On the Meridional Structure of Long-Lived Tropospheric Constituents

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We consider the general problem of the meridional distribution and seasonal variability of long-lived atmospheric constituents. Here “long-lived” means that the time scale of the constituent exceeds the atmospheric exchange time across the region of interest. The relevant time scale is the “dynamic lifetime,” defined here as the ratio of the total atmospheric loading of the constituent to the total global source. Provided that this quantity is large (greater than about 5 years for the troposphere), the constituent attains a “gradient equilibrium” in which spatial gradients of mixing ratio are weak and the meridional and seasonal structure take on a form which, for a given source, is independent of the dynamic lifetime. Analysis of this structure involves clarification of the nature of atmospheric exchange processes and leads us to highlight the nonuniqueness of such concepts as “interhemispheric exchange times.” These results are illustrated using a two-dimensional transport model to conduct simulations of idealized long-lived constituents with, in most cases, a simple mid-latitude northern hemisphere source and a sink with uniform decay rate. It is suggested that these results will aid the synthesis of observations of long-lived tropospheric constituents and will simplify the validation of transport models. More generally, this kind of approach might help to identify the effects of transport on trace constituent structures and thereby contribute to the understanding of the distributions of constituents which are not “long-lived” in the sense used here.

1. INTRODUCTION

The growing interest in tropospheric chemistry and, in particular, the monitoring of tropospheric trace constituents from widely separated sites around the world has increased the need for modeling studies to facilitate synthesis of the observations. While transport processes are typically more intense in the troposphere than in the stratosphere, significant meridional structure is observed for many constituents. It is therefore of some importance to enquire as to the relative importance of transport and chemistry in the determination of this structure. Amongst other things, this will help us to extract the maximum information from limited observations.

There are some gross structural characteristics, such as the reversed vertical gradient in the southern hemisphere of constituents of northern hemisphere origin, which appear to be common to many constituents. It seems clear that these properties must be of dynamical origin. To this end, we seek to identify those aspects of the global and seasonal behavior of trace constituents which may or may not be explained solely on the basis of the global and seasonal structure of transport processes. We do this by focusing attention on constituents which are “long-lived” (in a sense to be defined in the discussion that follows) and whose structure is determined solely by the effects of transport. Such constituents are uncommon (though we shall see, for example, that the chlorofluorocarbons meet the required criteria in a certain sense); however, the understanding gained by this kind of approach should tell us much about the effects of transport on other trace species.

There have been a number of modeling studies of the global and seasonal behavior of particular trace gases, using both two- and three-dimensional models. It is not in the spirit of the present study to attempt to simulate the structure of any particular, real, constituent; rather, a number of model experiments, using a two-dimensional transport model which is described in section 2, have been performed for hypothetical tracers with very simple “chemistry,” which in most cases is taken to mean a sink with a spatially and seasonally uniform rate constant. The source of these tracers in most of these experiments is confined to mid-latitudes of the northern hemisphere; however, some experiments were conducted with sources in either high northern latitudes or the tropics. We investigate the spatial and seasonal structure of these tracers and show how the spatial gradients and seasonal cycle of constituent mixing ratios depend on the “dynamic lifetime” (to be defined later) of the constituent. This allows us to make some general statements, based on simple reasoning and not restricted to the particular model used here, concerning the effects of transport processes on the spatial and seasonal structure of simple tropospheric constituents. These statements should aid the interpretation of observations of long-lived constituents and, more generally, help to identify those structural features which can be explained by transport alone and, conversely, those which appear to require other explanations.

2. FORMULATION OF THE MODEL

The model used for this study is a two-dimensional transport model representing the global atmosphere up to 10 mbar. This model was derived by Plumb and Mahlman [1987] (hereinafter referred to as PM) as a subset of the Geophysical Fluid Dynamics Laboratory (GFDL) three-dimensional general circulation/tracer model. The reader is referred to PM for details of the derivation procedure and a discussion of the properties of the zonally averaged transport processes which the model describes. The main features of the two-dimensional model are described here.

The continuity equation for the constituent mixing ratio $q(\phi, p)$ is

$$\frac{\partial q}{\partial t} = \frac{1}{a \cos \phi} \frac{\partial}{\partial \phi} \left( \frac{D_{xx}}{a \cos \phi} \frac{\partial q}{\partial \phi} + (D_{xx} + \chi) \frac{\partial q}{\partial z} \right)$$

$$+ \frac{1}{p} \frac{\partial}{\partial z} \left( \left[ \left( \frac{D_{zz} - \chi}{a \cos \phi} \right) \frac{\partial q}{\partial \phi} + D_{zz} \frac{\partial q}{\partial z} \right] \right) + S - \lambda q \quad (1)$$
The transport circulation is a simple, thermally direct circulation in the troposphere, with rising motion in the summer tropics and subsidence in middle latitudes, primarily in the winter hemisphere. The indirect "Ferrel cells," which are evident here, not only because we are mainly interested in tropospheric transport, but also because vertical diffusion (even when incorrectly represented as here) is not of primary importance in the stratosphere. The problem in the lower troposphere arose because of the breakdown of the derivation procedure at levels which are intersected by the Earth's surface. Values used in the lowest two model layers (see section 3) have been extrapolated from values above and may not be reliable. Recent work by P. Tans (private communication, 1987) on the seasonal cycle of tropospheric CO₂ using a model based on the transport coefficients used here suggests that vertical transport is too weak. Similar work by P. Hyson (private communication, 1987) has shown that the modeled cycle improves if the values of $K_{uv}$ are increased in the lowest two model layers. One further problem with the two-dimensional model, clearly evident in the comparison experiments of PM, is that the effective tropical "tracer tropopause" is too high (by about one grid increment), a problem caused by the vertical smoothing implicit in the calculation of the coefficients. While it would be straightforward in principle to remedy this deficiency, this has not been attempted for the present experiments.

