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Stratospheric Mean Ages and Transport Rates from Observations of Carbon Dioxide and Nitrous Oxide

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Measurements of stratospheric carbon dioxide (CO_2) and nitrous oxide (N_2O) concentrations were analyzed to investigate stratospheric transport rates. Temporal variations in tropospheric CO_2 were observed to propagate into the stratosphere, showing that tropospheric air enters the lower tropical stratosphere continuously, ascends, and is transported rapidly (in less than 1 month) to both hemispheres. The mean age *A* of stratospheric air determined from CO_2 data is approximately 5 years in the mid-stratosphere. The mean age is mathematically equivalent to a conserved tracer analogous to exhaust from stratospheric aircraft. Comparison of values for *A* from models and observations indicates that current model simulations likely underestimate pollutant concentrations from proposed stratospheric aircraft by 25 to 100 percent.

The chemistry of the stratosphere may be strongly perturbed by pollutants as a result of the long residence times (*1–10*) for gases and aerosols in the stratosphere. Rates of transport of pollutants into, within, and out of the stratosphere are thus important parameters that regulate stratospheric composition. The basic characteristics of stratospheric circulation are known from observations of trace gases such as water vapor and ozone (O_3) (*1*) and of particulates from nuclear tests (*10*) and volcanic eruptions (*11*). Air enters the stratosphere at the tropical tropopause, rises at tropical latitudes, and descends at middle and high latitudes to return to the troposphere (*1*, *12*). However, the rates for transport on global scales are poorly known, and quantitative information is critically needed to predict the response of stratospheric O_3 to climatic or chemical change (*13*) or to exhaust deposited in the stratosphere by proposed high-speed civil transports (HSCTs) (*14*).

We present here in situ observations of stratospheric CO_2 and N_2O concentrations that elucidate key aspects of stratospheric transport. Seasonal and annual variations in tropospheric CO_2 are shown to propagate into the stratosphere where CO_2 behaves as a conserved tracer (*15*); N_2O is nearly invariant in the troposphere and is removed by photolysis in the upper stratosphere. These tracers together provide an ideal probe of transport on time scales of importance in the stratospheric circulation.

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Tropospheric CO_2 concentrations oscillate seasonally, reflecting hemispheric cycles of photosynthesis and respiration; seasonal amplitudes near the surface are 1 part per million (ppm) in the south and 3 to 15 ppm (increasing poleward) in the north, with the two hemispheres 6 months out of phase (*16*). In addition, CO_2 increases annually on average by 1.4 ppm year^{-1} as a result of fossil fuel combustion (*17*). The 5 years of near global scale observations of CO_2 reported here reveal that air enters the stratosphere in all seasons and that seasonal cycles of CO_2 in the stratosphere have identical phase in the Northern Hemisphere and Southern Hemisphere. Analysis of the measurements establishes a lower limit for the rate of quasi-horizontal transport from the tropics to mid-latitudes of both hemispheres and defines mean upwelling velocities in the tropics and the mean age of stratospheric air in mid-latitudes. The results also provide critical diagnostics for models of stratospheric transport.

We obtained simultaneous measurements of CO_2, N_2O, and other species at altitudes between 9 and 21.5 km from November 1992 to February 1996, using NASA's ER-2 aircraft. The longest interval between observations was 5 months, with near monthly time resolution between February and November 1994. Sampling included tropical, middle, and high latitudes from 70°S to 61°N (*18*).

Seasonal and annual changes in tropospheric CO_2 propagate across the tropical tropopause and slowly ascend, as shown by vertical profiles of CO_2 in the tropics (Fig. 1A). Maxima in CO_2 were observed at a potential temperature $\theta \approx 435$ K [corresponding to a pressure-altitude of 19 km (*19*)] in October 1994 and in November 1995. A minimum was observed at a similar altitude in February 1996. The observed extrema represent the annual maxima (or minimum) that entered the stratosphere 3 to 4 months earlier at the tropical tropopause at 390 K (~16 km). The 2-ppm increase from October 1994 to November 1995 reflects the trend in tropospheric CO_2 over that time interval. The amplitude of the seasonal variation at 435 K is about 80% of the variation for air entering the stratosphere (see below), a notably small attenuation indicating that vertical advection dominates both vertical diffusion and mixing of older air from mid-latitudes into the tropics (*20–22*).

