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Age as a diagnostic of stratospheric transport

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Abstract. Estimates of stratospheric age from observations of long-lived trace gases with increasing tropospheric concentrations invoke the implicit assumption that an air parcel has been transported intact from the tropopical tropopause. However, because of rapid and irreversible mixing in the stratosphere, a particular air parcel cannot be identified with one that left the troposphere at some prior time. The parcel contains a mix of air with a range of transit times, and the mean value over this range is the most appropriate definition of age. The measured tracer concentration is also a mean over the parcel, but its value depends both on the transit time distribution and the past history of the tracer in the troposphere. In principle, only if the tropospheric concentration is increasing linearly can the age be directly inferred. We illustrate these points by employing both a one-dimensional diffusive analog of stratospheric transport, and the general circulation model (GCM) of the Goddard Institute for Space Studies (GISS). Within the limits of the GCM, we estimate the time over which tropospheric tracer concentrations must be approximately linear in order to determine stratospheric age unambiguously; the concentration of an exponentially increasing tracer is a function only of age if the growth time constant is greater than about 7 years, which is true for all the chlorofluorocarbons. More rapid source variations (for example, the annual cycle in CO_2) have no such direct relationship with age.

Introduction

Many of the radiatively and chemically important trace gases in the stratosphere have tropospheric origin. The distribution and evolution of these gases is controlled by the stratospheric circulation and troposphere-stratosphere exchange. One measure of the ability of the atmosphere to transport trace gases into the stratosphere is the age of stratospheric air [*Bischof et al.*, 1985]. This timescale has been used, for example, in estimating ozone depletion potentials of HCFCs in the winter polar stratospheric vortex [*Pollock et al.*, 1992], as well as polar stratospheric vortex concentrations of inorganic chlorine (L. E. Heidt et al., unpublished manuscript, 1993). Age will likely become an increasingly useful conceptual tool as monitoring of anthropogenic trace gases intensifies. It is therefore important to define this time scale explicitly, and to determine the necessary criteria a tracer must satisfy in order that its measurements yield age unambiguously, or, conversely, that its concentration may be deduced from a knowledge of age.

Quantitative estimates of age have been obtained from measurements of tracers with negligible stratospheric sources and sinks, and whose tropospheric concentrations are systematically increasing with time, by observing the time lag between stratospheric and tropospheric concentrations. Thus, *Schmidt and Khedim* [1991] found a typical age of about 5 years for middle stratospheric air in middle and high latitudes on the basis of measured CO_2 concentrations, while *Pollock et al.* [1992] obtained a value of 3–5 years for lower stratospheric air from observations of CF_3CF_2Cl (CFC-115). *Hall and Prather* [1993] determined the spatial distribution of stratospheric age from general circulation model experiments with a CO_2-like tropospheric source, but also drew attention to the ambiguities introduced into the calculation by seasonal and interannual fluctuations in tropospheric concentrations.

Our goal in this paper is to clarify the meaning of age. It is tempting to interpret the age of a stratospheric air parcel simply as a transit time from the tropical tropopause. However, air parcels do not maintain their integrity over such climatological timescales; an air par-

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cel injected into the stratosphere is rapidly torn apart by wave-induced, irreversible mixing processes. This point was identified by *Plumb and Ko* [1992] as crucial in the establishment of the equilibrium distributions of long-lived tracers and of the observed tracer correlations. Any given stratospheric air parcel therefore comprises many components, each with a different history since last leaving the troposphere [*Hall and Prather*, 1993]. It is strictly correct to speak, not of the transit time of an air parcel as a single quantity, but rather of a statistical distribution of transit times of all the irreducible elements that comprise the parcel. In mathematical terms, this distribution is the Green's function for the differential operator governing the transport of tracer from the tropical upper troposphere. *Kida* [1983] also recognized the statistical nature of age. In keeping with his terminology, we call the distribution of transit times the age spectrum.

Long-lived trace gases of tropospheric origin, and whose tropospheric concentration varies in time, label air entering the stratosphere. The measured tracer concentration of a stratospheric air parcel is a mean value over the distribution of irreducible elements comprising the parcel. One can, of course, define a time scale equal to the time by which the stratospheric concentration lags that of the tropical tropopause. It is important to recognize, however, that in general such a time scale depends on the history of the tracer source. Time lag is thus a nonunique expression of transport time scales. We therefore choose to define the age of a stratospheric air parcel in a species-independent way, as the mean over the age spectrum. While this definition departs from the more conventional time lag, we shall in fact show that the age thus defined is equal to the time lag for a tracer whose tropospheric concentration has grown linearly for a sufficiently long time.

In what follows, we first present a formal and general exposition of these arguments, and follow by illustrating their implications in a simple one-dimensional diffusion model. The spatial dependence of age and the age spectrum in the stratosphere is discussed using results from a three-dimensional chemical transport model (CTM) employing winds computed from the stratospheric general circulation model (GCM) at the Goddard Institute for Space Studies (GISS). Within the limits of the model, we find the age of stratospheric air to range up to 4 years. In addition, the stratospheric time lag in tracer concentration from tropospheric values undergoing an increasing trend is approximately equal to age for species whose exponential growth time constant is greater than about 7 years. The structure of the age spectrum, and its height dependence, explains in a natural way both the distribution of linearly increasing tracers such as CO_2 and the rapid attenuation with height of the annual cycle of CO_2.

General Development

The mixing ratio n of a conserved tracer satisfes the continuity equation

$$\frac{\partial n}{\partial t} + \mathcal{L}(n) = 0 \tag{1}$$

where $\mathcal{L}$ is the relevant differential transport operator. In order to keep the discussion as simple as possible, we assume that the transport described by $\mathcal{L}$ is stationary in time, thus neglecting seasonal and interannual variations in transport. We shall see that seasonality of transport is a relatively small effect in the GISS GCM when the distribution of tracer in the stratosphere is considered over a number of years. The influences of transport variations are discussed later.

Now, if $n = n(P,t)$ is prescribed at some point P_0, such that $n(P_0,t) = 0$ for $t < 0$, then the general solution at some other point P is

$$n(P,t) = \int_0^t n(P_0, t-t')G(P,P_0,t')dt' \tag{2}$$

The function G is the Green's function, such that if $n(P_0,t) = \delta(t-t_0)$, where $\delta(t)$ is the Dirac delta function, then $n(P,t) = G(P,P_0,t-t_0)$. (We shall later use this relationship to determine G.) If $n(P_0,t) = \Theta(t)$, where Θ is the Heaviside function, the long-time solution must be $n(P,t) = 1$ for all points P, whence

$$\int_0^\infty G(P,P_0,t')dt' = 1 \tag{3}$$

The forms of equations (2) and (3) suggest a straightforward interpretation of the Green's function in terms of transport timescales. We identify the point P_0 as the equatorial tropopause, where we believe most air enters the stratosphere [*Brewer*, 1949, *Holton*, 1990]. Air parcels leaving this region are rapidly torn apart by irreversible mixing processes associated with breaking waves [e.g., *McIntyre*, 1992 and references therein]. Tracer variance will cascade down to ever smaller scales, until ultimately it is detroyed by molecular diffusion. A parcel of air at point P in the stratosphere, therefore, contains rich fine-scale tracer structure. Following *Plumb and McConalogue* [1988], we consider this parcel as composed of a continuum of irreducible material elements, each of which conserved its mixing ratio from the time it left P_0. Each element has, in general, traversed a unique path with a unique transit time. (For the sake of this argument, it does not matter whether these elements retain their separate identities or mix to homogenize the parcel, since we shall regard the mean parcel mixing ratio as the quantity of interest. In reality, we expect only very recent transport to be manifest as microstructure within the parcel, as older components have been thoroughly mixed by small-scale processes.) We are thus led, in light of (2), to interpret the Green's function $G(P,P_0,t)$ as the distribution of transit times (i.e., the age spectrum) within the air parcel such that the mass fraction of the air parcel with transit times between t' and $t'+dt'$, and which therefore at time t has mixing ratio $n(P_0,t-t')$, is $G(P,P_0,t')dt'$; the definition of the age (the average of the component transit times) follows immediately as

