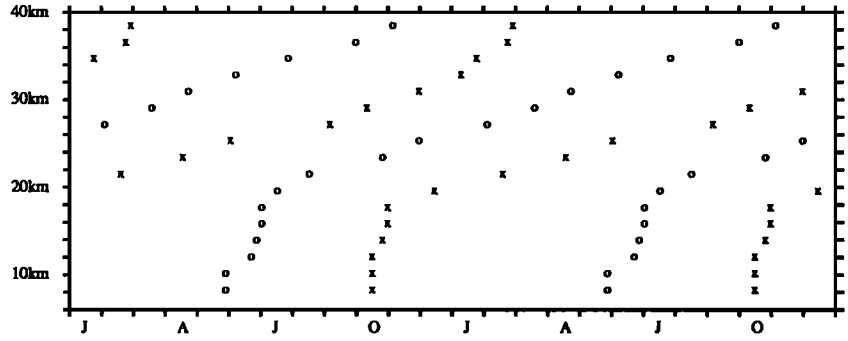

Fig. 7. Phase of the maximum (circles) and minimum (crosses) of the annual cycle in CO_2 concentration at 4°N as a function of time and pressure altitude in response to a forced cycle at the surface.

lower stratosphere (20 km) has reached about half its final value, but essentially there is no response yet in the tropical or mid-latitude middle stratosphere (30 km). Comparison of the 20 km time series at 4°N and 44°N (not shown) reveals a time scale for latitudinal transport in the lower stratosphere of about 6 months. The mixing ratio at 4°N and 30 km reaches 0.5 ppm in just under 2 years and 0.8 ppm after about 3 years.

Irregular variations in CO_2 associated with the ENSO suggest that a source of CO_2 is followed one to two years later by an approximately equivalent sink (for example, see *Keeling et al.* [1989*a*]). Because we are interested in perturbations about the linear growth, we combine this pulsed source with an equal but opposite pulsed sink one year later. This is done by subtracting each time series of the pulsed-source simulation from itself lagged by one year. This experiment simulates a perturbation about the linear growth, but without a shift in the long-term trend. It represents effectively a single cycle of a two year oscillation in CO_2 emmisions. Time series at selected locations are shown in Figure 8 .

With cancelling plus/minus pulses in CO_2 a year apart, the troposphere recovers rapidly: no signal is apparent at the 5% level after the second year (see the 12 km series in Figure 8). The stratosphere, however, carries a memory of the pulses for several years. At 20 km in the tropics, the response to the troposphere is more rapid, and the net signal (0.9 ppm after one year) falls to less than 0.1 ppm after 2.5 years. At 20 km in mid-latitudes, however, the CO_2 remains elevated by more than 0.1 ppm into the fourth year. At 30 km, the impact of the initial pulse is not detectable until the beginning of the second year, but a positive anomaly remains apparent at the 0.1 ppm (10%) level after 5.5 years. Thus, irregular perturbations about the linear growth in CO_2 will be manifest in the stratosphere, and a long-term net change in stratospheric CO_2 can occur.

The Effect of Methane Oxidation

Although CO_2 is approximately conserved in the stratosphere, CH_4 oxidation provides a small but detectable source. We have made a first-order estimate of the CH_4 oxidation based on our model's simulation of N_2O (T. M. Hall and M. J. Prather, manuscript in preparation, 1993) and the observed correlation of N_2O and CH_4 from the balloon profiles of *Schmidt et al.* [1991]. Figure 9 shows the annual mean concentration of CO_2 as a function of latitude and pressure altitude corrected for the CH_4 source. Compared with the parallel Figure 2, one can see the systematic shift in CO_2 isopleths, corresponding to about 0.75 ppm offset in