Observations of CO_2 at northern and southern mid-latitudes show that seasonal variations in CO_2 are transported rapidly from the tropics poleward. However, the seasonality at mid-latitudes is not as well preserved with respect to altitude (or θ) (for

example, Fig. 1B) as in the tropics. Planetary-scale waves in the extratropics induce both large-scale reversible air displacements and horizontal mixing (*12*), resulting in air parcels at a given altitude that have a diverse range of transport histories rather than the relatively narrow distribution found in the tropics (*3*, *22*, *23*). Seasonal variations in CO_2 at mid-latitudes are evident when CO_2 is plotted against a seasonally invariant tracer such as N_2O (for example, Fig. 1C), which removes the variability associated with reversible displacements (*24*) and, to some extent, with irreversible mixing (*25*).

Compact relations between CO_2 and N_2O as in Fig. 1C were consistently observed in the extratropics on the 87 flights from 1992 to 1996, defining the seasonal and interannual evolution of stratospheric CO_2 concentrations. On several occasions data were obtained in both southern and northern mid-latitudes within a 3-week period; the resulting scatterplots within each period are nearly identical (Fig. 2). Air at mid-latitudes in both hemispheres for which θ = 380 to 400 K and N_2O = 305 to 310 parts per billion (ppb) exhibited CO_2 seasonal changes identical in phase and amplitude to changes at the tropical tropopause (Fig. 3A; see also Fig. 1C) (*26*). These observations indicate (i) that inputs from the extratropical troposphere to altitudes above θ = 380 to 400 K are insignificant (*27*), (ii) that air enters the tropical stratosphere continuously throughout the year, and (iii) that air is transported rapidly (<1 month) from the tropics to mid-latitudes of both hemispheres at altitudes near the tropopause (*28*). At higher altitudes (higher θ, lower N_2O concentrations), the CO_2 cycle is more attenuated at mid-latitudes than in the tropics (Fig. 1C) and is completely damped above 19 km (440 K). The complete attenuation may reflect limited transport of air from the tropics to mid-latitudes at these altitudes or simply the dominance of dispersive mixing.

Several important transport quantities can be derived from the phase velocity of

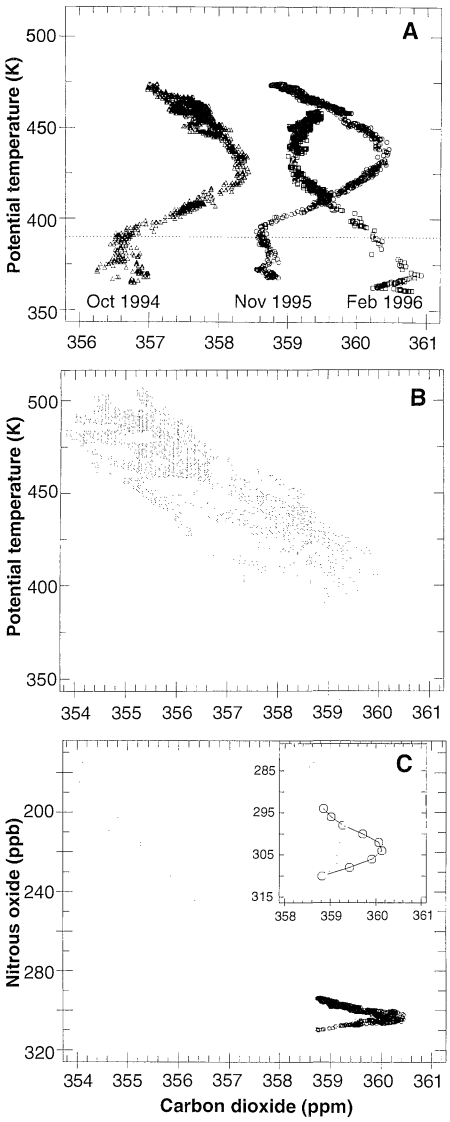

Fig. 1. (**A**) Vertical profiles of CO_2 at tropical latitudes, plotted versus potential temperature (*19*) for flights on 24, 26, and 29 October 1994 (7°S to 7°N; six profiles), 5 November 1995 (2°S to 4°N; two profiles), and 13 February 1996 (2°S to 3°N; two profiles). The dotted line denotes the tropopause at 390 K (~16 to 17 km). (**B**) Profiles of CO_2 at mid-latitudes (35° to 65°N) in November 1995. (**C**) Scatterplot of simultaneous measurements of CO_2 versus N_2O, with N_2O used as a pseudo-vertical coordinate for November 1995; o, tropical data from (A); +, mid-latitude data from (B). (Inset) Same data on an expanded scale with CO_2 averaged in 1-ppb intervals of N_2O.