$$\Gamma(P,P_0) = \int_0^\infty t\, G(P,P_0,t)dt \tag{4}$$

Returning now to (2), it can be seen that, if the air parcel at P traveled intact (without mixing) from P_0 in a time t_0, then $G = \delta(t - t_0)$. The age is $\Gamma = t_0$, and

$$n(P,t) = n(P_0, t - \Gamma) \tag{5}$$

In the case of simple advection, therefore, the mixing ratio at P simply lags that at P_0 by a time equal to the age of the air pacel at P, whatever the history of the source $n(P_0, t)$.

In the more general case, however, when the age spectrum G has a finite width, it is not apparent from (2) that such a relationship holds. Indeed, we shall show by example below that it does not. It does hold, however, in the long-time limit with a linearly growing source

$$n(P_0,t) = \begin{cases} 0 & \text{if } t < 0 \\ \gamma t & \text{otherwise} \end{cases} \tag{6}$$

Then (2) gives

$$\begin{aligned} n(P,t) &= n(P_0,t)\int_0^t G(P,P_0,t')dt' \\ &- \gamma \int_0^t t' G(P,P_0,t')dt' \end{aligned} \tag{7}$$

As $t \rightarrow \infty$, by (3) and (4), this reduces to (5). Such a limit is valid for t greater than a few years, so that, as we shall see, it is in the region where $G(P, P_0, t)$ asymptotically approaches zero. This is equivalent to integrating beyond the transient response of the system defined by (1) and (6). Therefore, as recognized by *Hall and Prather* [1993], the association of age with lag time is valid for linearly growing sources (of which CO_2 is the best known example, albeit with additional annual and interannual components).

Consider now, however, an exponentially growing source (such as CFCs over much of the past two decades)

$$n(P_0,t) = \begin{cases} 0 & \text{if } t < 0 \\ n_0(e^{\sigma t} - 1) & \text{otherwise} \end{cases} \tag{8}$$

for $\sigma > 0$. From (2), for large time,

$$\begin{aligned} n(P,t) &= n_0\left(e^{\sigma t}\int_0^\infty e^{-\sigma t'} G(P,P_0,t')dt' - 1\right) \\ &= n(P_0, t - \Gamma_e) \end{aligned} \tag{9}$$

thus defining a lag time for the tracer

$$\Gamma_e = -\frac{1}{\sigma}\ln \int_0^\infty e^{-\sigma t'} G(P,P_0,t')dt' \tag{10}$$

In general, this does not reduce to (4) (except, as we shall see, in the limit of small σ) and is a function of σ. Therefore interpreting as age the lag time between stratospheric and tropospheric mixing ratios for exponentially growing tracers is not strictly correct, and will lead to different values for tracers growing at different rates. Whether this is a serious problem in practice depends on how small σ must be for Γ_e to be a valid approximation to (4).

Changing time variables in (10) to $t - \Gamma$, and expanding the exponent in σt to $O(\sigma^2)$ yields

$$\Gamma_e \sim -\frac{1}{\sigma}\ln e^{-\sigma\Gamma}(1 + \sigma^2\Delta^2) \tag{11}$$

or

$$\Gamma_e \sim \Gamma(1 - \sigma\frac{\Delta^2}{\Gamma}) \tag{12}$$

where Δ is the width of the age spectrum, defined by

$$\Delta^2(P,P_0) = \frac{1}{2}\int_0^\infty (t-\Gamma)^2 G(P,P_0,t)dt \tag{13}$$

Therefore $\Gamma_e \sim \Gamma$ to $O(\sigma)$ is a good approximation to the extent that

$$\frac{1}{\sigma} \gg \Delta(\frac{\Delta}{\Gamma}) \tag{14}$$

The Δ factor on the right hand side of the inequality determines the absolute nearness of the time dependence to linearity over the width of the age spectrum, and, therefore, the absolute difference of the lag time from the age. The Δ/Γ factor scales this absolute difference to the age. We will use approximation (12) in conjunction with GCM results to estimate the error in age as determined by exponentially growing long-lived tracers of tropospheric origin such as CFCs.

As a third example (with CO_2 seasonality in mind), consider a periodic source

$$n(P_0,t) = Re\ [in_0\ e^{i\omega t}] \tag{15}$$

For large time, (2) now gives

$$n(P,t) = Re\ [in_0\ Ae^{i\omega(t-\Gamma_\omega)}] \tag{16}$$

where A, the amplitude, and Γ_ω, the phase lag time, are real, and

$$Ae^{-i\omega\Gamma_\omega} = \int_0^\infty e^{-i\omega t'} G(P,P_0,t')dt' \tag{17}$$

In the long-period limit where

$$\frac{1}{\omega} \gg \frac{\Delta^2}{\Gamma} \tag{18}$$

(17) reduces to

$$Ae^{-i\omega\Gamma_\omega} \sim (1 - \omega^2\Delta^2)e^{-i\omega\Gamma} \tag{19}$$

to $O(\omega^2)$, whence $\Gamma_\omega \sim \Gamma$; once again there is a simple time lag whose value is the age. As we shall see below, however, an annual cycle in the stratosphere does not satisfy this limit, in which case we cannot associate the time lag of the phase with age.

One-Dimensional Diffusion Analog

Selection of a model for the atmospheric transport illustrates the general statements above by providing an explicit Green's function. Much use has been made of two-dimensional models employing advective-diffusive continuity equations, within which wave-induced mixing is parameterized by diffusivity matrices. For tracers of long enough chemical lifetimes, the transport may be further reduced to a one-dimensional flux gradient relation [*Holton*, 1986, *Plumb and Ko*, 1992]. Motivated by this work, we select a one-dimensional diffusive model. However, because our goal at this point is not to quantify stratospheric transport, but rather to illustrate the points of the previous discussion, we choose a diffusion equation simple enough to allow analytic solution: mass-weighted diffusion with constant and uniform coefficients. This system has enough likeness to stratospheric transport to yield insight into the meaning of age and the age spectrum. In the following section, we employ a GCM to estimate relevant time scales quantitatively.

The tracer continuity equation for this idealized one-dimensional system is

$$\rho\frac{\partial n(z,t)}{\partial t} = K\frac{\partial}{\partial z}(\rho\frac{\partial n(z,t)}{\partial z}) \qquad (20)$$

where $\rho = \rho_0 e^{-z/H}$ is the density of air, n the tracer mass mixing ratio, and K a constant diffusion coefficient. The age spectrum of this system at z is the Green's function, the solution to (20) with the boundary condition $n(0,t) = \delta(t)$, which is

$$G(z,t) = \frac{z}{2\sqrt{\pi K t^3}}\exp\left[\frac{z}{2H} - \frac{Kt}{4H^2} - \frac{z^2}{4Kt}\right] \qquad (21)$$

In Figure 1 we plot $G(z,t)$ versus t for different values of z/H. The location of the peak of $G(z,t)$ occurs at greater t as z increases. In addition the width increases and the magnitude decreases.