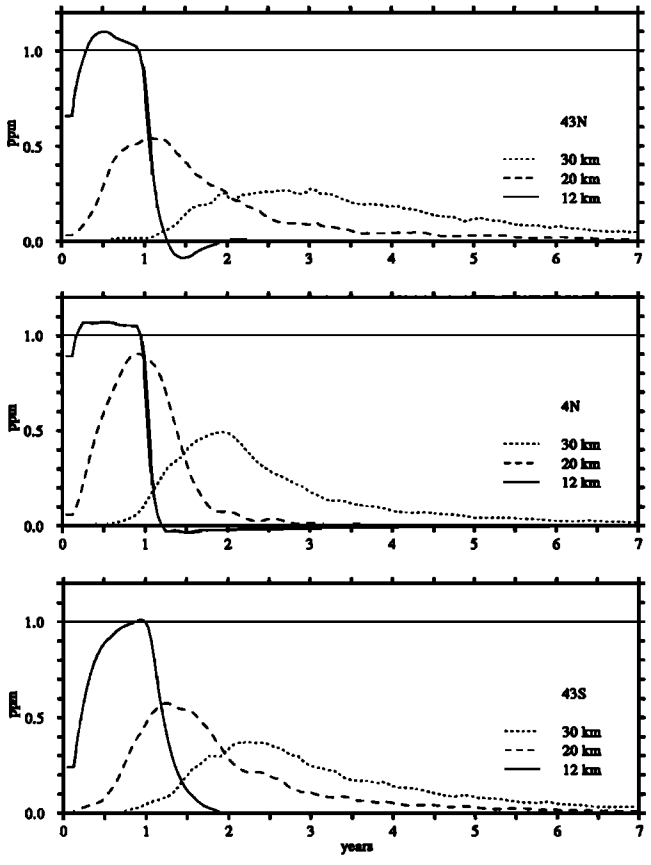

Fig. 8. Time series of predicted zonal mean CO_2 concentrations resulting from a pulsed source on January 1 and an equal sink 12 months later. Results are shown for altitudes of 12 km, 20 km, and 30 km at latitudes of 43°S, 4°N, and 43°N. The individual pulses are equivalent to a global increase/decrease of 1 ppm.

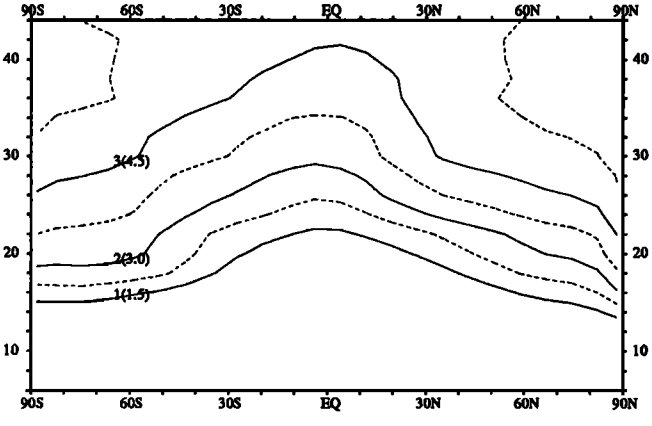

Fig. 9. CO_2 corrected for CH_4 oxidation as a function of latitude and pressure altitude. See details in Figure 2. The CH_4 source of CO_2 is based on the N_2O distribution calculated with the same model and an empirical N_2O-CH_4 correlation from balloon observations [*Schmidt et al.*, 1991].

the middle stratosphere. The methane source of CO_2, which increases with height, contributes to the appearance of the region of weak vertical gradient observed in the middle and upper stratosphere [*Schmidt and Khedim*, 1991].

CONCLUSIONS

Carbon dioxide is a unique tracer of stratospheric circulation: its atmospheric distribution is maintained by variations in the CO_2 concentration as forced from the surface, rather than by stratospheric chemistry. Our three-dimensional stratospheric circulation model reproduces the gross features observed at mid- and high northern latitudes, i.e., the middle and upper stratosphere have about 6 ppm less CO_2 than the upper troposphere. It predicts that surfaces of constant CO_2 mixing ratio bulge upward in the tropics and slope down towards the poles like those of other long-lived chemical tracers such as N_2O and CH_4. The simulated CO_2 flux into the stratosphere happens almost entirely in the tropics; this predicted lack of extratropical exchange can be tested by detailed measurements across the tropical and mid-latitude tropopause. In particular, the propagation into the mid-latitude stratosphere of the regular growth and annual and longer period fluctuations should proceed from the tropical stratosphere, rather than the mid-latitude troposphere. Consequently, for example, measurements through the tropopause should reveal a much greater attenuation of annual cycle amplitude at mid-latitudes than in the tropics.

The regular rate of CO_2 increase observed in the troposphere provides a time-label for air entering the stratosphere. In the stratosphere, transport processes irreversibly mix a range of CO_2 values; the only information retained after the mixing is the mean concentration. This mean concentration is equivalent to a mean "age" (time since last contact with the troposphere) only if the CO_2 concentration varies linearly with time. Fluctuations in the CO_2 growth rate complicate age determinations. Our simulation indicates that the annual cycle must be accounted for in the lower stratosphere at ER-2 altitudes, but is negligible in the middle and upper stratosphere. Fluctuations of longer period, such as those associated with the ENSO, are less attenuated in the stratosphere than the annual cycle. In our model, a single biennial plus-minus pulse has a signature lasting over five years in the middle and upper stratosphere. Such long-term variations in stratospheric CO_2 could be misconstrued as age fluctuations when, in fact, troposphere-to-stratosphere transport processes need not have changed. However, the response of the model stratosphere to the idealized ENSO forcing with fixed circulation is still small compared to the balloon-observed variability, suggesting a dominant role for interannual transport variations. As a further complication, these interannual changes (for example, the weakening of the Hadley cell associated with ENSO and the quasibiennial oscillation (QBO) of the zonal wind direction in the tropical stratosphere) may be correlated with source variations. The potential stratospheric offsets in CO_2 concentration produced by these correlations could lead to distributions substantially different than predicted here.

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