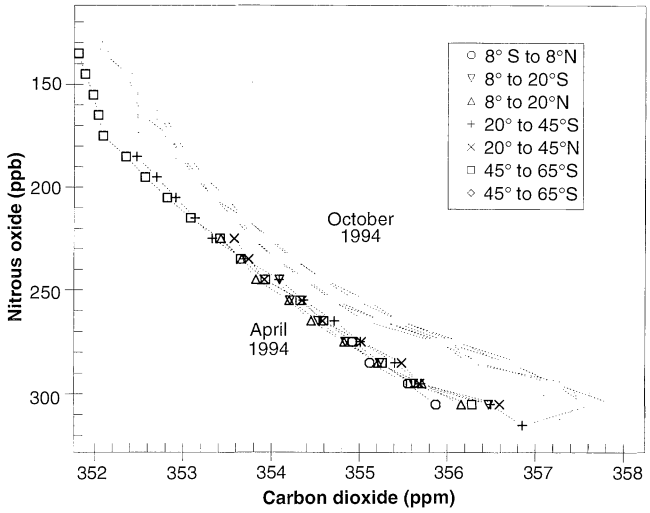

Fig. 2. Scatterplot of CO_2 concentrations, binned and averaged in 10-ppb intervals of N_2O and with respect to latitude, versus N_2O for March through April 1994 (black) and October through November 1994 (gray).

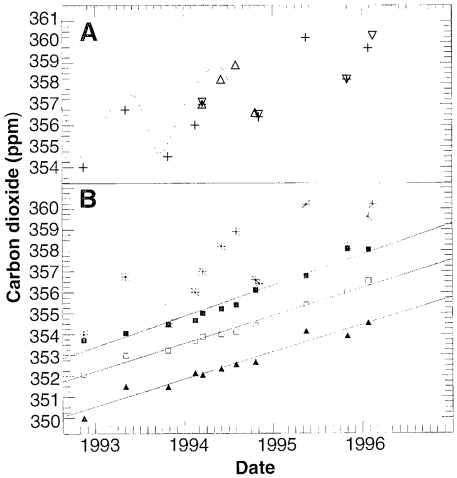

Fig. 3. (**A**) The boundary condition for CO_2 concentrations in air entering the stratosphere irreversibly, as determined by the mean of CO_2 in air with $N_2O \approx 310$ ppb, CO ≈ 30 to 40 ppb, $\theta >$ 380 K, and location above the tropopause altitude. The symbols denote whether the mean was obtained from air sampled in the tropics (∇) or the northern (+) or southern (△) extratropics. Curves show the average of monthly mean surface CO_2 concentrations for Mauna Loa (19°N) and Samoa (14°S) from the National Oceanic and Atmospheric Administration Climate Monitoring and Diagnostics Laboratory Flask Network data (dotted line) and the average delayed by 2 months (solid line). (**B**) Temporal evolution of stratospheric CO_2 concentrations, binned by values of simultaneous N_2O measurements, and the associated mean age of the air: (*), $N_2O \approx 310$ ppb, age = "0" [the boundary condition in (A)]; (■), N_2O = 275 to 280 ppb, age = 2.0 years; (□) 240 to 245 ppb, age = 3.0 years; (▲): 170 to 175 ppb, age = 4.5 years. Ages have 1σ standard deviations of 0.2 year. Lines are linear least squares fits to the CO_2 concentrations, yielding growth rates of 1.5, 1.3, and 1.3 ± 0.1 ppm year^{-1}, respectively.