The age of the diffusive system at a point z is given by (4), the first moment of the Green's function. The integration produces

$$\Gamma = \frac{H}{K}z \qquad (22)$$

As must be the case, the age increases monotonically with distance from the source.

The age, given by (4), is the lag time in tracer concentration at z if the time dependence is linear at $z = 0$ (or, more specifically, obeys (6)). On the other hand, if the time dependence is exponential with time constant σ^{-1}, one obtains, as a lag time, Γ_e given by (10). Evaluating (10) with the Green's function (21) yields

$$\Gamma_e = \frac{z}{2\sigma H}\left[\sqrt{1+\frac{4\sigma H^2}{K}} - 1\right] \qquad (23)$$

Notice that Γ_e is σ dependent: different rates of increase produce different lags.

Under what conditions does the exponential lag time approximate the true age? According to our general development, this is the case to the extent that $\sigma^{-1} \gg \Delta^2/\Gamma$. The width Δ of the age spectrum, obtained by substituting (21) into (13), is

$$\Delta = \frac{\sqrt{H^3 z}}{K} \qquad (24)$$

Given (22), the appropriate limit is therefore

$$\frac{1}{\sigma} \gg \frac{H^2}{K} \qquad (25)$$

This is simply the timescale for diffusion over a scale height. It is straightforward to verify from (23) that $\Gamma_e \sim \Gamma$ in this limit.

As the second example, consider an oscillating boundary condition for equation (20). With a time dependence at $z = 0$ given by (15) and the Green's function (21), the steady state ($t \rightarrow \infty$) solution (2) is

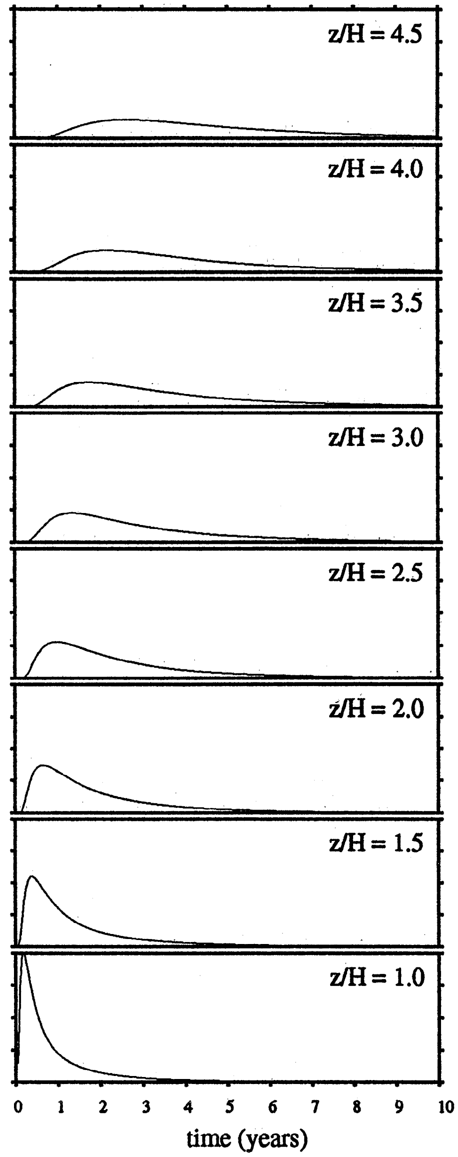

Figure 1. The Green's function (the response to a delta function time dependence at the tracer source) for the one-dimensional mass weighted diffusion equation plotted for different values of z/H, as labeled. The amplitudes are normalized to peak at unity for $z/H = 1$. Here, $K = 3.1$ m^2/s.

$$n(z,t) = A\cos[\omega(t - \Gamma_\omega)] \quad (26)$$

where

$$A = n_0 \exp[-\frac{z}{2H}(1 + \frac{16\omega^2 H^4}{K^2})^{1/4}\cos\phi + \frac{z}{2H}] \quad (27)$$

$$\Gamma_\omega = \frac{z}{2H\omega}(1 + \frac{16\omega^2 H^4}{K^2})^{1/4}\sin\phi \quad (28)$$

and

$$\phi = \frac{1}{2}\tan^{-1}(\frac{4H^2\omega}{K}) \quad (29)$$

This is diffusive wave propagation. The phase shift $\omega\Gamma_\omega$ increases linearly with height, while the amplitude decays exponentially. The higher the frequency, the more rapid the amplitude decay.

The oscillating source produces a time lag in phase (28) that depends not only on the diffusive transport, but also on the frequency of forcing. However, like the exponential case, in the low frequency limit (18) the amplitude becomes simply $A \sim n_0$ to $O(\omega)$, while (28) reduces to $\Gamma_\omega \sim \Gamma$ to $O(\omega^2)$. In this limit, the oscillation is approximately linear over the width of the age spectrum.

In the stratosphere, as we shall see, the GCM determined values of Δ^2/Γ range from 0.4 to 0.8 years, so that annual cycles ($\omega = 2\pi/1\ year$) cannot be considered low frequency. In the high-frequency limit, $\omega^{-1} \ll \Delta^2/\Gamma$, the oscillation period is much less than the time to diffuse over a scale height. This limit is equivalent to wave propagation in a diffusive system with no mass weighting ($H \to \infty$). From (27) and (28),

$$A \sim n_0 e^{-z\sqrt{\omega/2K}} \quad (30)$$

and

$$\Gamma_\omega \sim \frac{z}{\sqrt{2K\omega}} \quad (31)$$

The amplitude decays by e^{-1} in a wavelength $\sqrt{2K/\omega}$. Note that the phase lag decreases with increasing ω. From (28), $\Gamma_\omega < \Gamma$ for all $\omega > 0$, consistent with the GCM result that the CO_2 annual cycle propagates more rapidly than the increase with height of the stratospheric age [*Hall and Prather*, 1993].

Three-Dimensional Transport Simulations

In order to determine the role of the age spectrum in summarizing atmospheric transport ability, we perform three-dimensional stratospheric transport simulations of hypothetical conserved tracers. The use of such a model allows us to quantify (within the limits of the model) timescales of the spectrum and therefore to make predictions on the accuracy of associating the lag times of various tracers with stratospheric age. The simulations employ a CTM which utilizes winds computed by the stratospheric version of the GISS GCM. We first briefly summarize the computer model, and then discuss the simulations and their results.

The Chemical Transport Model

Prather et al. [1987] documented a tropospheric version of the CTM, and the stratospheric version has been employed to study various trace gases [*Prather et al.*, 1990*a*, 1990*b*; *Hall and Prather*-1993]. The CTM employs the lowest 21 layers of winds computed from the 23-layer stratospheric version of the GISS GCM [*Rind et al.*, 1988]. The winds advect tracer every 8 hours with an algorithm that limits numerical diffusion by conserving second-order moments of tracer distribution within each grid box [*Prather*, 1986]. Transport by wet and dry convection occurs in the troposphere as follows: every time step the CTM moves a fraction of tracer mass in a grid box at level k to level m ($m > k$). This fraction is determined by the monthly averaged frequency of convective events in the GCM connecting m and k at each horizontal grid point. Information of the first- and second-order vertical moments is lost during the transport, but horizontal information is retained. Subsidence within the grid box to balance the convective mass flux is treated as vertical advection. To parameterize horizontal dispersal associated with deep convection, the CTM diffuses tracer during such events with a coefficient tuned to tropospheric tracer observations. No convective or diffusive transport is included in the stratosphere. See *Prather et al.* [1987] for further details, and *Hansen et al.* [1983] for a description of the GCM convection.