the CO_2 seasonal signal and from the time lag of stratospheric CO_2 concentrations with respect to the troposphere once the seasonal signals have damped (or averaged) out. Effective upwelling velocities in the tropics can be derived from the altitudes of the extrema in the CO_2 profiles (Fig. 1A) and the time interval between the observation and the date on which the CO_2 extrema entered the stratosphere (*29*), if the locations of the extrema are not significantly altered by mixing with mid-latitude air. The data in Fig. 3A show that the boundary condition for stratospheric CO_2 is accurately represented by the mean of ground-based observations from southern and northern subtropical stations [Samoa (14°S) and Mauna Loa (19°N), respectively], retarded by 2 months. Knowledge of this boundary condition allows estimates of the transit time for seasonal extrema within ± 2 weeks. The resulting ascent rates (Table 1) are remarkably similar to seasonally resolved velocities computed from radiative heating rates based on the use of satellite and climatological data (*30*) but are independent of radiative transfer models. Velocities derived from CO_2 extend to lower altitudes into the region near the tropical tropopause where air enters the stratosphere and heating rates are small.

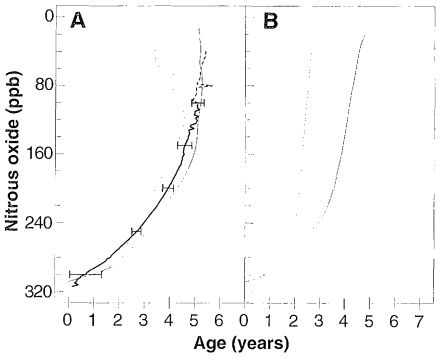

Fig. 4. (**A**) Mean ages of mid-latitude air for 1992 through 1996 derived from all CO_2 observations (binned in 2-ppb intervals of N_2O), plotted against N_2O (as a pseudo-vertical coordinate) (heavy solid line). Representative standard deviations of ages for a given value of N_2O are shown by horizontal bars and are ≤0.2 year for N_2O < 280 ppb. For N_2O < 90 ppb (heavy dashed line), data were obtained in May 1993 and October 1994 only and represent air that had descended in the polar vortices. The thin lines show results for a tracer model with transport based on a monthly (solid) or annual (dashed) mean residual circulation. (**B**) Model results for the tropics (±15°). Observations of CO_2 in low N_2O and high-altitude tropical air have not yet been acquired.

The mean age (*A*) of air at a point in the stratosphere is commonly defined as the time lag between the observed stratospheric concentration and tropospheric values for a conserved tracer with a time-varying concentration in the troposphere, for example, CO_2 (*2*, *3*) or sulfur hexafluoride (SF_6) (*9*, *31*). Hall and Plumb (*23*) showed that, if the tropospheric concentration of the trace gas increases linearly with time, this time lag is identical to an average over the ensemble of transit times from the troposphere and therefore represents the true average time (or "mean age") since the air entered the stratosphere. We calculated mean stratospheric ages from CO_2 data, using a linear fit to the deseasonalized boundary condition in Fig. 3A after correcting for methane (CH_4) oxidation (*15*). For altitudes at which seasonal variations in CO_2 have damped out, that is, higher than 3 to 5 km above the tropopause at mid-latitudes (N_2O < 280 ppb), remarkably precise ages for a given value of N_2O were obtained for all observations from 1992 to 1996, including 2.0 ± 0.2 years for N_2O = 275 to 280 ppb and 4.5 ± 0.2 years for N_2O = 170 to 175 ppb (Fig. 3B), and values exceeding 5 years for low N_2O air in the polar vortices (Fig. 4A) (*32*). These ages are in good agreement with those derived from SF_6 (*9*, *33*). The uniformity of the ages derived from CO_2 data obtained frequently over a 5-year period and the agreement with ages from SF_6 observations indicate that negligible errors are introduced by the CO_2 seasonal cycle or by interannual variations in CO_2 growth rates.

The mean age *A* is a quantity closely related to the mean residence time for pollutants with stratospheric sources (exhaust from HSCTs, for example) that can be accurately determined from atmospheric observations. We show here that *A* is mathematically equivalent to a conserved pseudo-tracer with unit stratospheric input rate and zero tropospheric concentration. The mass continuity equation for the concentration C of a conserved tracer is

$$\partial C/\partial t + \nabla\cdot\Phi_C = 0 \qquad (1)$$

where Φ_C is the flux. If C at the tropopause (z = 0) increases linearly with time t [$C(x, y, 0; t) = C_0 + bt$] and Φ_C = 0 normal to the upper boundary, we can define $A = -(C - C_0 - bt)/b$, giving

$$\partial A/\partial t + \nabla\cdot\Phi_A = 1 \qquad (2)$$

The right side of Eq. 2 represents a uniform "source" (aging rate), and the concentration (age) at the lower boundary is $A(x, y, 0; t) = 0$.

A simple model that solves Eq. 2, with the second term computed from the monthly residual circulation (*34*) averaged into a tropical and two mid-latitude zones, reproduces the distribution of *A* in the extratropics derived from CO_2 (Fig. 4, A and B) (*35*). The agreement is surprising because the residual circulation in the model is completely advective; it may be explained by noting that model ages are 20% smaller at mid-latitudes and almost 40% smaller in the tropics if the residual circulation is averaged annually instead of monthly. We therefore infer that fluctuations in the sign and magnitude of the residual circulation on monthly time scales provide significant mixing in the model on global scales, sufficient to match the mean ages derived from the observations.

The pseudo-tracer *A*, constrained by observations, represents a critical parameter to test models used to predict the distributions of pollutants from HSCTs. For HSCTs flying at Mach 2.4 with an emission index of 15 g of NO_x per kilogram of fuel burned (*14*), the model described above predicts peak enhancements in stratospheric nitrogen oxides (ΔNO_y) of >5 ppb for 15° to 90°N and >3 ppb for 15°N to 15°S, or 25 to 100% greater than predicted by models used in recent assessments (*14*). This difference is consistent with the underestimation of mean ages derived from SF_6 distributions simulated by assessment models (*36*). Hence, these models likely underestimate ΔNO_y resulting from HCSTs by 25 to 100%.

These measurements of CO_2 and N_2O provide quantitative information on stratospheric transport rates and unique constraints on stratospheric models, including (i) a lower limit of 1 month on rates for the dispersal of tracers from tropical to mid-latitudes of both hemispheres; (ii) seasonally resolved tropical upwelling velocities in the range 0.2 to 0.3 mm s^{-1};

Table 1. Upwelling velocities in the lower tropical stratosphere.

Date of tropical profile	Potential temperature range (K)	Time period for ascent from tropopause	Transit time (days)	Ascent rate (K day^{-1})	Ascent rate (10^{-3} m s^{-1}) (*38*)
24, 26, 29 Oct. 1994	390 → 430 ± 5	1 July → 26 Oct.	118 ± 14	0.34 ± 0.06	0.19 ± 18%
5 Nov. 1995	390 → 438 ± 4	1 July → 5 Nov.	128 ± 14	0.38 ± 0.07	0.21 ± 18%
13 Feb. 1996	390 → 440 ± 10	15 Nov. → 13 Feb.	90 ± 14	0.56 ± 0.14	0.31 ± 25%

and (iii) accurate mean ages for the lower and middle stratosphere, including 2 years at altitudes of 15 km and >5 years for altitudes above 25 km. Values for A represent a powerful diagnostic for stratospheric models, readily computed by the use of a simple pseudo-tracer. Reproducing the observed temporal and spatial evolution of the relation between CO_2 and N_2O represents a further challenging test. With this information it should be possible to improve models and provide enhanced confidence in predictions of environmental effects on stratospheric O_3.