The horizontal resolution is 7.83° latitude by 10° longitude. In the vertical, nine sigma layers represent the surface to 100 mbar, while the 12 stratospheric layers lie between fixed pressure levels reaching to approximately 0.05 mbar. In the simulations reported here, we recycle a single year of GCM winds; no interannual variation in transport is included. The vertical resolution of the model, approximately 5-km pressure altitude in the stratosphere, is effectively increased by using the second-order moments of the tracer distributions to diagnose mixing ratios every 2 km.

Simulation and Results

Hall and Prather [1993] performed simulations of the trend and annual cycle of CO_2 using the same CTM and year of GCM winds employed here. A major goal of their work was to test the transport properties of the stratospheric GCM by comparing the CO_2 results to balloon sounding observations [*Schmidt and Khedim*, 1991], and to make predictions to be tested by lower stratospheric ER-2 flights. Carbon dioxide is an excellent tracer with which to test the model's transport properties because it is essentially inert in the middle atmosphere, and therefore does not depend on photochemical modelling. The CTM compares favorably to observations, with modeled and observed CO_2 profiles

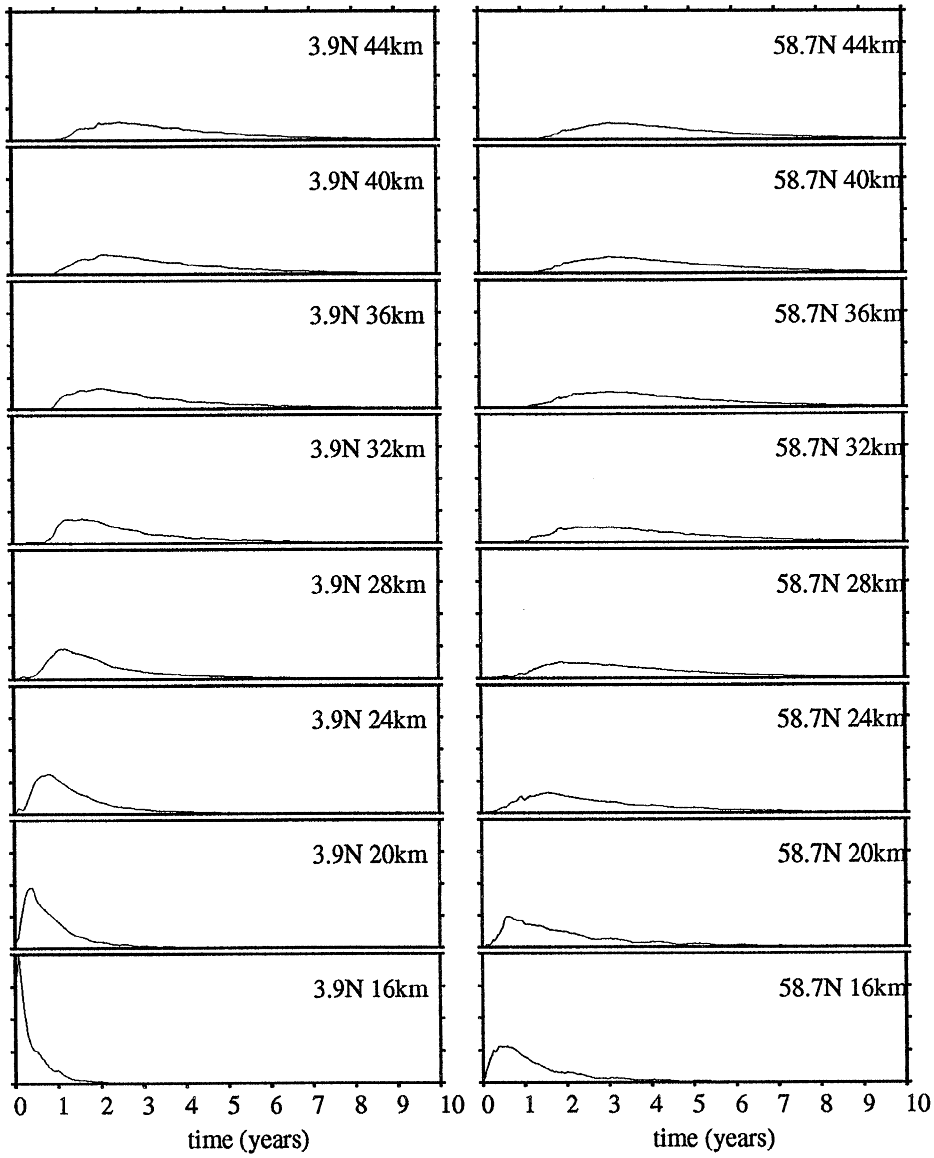

Figure 2. The zonal-mean time series response to a pulsed surface source. Pressure altitudes and latitudes are displayed as labeled, and the amplitudes have been normalized such that the peak value at 3.9°N and 16 km is unity. The time series may be interpreted an the age spectrum.

showing a difference from the troposphere to stratosphere of 4 to 6 ppm, depending on latitude. This reasonable representation of stratospheric transport encourages us to use the model as a tool to explore further the nature of this transport.

Because our goal here is to use the GCM in order to gain insight on the meaning of age as a transport time scale, we are not constrained to model a real tracer. To this end, we model the evolution and distribution of a perfectly conserved tracer with source only at the surface. We choose a time dependence for the tracer's surface boundary condition in order that the response approximate the Green's function for the transport equation solved by the model: an initial non-zero value (source) at some point at the surface, followed by a boundary condition of zero at this point.

The mechanics of our simulation are as follows. We initialize the atmosphere on January 1 to have zero tracer concentration everywhere, except for a zonal band centered on 3.9°S extending 3.9° in latitude on either side, and from the surface to approximately 1 km (the first three layers of the model atmosphere). In this band the initial concentration is finite (the magnitude, 1000 ppm, is arbitrary). Afterwards, this latitudinal band is maintained at zero. Although the source is obviously not point-like, this has no significant bearing on the stratospheric response. The zonal extent matters little because the atmospheric distribution loses zonal asymmetry rapidly with height. Further, because most tracer enters the stratosphere through the tropical tropopause, the latitudinal structure of the sources and sinks also matters little. We explore this point by comparing the responses to the 3.9°S source to an otherwise identical simulation with a source at 43.1°N.

Figure 2 displays the zonal-mean time series response of the stratosphere at altitudes of 16 km, 20 km, 24 km, 28 km, 32 km, 36 km, 40 km, and 44 km, and latitudes of 3.9°N and 58.7°N. (Throughout, we use as a vertical coordinate pressure altitude defined as $z(km) = 16 \log_{10}(1000/p)$, where p is the pressure in millibars. This way, the bottom of the model stratosphere, fixed at 100 mbar, is exactly 16 km.) The source is at 3.9°S. The time series at 16 km and 3.9°N is approximately the reponse at the tropical tropopause. Here, the signal is most peaked and least delayed from $t = 0$. This confirms the tropical tropopause as the primary entry point to the stratosphere. The pulse then spreads and attenuates as tracer propagates latitudinally and vertically. In the tropical middle stratosphere, no signal is apparent before about 1 year, and at high latitude before about 1.5 years. The spread with height of the pulse is a result of the irreversible mixing of air induced by the stratospheric circulation. In fact, the shapes of the time series are qualitatively like equation (21) shown in Figure 1, the age spectrum for the one-dimensional mass-weighted diffusive system.