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15. The concentration of stratospheric CO_2 is conserved except for small quantities produced by CH_4 oxidation. In our analysis of mean age, we accounted for this input using simultaneous measurements of CH_4 (C. Webster *et al.*, personal communication), assuming tropospheric CH_4 concentrations of 1.7 ppm (by volume) (ppmv) for 1986 through 1996 [E. J. Dlugokencky, L. P. Steele, P. M. Lang, K. A. Masarie, *J. Geophys. Res.* **99**, 17021 (1994)].
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17. The global rate of increase for tropospheric CO_2 was 1.4 ppm $year^{-1}$, averaged over 20 years or over the last 6 to 10 years (*16*).
18. We measured CO_2 at 4 Hz (then median-filtered to 0.5 Hz) using nondispersed infrared absorption; the long-term precision was ±0.05 ppmv (1σ), derived from 215 in-flight determinations of CO_2 (from 86 flights) for a "surveillance" standard, sampled at 1- to 2-hour intervals through the sample line and refilled after 15 to 20 flights. We also monitored the concentration of the surveillance cylinder on the ground, using independent standards traceable to the Scripps-World Meteorological Organization scale (*4*). We measured N_2O at 1 Hz, using tunable diode laser spectroscopy [J. R. Podolske and M. Loewenstein, *Appl. Opt.* **32**, 5324 (1993)], CO and CH_4 at 0.3 Hz by tunable diode laser spectroscopy [C. R. Webster, R. D. May, C. A. Trimble, R. G. Chave, J. Kendall, *ibid.* **33**, 454 (1994)], temperature profiles at 0.1 Hz by microwave radiometry [B. L. Gary, *J. Geophys. Res.* **94**, 223 (1989)], and static temperature and pressure desampled to 1 Hz (T. P. Bui, K. R. Chan, S. W. Bowen, personal communication). Field campaigns, sites, and latitudes sampled were as follows: *SPADE*, Moffett Field, CA, November 1992: 20°N to 40°N; April through May 1993: 14°N to 60°N; October 1993: 14°N to 60°N; *ASHOE/MAESA*, California, Hawaii, Fiji, and Christchurch, New Zealand, February 1994: 35°N to 61°N; March through April 1994: 68°S to 38°N; May through June 1994: 69°S to 20°S; July through August 1994: 69°S to 19°S; October through November 1994: 70°S to 60°N; *STRAT*, California and Hawaii, May 1995: 14°N to 62°N; October through November 1995: 2°S to 59°N; January through February 1996: 2°S to 53°N.
19. Potential temperature (θ) is the temperature of air if compressed adiabatically to 1 atm; it is conserved in the absence of energy exchange with the environment (as when air moves adiabatically). On average, θ increases monotonically with altitude.
20. The attenuations are consistent with a 30 to 50% admixture of mid-latitude air into the tropics, an amount indicated by the studies of C. M. Volk *et al.* [*Science* **272**, 1763 (1996)], K. Minschwaner *et al.* [*J. Geophys. Res.* **101**, 9433 (1996)], and L. M. Avallone and M. J. Prather (*ibid.*, p. 1457).
21. A seasonal water vapor cycle has also been shown to persist as air is advected vertically in the tropics (*37*). The phase relation between the seasonal cycles of CO_2 and water vapor is preserved as air is transported to mid-latitudes [K. A. Boering *et al.*, *Geophys. Res. Lett.* **22**, 2737 (1995)]. At present, the boundary condition for water vapor concentrations in air entering the stratosphere is not accurately known (this quantity is not known because of the complex physical-chemical mechanisms associated with the transport from the troposphere to the stratosphere).
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25. The interplay between vertical advection and mixing by planetary-scale waves produces nearly parallel surfaces of constant mixing ratio for long-lived tracers at mid-latitudes, providing compact scatterplots between pairs of tracers [J. R. Holton, *J. Geophys. Res.* **91**, 2681 (1986); J. D. Mahlman *et al.*, *ibid.*, p. 2687; R. A. Plumb and M. K. W. Ko, *ibid.* **97**, 10145 (1992)].
26. Potential temperatures from 380 to 400 K divide the stratosphere into the "overworld," where parcels moving adiabatically remain above the tropopause at all latitudes, and the "middleworld," where parcels may enter or exit the stratosphere adiabatically at low latitudes (*12*). To rise into the overworld, air must cross θ = 380 K in the tropics. Figure 3A shows the boundary condition for CO_2 entering the overworld.