The high-latitude time series in the lower stratosphere show annual fluctuations, reflecting annual variations in model transport. However, in viewing the pulse over its entire history, the seasonality is clearly not the dominant characteristic. This observation justifies our discussion of the age spectrum as the Green's function of a stationary transport process.

Zonal mean contours of tracer mixing ratio are shown in Figure 3 for the averaged time periods 0.5–1.5 years, 1.5–2.5 years, 2.5–3.5 years, and 3.5–4.5 years with the source at 3.9°S. In the first period the pulse peak has passed through the tropical tropopause. By the end of year 3, it has reached the upper stratosphere, and is significantly spread out in latitude. Meanwhile, below, the tracer depletion from the zero boundary condition has propagated through the tropical tropopause and into the stratosphere. In year 4, the vertical tracer gradient is positive throughout the model stratosphere. Also, in year 4 the tracer isopleths have approximately assumed the shape common to all long-lived tracers: a bulge in the tropics and downward slope toward either pole dictated by a balance of the Brewer-Dobson circulation

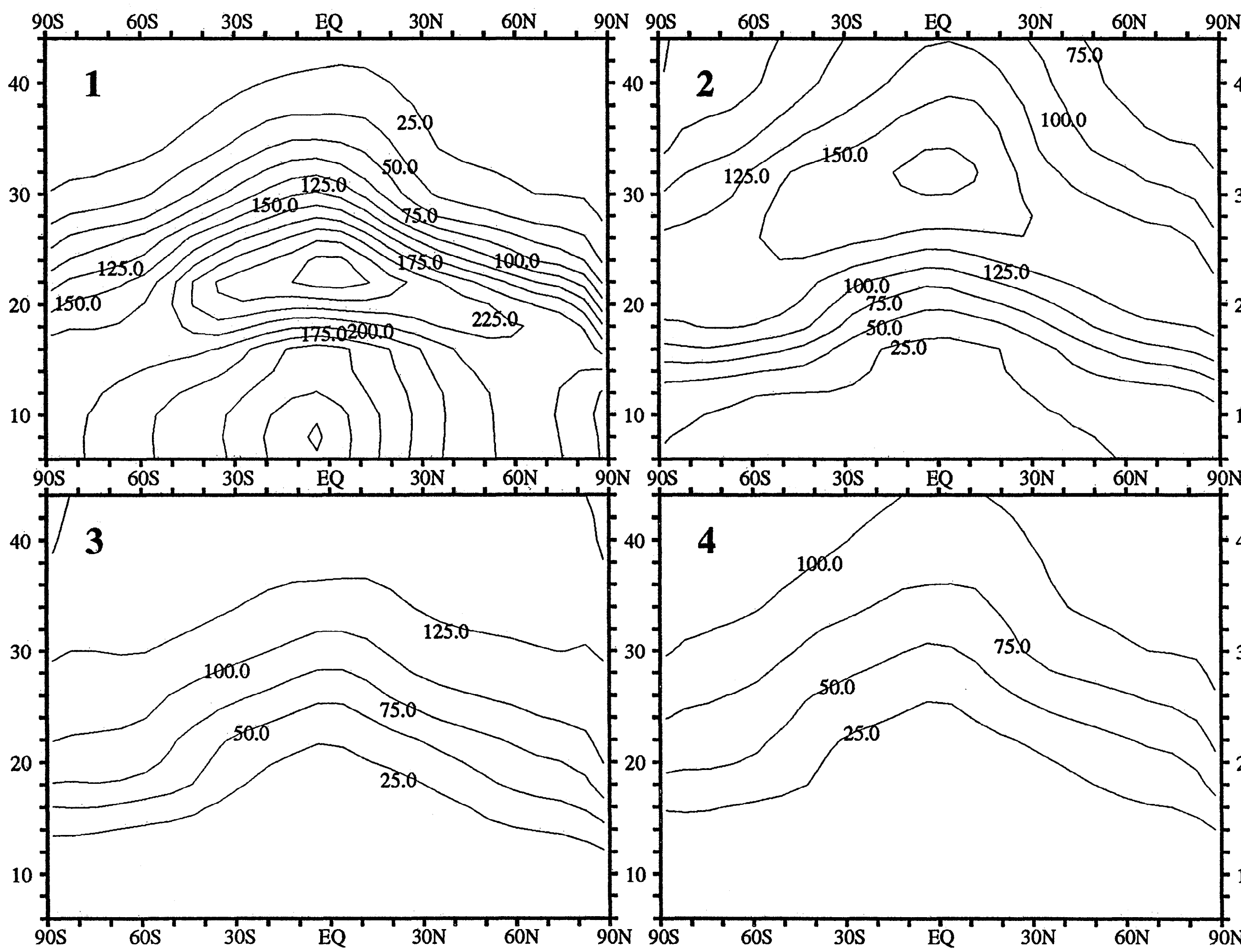

Figure 3. Zonal mean mixing ratio contour plots of the response to a pulsed source at the surface near the equator. Averaged time periods from years 0.5–1.5, 1.5–2.5, 2.5–3.5, and 3.5–4.5 are shown labeled 1 through 4. This response may be interpreted as the stratospheric age spectrum. Contours are labeled in ppm, but only relative values of the mixing ratios are significant. The vertical coordinate is pressure altitude, labeled in kilometers.

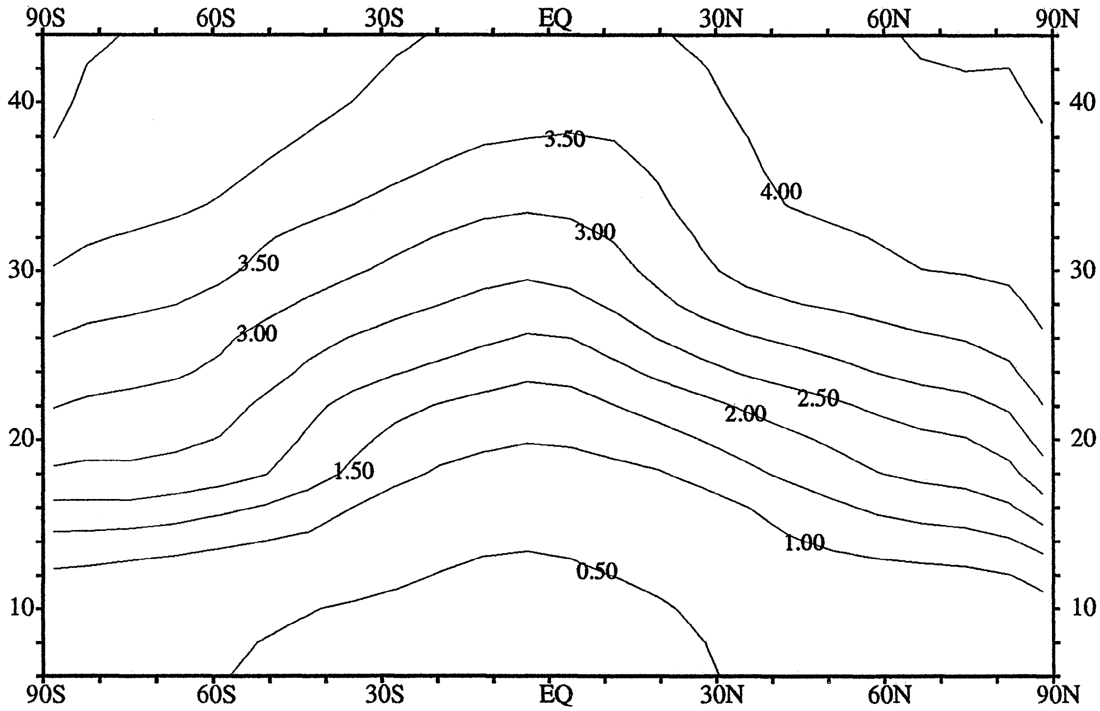

Figure 4. Contours of constant age determined by computing the first moments of the zonal mean age spectrum at each latitude and height. The source is located at 3.9°S. The contours are labeled in years, and the pressure altitudes in kilometers.