27. Seasonal variations in CO_2 in the northern extratropical troposphere have amplitudes that are too large and those in the south have amplitudes that are too small and have the incorrect phase to be consistent with the stratospheric observations in the overworld (*26*). The three-dimensional model results of Hall and Prather (*3*) simulating stratospheric CO_2 concentrations are consistent with observed features; their model results and the few measurements available to date [T. Nakazawa, K. Miyashita, S. Aaki, M. Tanaka, *Tellus B* **43**, 106 (1991); H. Matsueda and H. Inoue, *Atmos. Environ.* **30**, 1647 (1996)] suggest seasonal variations in the upper tropical troposphere similar to the observed boundary condition in Fig. 3A.
28. Volcanic aerosols from Mount Pinatubo were rapidly transported from tropical to northern and southern mid-latitudes at altitudes below 21 km (*11*); CO_2 data show analogous transport for gases continuously entering the tropical stratosphere from the troposphere.
29. Modeling of the age spectrum for a conserved tracer with a seasonal cycle (*22*) indicates that bulk "modal" transit times may be derived from the phase lag time of a propagating periodic signal in the lower tropical stratosphere because advection dominates dispersion in this region.
30. Eluszkiewicz *et al.* (*34*) computed upwelling velocities of 0.23 and 0.30 mm s^{-1} for July through October and November through February, respectively, averaged from the tropopause to 21 km for 15°S to 15°N. Velocities calculated by Rosenlof (*7*) were 0.34 and 0.09 mm s^{-1} in January and July 1993, respectively, for upwelling at 50 mbar (21 km) from 10°S to 10°N, with an annual mean of 0.21 mm s^{-1}. Uncertainties were estimated to be ± 50% in the lower stratosphere and higher near the tropopause. Tropical CO_2 profiles (3°S to 3°N) obtained in March 1994 up to a maximum altitude of 460 K decreased monotonically from the tropopause, suggesting that the minimum had propagated to θ ≥ 460 K and yielding a mean ascent rate of ≥ 0.56 K/day (or 0.31 mm s^{-1}) over the period 15 November through 22 March. The propagationof seasonal cycles in total hydrogen (H_2O + 2 CH_4) to higher altitudes from satellite observations led Mote *et al.* (*37*) to suggest ascent rates of about 0.4 mm s^{-1} for northern winter and about 0.2 mm s^{-1} for the rest of the year.
31. J. W. Elkins *et al.*, *Geophys. Res. Lett.* **23**, 347 (1996); L. S. Geller *et al.*, in preparation.
32. The mean age for the mid-stratosphere (for example, 5.0 years at N_2O = 110 ppb in Fig. 4A) is 1.5 to 3 times longer than reported values of the turnover time for mass in the stratosphere, $\tau_m \equiv$ (stratospheric mass)/(flux through the tropical tropopause) (*5–8*). For example, Rosenlof and Holton (*6*) reported values in the range 2 to 3 years from calculations of the residual circulation in which the velocities for vertical advection were consistent with those in Table 1. However, mean age may differ from the turnover time as a result of mixing. Consider a tube of length L that receives air flow with velocity v and has a CO_2 concentration at the inlet that is increasing linearly with time t, $C(0) = C_0 + bt$. In the absence of dispersion, the concentration at point x is $C(x, t) = C_0 + b(t - x/v)$ and the mean concentration is $C_{mean}(t) = C_0 + b(t - \tau/2)$, where $\tau \equiv L/v$; in a well-mixed tube, $C_{mean}(t) = C_0 + b(t - \tau)$. Thus, the mean ages (that is, the lag times with respect to the boundary concentration) are τ/2 and τ, respectively.
33. Simultaneous measurements of SF_6 were obtained in October through November 1994 (*31*); ages derived from the time lag between stratospheric observations and the concentration measured in the upper tropical troposphere, agree to better than 0.2 year between CO_2 and SF_6. Ages derived from the time lags with respect to global surface mean data for both CO_2 and SF_6 also agree to within 0.3 year but are longer by 0.3 to 0.7 year than those derived from the apparent boundary conditions for CO_2 and SF_6 in the upper tropical troposphere.
34. J. Eluszkiewicz *et al.*, *J. Atmos. Sci.* **53**, 217 (1996).
35. A tropical pipe model [R. A. Plumb, *J. Geophys. Res.* **101**, 3957 (1996)] was constructed to include three regions, divided at 15°N and 15°S. A detailed model description will be presented elsewhere (S. C. Wofsy *et al.*, in preparation).
36. M. K. W. Ko, personal communication.
37. P. W. Mote *et al.*, *J. Geophys. Res.* **101**, 3989 (1996).
38. We converted the observed ascent rates in kelvin per day to velocities in meters per second using National Meteorological Center temperature profiles from the Halogen Occultation Experiment temperature retrieval on-board the Upper Atmospheric Research Satellite (B. Pierce, personal communication).

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