and rapid quasi-horizontal mixing. The tracer is said to be in slope equilibrium [*Holton*, 1986, *Mahlman et al.*, 1986, *Plumb and Ko*, 1992]. In particular, the isopleths approximate the shape of long-lived tracer isopleths exhibiting linear increase, and therefore, are nearly coincident with contours of constant age. This is the time at which the upper limits of the integrals in equation (7) may be taken to infinity; we are in the tail region of the age spectrum everywhere in the model atmosphere. The only subsequent evolution is a gradual diminishing of gradient, as the concentration approaches zero every where in the model atmosphere.

Contours of constant first moment of the time series are, according to our previous discussion, contours of constant age with respect to the source at 3.9°S. These are shown in Figure 4 . The shapes of the contours are nearly identical to those in Figure 2 of *Hall and Prather* [1993], a plot of age computed from the stratospheric lag in modeled CO_2 with respect to the surface. This similarity reveals the insensitivity of the stratosphere to the geographic structure of the surface boundary condition. The boundary condition of the "steady growth" simulation *Hall and Prather* [1993] was global and uniform, while the present condition is applied only at a 7.8° latitude band centered at 3.9°S. To further test the influence of boundary condition location, we plot in Figure 5 the age contours with respect to a source centered at 43.1°N. The shapes of the contours in the stratosphere are again nearly identical. Figure 6 contours the difference in age with respect to 43.1°N and 3.9°S. In the stratosphere, there is very little variation, with the values for the 43.1°N source everywhere exceeding those for the 3.9°S source by about 0.7 years, presumably the time for the pulse to spread from the surface at 43.1°N to the tropical tropopause. That stratospheric age is a minimum for the 3.9°S source is consistent with a tropical entry point of air into the stratosphere. In the troposphere, however, the age distribution is sensitive to the source, and it is only possible to speak of age with respect to the particular source, consistent with the requirement of realistic representations of surface sources and sinks for proper tropospheric tracer modeling. Such modeling is not the emphasis of our work here.

The width, as defined by (13), of the age spectrum provides further insight to the modeled stratospheric transport. In Figure 7 we contour the zonal mean of this quantity, for the source at 3.9°S. In the lower stratosphere, the model's age spectrum spreads with height. This spread continues throughout the model atmosphere in tropical latitudes. A comparison of Figure 4 and Figure 7 reveals that in the lower stratosphere, the contours of age and spectral width are nearly parallel. However, with increasing height, the shape of the age spectrum becomes less uniform on surfaces of constant age; for a given age, the spectrum is more narrow in the tropics than at middle and high latitudes. This is due to the mean upward advective transport in the tropics of the Brewer-Dobson cell. Age increases with distance from the source for both advective and diffusive transport; however, diffusive transport also causes the age spectral width to increase, unlike advective transport (an advected delta function remains a delta function). The annual mean advection of the Brewer-

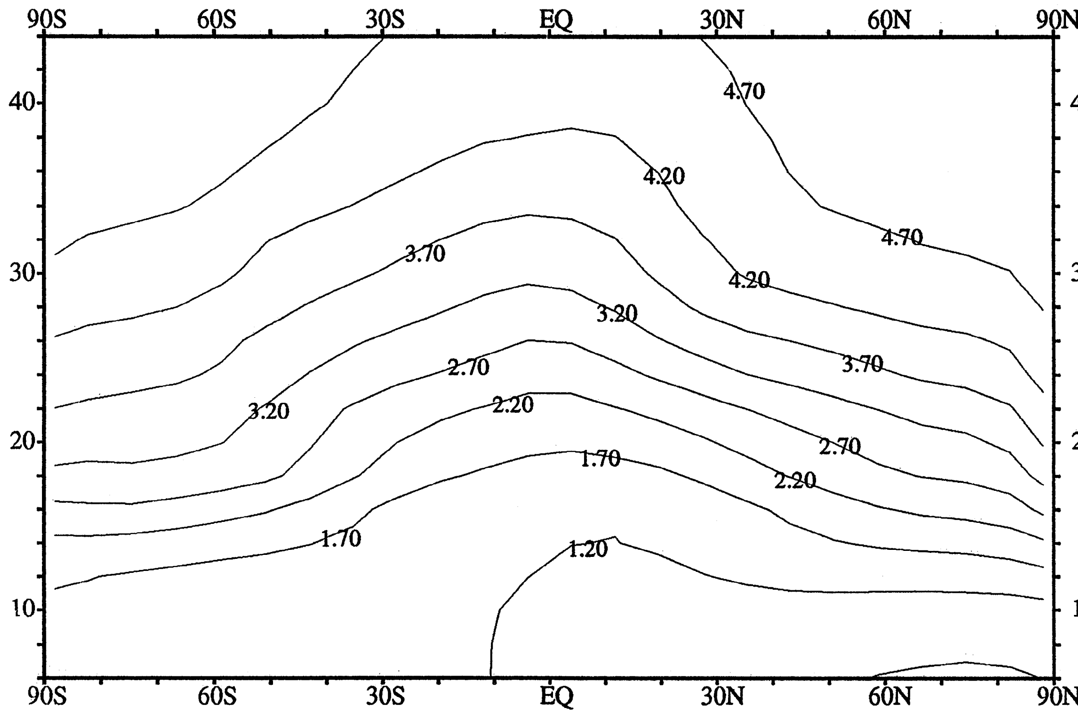

Figure 5. Identical to Figure 4, but with a source located at 43.1°N.

Dobson cell is poleward at midlatitudes and downward at high-latitudes. Consistent with this, above the lower stratosphere the age spectral width contours turn over; at high latitudes the values do not increase monotonically with height, but rather reach a maximum in the lower stratosphere.

Once we understand the propagation of a pulse into the stratosphere, we may infer the propagation of a sinusoid. Figure 8 displays contours of the zonal mean amplitude of a propagating annual cycle determined by convolving $\cos \omega t$ (where $\omega = 2\pi/1$ *year*) and the model's age spectrum for the source at 43.1°N. The amplitude is normalized to agree at the tropical tropopause with the modeled CO_2 annual cycle of *Hall and Prather*

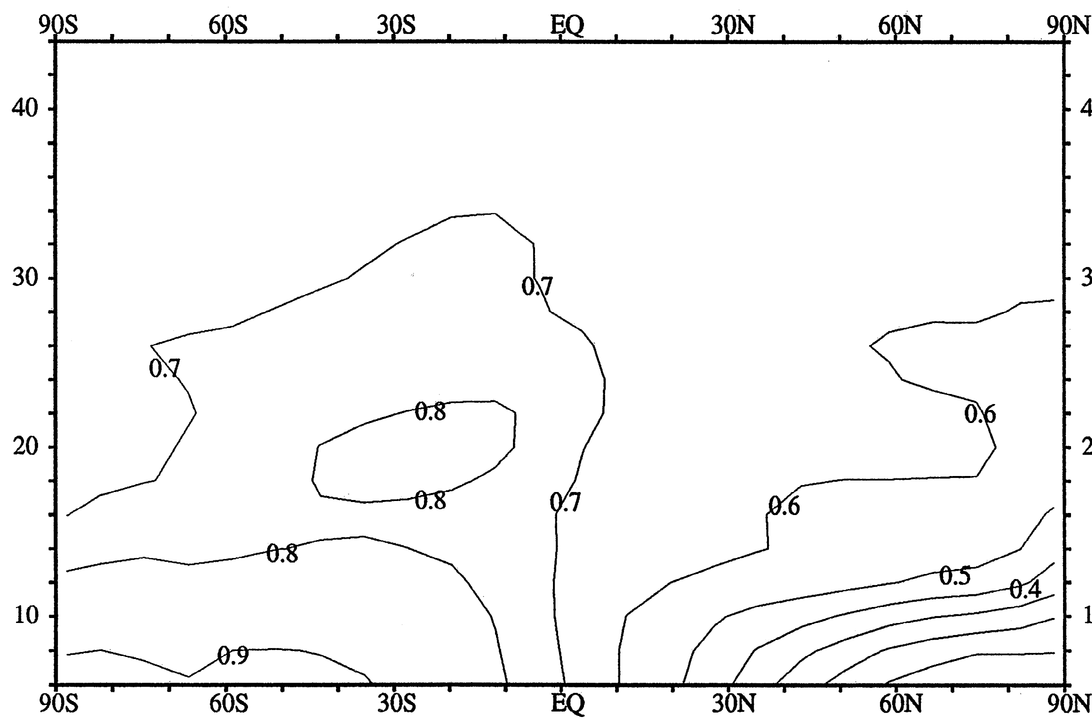

Figure 6. Contours of the age difference between a source at 43.1°N and 3.9°S. Contours are labeled in years, and pressure altitudes in kilometers.

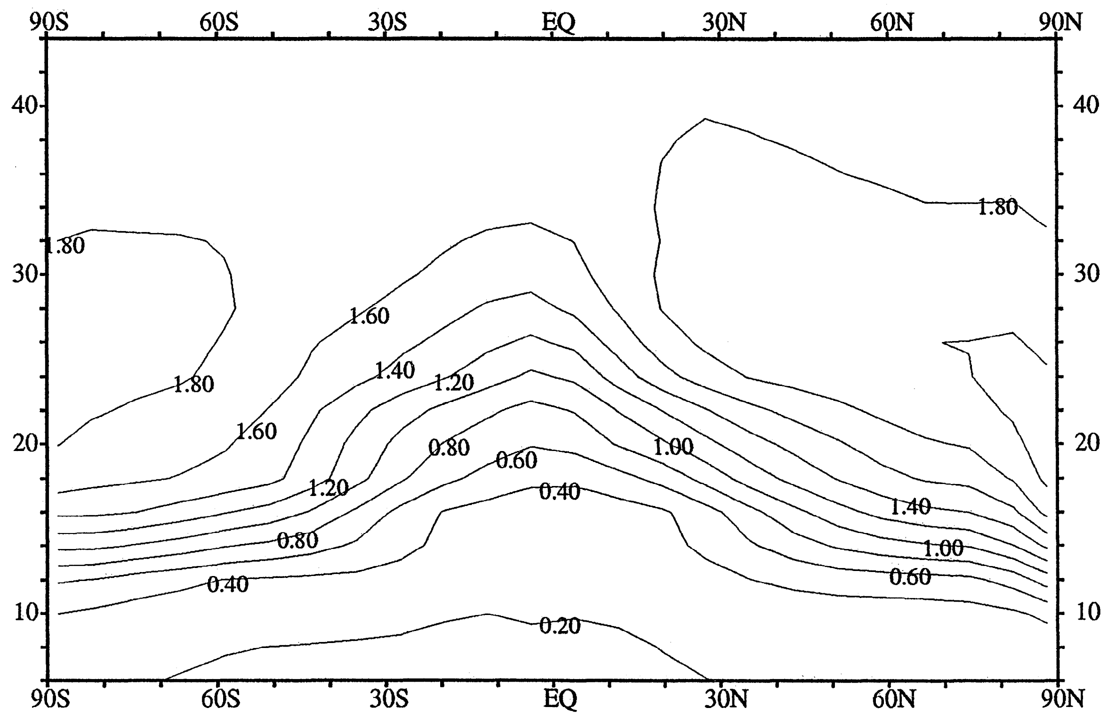

Figure 7. Contours of zonal mean width Δ of the age spectrum as defined by expression (13). The contours are labeled in years, and pressure altitudes in kilometers.

[1993] (see their Figure 4). Although the tropospheric details of their amplitude distribution differ slightly from our Figure 8, in the stratosphere they are nearly identical. The amplitude attenuates rapidly with distance from the source due to the spread of the age spectrum; with increasing spectral width, more sinusoid cycles contribute to the averaging which constitutes the reponse. The time lag in phase (not shown) increases with distance from the source, but less rapidly than age. Values of the timescale Δ^2/Γ, contoured in Figure 9 ,

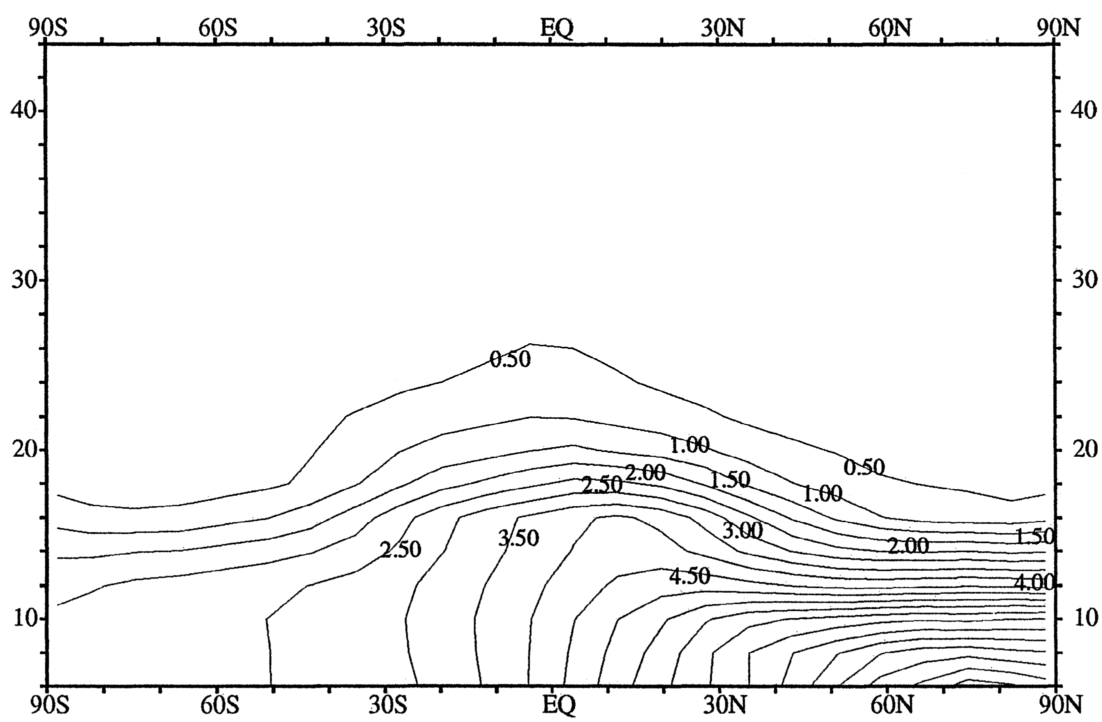

Figure 8. Contours of zonal mean peak-to-peak amplitude of a propagating signal due to an annual cycle in the surface source. The propagating response is determined by convolving the age spectrum with $\cos \omega t$ at each point in the model atmosphere. Amplitude at the tropical tropopause is normalized to the value determined by the simulation of *Hall and Prather* [1993]. The contours are labeled in ppm, and the pressure altitudes in kilometers.

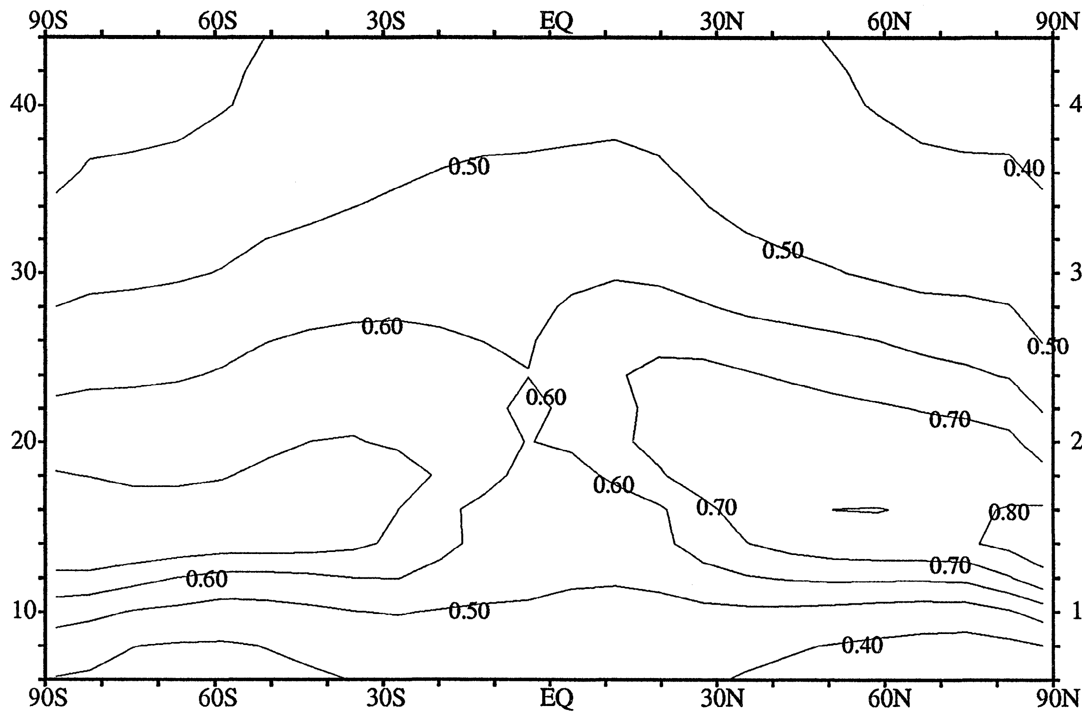

Figure 9. The ratio of Figure 4 and Figure 7. The contours are labeled in years, and the vertical coordinate is pressure altitude in kilometers.

range in the stratosphere from 0.4 to 0.8 years, so that the modeled annual cycle is not in the low-frequency limit (18).

Discussion and Summary

The age of air at a location in the stratosphere is a timescale summarizing the ability of the atmosphere to transport material to that location from the troposphere. This timescale is a statistical quantity, a mean value over a spectrum of transit times of the irreducible elements comprising an air parcel. The stratospheric time lag in mixing ratio of tracers with tropospheric origin is identical to the age if the tracer time trend is linear. In principle, only then will the distribution of tracer mixing ratio in the parcel be equivalent to the age spectrum.

Values of age are increasingly quoted for the high-latitude lower stratosphere, and used to predict concentrations of constituents, such as chlorine compounds, playing important roles in the heterogeneous chemistry responsible for high-latitude ozone destruction *Solomon,* 1990; *Pollock et al.,* 1992; L. E. Heidt et al., unpublished manuscript, 1993]. Long-lived atmospheric constituents with systematic increase in time are necessary to determine age. Thus, CO_2 is an obvious choice [*Bischof et al.*, 1985, *Schmidt and Khedim*, 1991], as is CFC-115 [*Pollock et al.*, 1992]. Note, however, that because isopleths of CO_2, or of a hypothetical, linearly growing, conservative tracer, assume the slope equilibrium configuration common to weakly nonconservative (long-lived) tracers, mixing ratios of the former will, following the arguments of *Plumb and Ko* [1992], display a compact relationship with those of the latter. Since the mixing ratio of the growing conservative species is a measure of age, it follows that such a relationship can be used to calibrate the long-lived steady species against age. Thus, species such as N_2O can be used as a measure of age. The implications of this for the modeled CO_2-N_2O relationship will be explored in a forthcoming paper (T. M. Hall and M. J. Prather, manuscript in preparation, 1993).

In the theoretical development presented above, we have restricted attention to steady (annual mean) transport. In reality, of course, seasonal fluctuations of stratospheric transport will produce corresponding fluctuations of the surfaces of constant age, just as they will for the surfaces of long-lived trace species such as N_2O. In the winter polar vortex, old air apparently descends throughout much of the depth of the stratosphere, as is demonstrated in the meridional cross section of CH_4 shown by *Tuck et al.* [1993]. (This effect is present, but underestiamted in the GCM.) Thus the age at some location will show substantial annual variation, especially in high latitudes.

Interannual variability of transport is not included in this analysis, and we are thus unable to assess its effects. It seems likely, however, that substantial and irregular variability on time scales comparable to or less than the age spectral width would seriously undermine the generality, and therefore the usefulness, of the age concept.

The GCM results have allowed us to quantify the criteria the time dependence of a tracer must satisfy in order for age to be inferred from stratospheric lag in concentration. For long-lived tracers exhibiting approximate exponential increase in tropospheric concentration, such as CFCs, age may be inferred from the lag in stratospheric concentration to the extent the exponential time constant is greater than Δ^2/Γ, as discussed above. From Figure 9, this time scale varies from 0.4 to 0.8 years in the model stratosphere. Using the general result (12), and a value $\Delta^2/\Gamma = 0.7$, we estimate a 10% difference between concentration lag time and age for a 7 year exponential growth time constant. Even the most rapidly growing CFCs are increasing more slowly than this. As an example, consider CFC-115 (CF_3CF_2Cl), which *Pollock et al.* [1992] use to estimate the age of air in the polar stratospheric vortex by observing the time lag from the tropospheric concentration. Their time series of tropospheric mixing ratio from 1980 to 1992 is fit well by $Ae^{\sigma t} + B$ with $\sigma^{-1} = 16$ *years*. We estimate that the observed time lag in CFC-115 concentration differs from the age by about 4%, a difference much smaller than the observed variability. A periodic variation in tropospheric source also propagates into the stratosphere at a rate different than that for a linear trend. Recasting (28) in terms of Δ^2/Γ and using a value 0.7 years yields a period of about 18 years for $\Gamma_\omega = 0.9\Gamma$. Periods less than this (the annual cycle and ENSO-related source variations) may therefore complicate age determinations.

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