Apart from the deficiencies in the two-dimensional representation of the GCM, the extent to which the model is a faithful representation of the real atmosphere is of course also limited by deficiencies of the GCM. These are noted by PM and discussed in more detail by Mahlman and Maxim [1978, and references therein]. In common with most GCMs, the model suffers from an overly cold stratospheric polar night, which is presumably an indication of an underestimate of transport processes in the winter stratosphere [see World...
This problem is potentially serious for stratospheric tracers but of lesser significance here. The most obvious shortcoming in the troposphere is that the transient eddies are somewhat weak in middle latitudes, which presumably implies that the transport in the present model is similarly weak. However, in the absence of any reliable estimates of tropospheric mixing rates, the magnitude of the error is difficult to assess.

3. Model and Experimental Details

The model solves (1) on a grid which is largely dictated by that of the parent GCM (see PM for details). The grid on which mixing ratio \( q \) is defined is regular in latitude with 76 points from pole to pole (that is, a grid increment of 2.4°) and irregular in height, the 10 vertical levels being located at \( p = 1000, 835, 685, 500, 315, 190, 110, 65, 38, \) and 10 mbar. The lower boundary is assumed to be flat, with no representation of, for example, the Antarctic plateau. The transport coefficients are defined on points midway in latitude and height (not \( p \)) between these points. The prediction equation (1) for the mixing ratio \( q \) is written in matrix form

\[
\frac{\partial}{\partial t} q = A \cdot q + S
\]

where \( A \) is the matrix of coefficients formed by taking finite differences of the transport terms on the right-hand side of (1) and adding the sink. We use an implicit scheme to integrate (2), writing it as

\[
(q^{t+\delta t} - q')/\delta t = \frac{1}{2} A \cdot (q^{t+\delta t} + q') + S
\]

whence

\[
q^{t+\delta t} = B \cdot q' + C
\]

where

\[
B = (I - 0.5\delta t A)^{-1} \cdot (I + 0.5\delta t A)
\]

and

\[
C = \delta t (I - 0.5\delta t A)^{-1} \cdot S.
\]

All experiments begin on January 1 of year 1; the initial tracer field is set to zero everywhere. The transport coefficients are updated at the start of each calendar month to the appropriate monthly values. Only surface sources are prescribed; in most cases we use a northern hemisphere mid-latitude source \( N \), setting

\[
S = S_N \sin^2 (2\phi) \quad \phi > 0
\]

\[
S = 0 \quad \phi \leq 0
\]

in the lowest layer, where \( S_N = 1.0 \times 10^{-4} \) mixing ratio units (MRU) per second, and \( S = 0 \) elsewhere. The mixing ratio units are of course arbitrary in the context of this study, unless it is desired to relate the numerical results to reality; this will be addressed later. One experiment reported here uses a high-latitude source \( M \), with

\[
S = S_M \cos^2 (3\phi) \quad \phi > 30^\circ
\]

\[
S = 0 \quad \phi \leq 30^\circ
\]

where \( S_M = 4.821 \times 10^{-4} \) MRU s\(^{-1}\). For one further experiment, we use a tropical source \( E \), for which

\[
S = S_E \cos^2 (9\phi) \quad |\phi| < 10^\circ
\]

\[
S = 0 \quad |\phi| \geq 10^\circ
\]

where \( S_E = 3.062 \times 10^{-4} \) MRU s\(^{-1}\).

The value of the constants \( S_N, S_M, \) and \( S_E \) have been chosen such that the globally integrated source is the same in all three cases, \( 3.645 \times 10^{17} \) t yr\(^{-1}\) MRU. This means, for example, that if the total global source of constituent is 364.5 Mt yr\(^{-1}\), the unit MRU should be interpreted as parts per \( 10^9 \) (by mass).

In most cases we use a global sink (GS), for which the sink \( \lambda \) is defined to be constant everywhere. For one experiment, however, we define a stratospheric sink (SS):

\[
\lambda = 0 \quad p > 65 \text{ mbar}
\]

\[
\lambda = (2 \text{ years})^{-1} \quad p = 65 \text{ mbar}
\]

\[
\lambda = (1 \text{ year})^{-1} \quad p = 38 \text{ mbar}
\]

\[
\lambda = (0.5 \text{ year})^{-1} \quad p = 10 \text{ mbar}
\]

The basic experiments described here are listed in Table 1.

4. Meridional Tracer Structure

Three experiments, \( N1, N2, \) and \( N3, \) were run with the source \( N \) given in (7), with a globally uniform sink rate \( \lambda \) such that \( \lambda^{-1} \) is 5, 0.5, and 50 years, respectively. These experiments were run for 20, 5, and 100 years, respectively, by which time the first two experiments were very close to equilibrium; the global mean mixing ratio for experiment \( N3 \) was, at 100 years, still 12% less than its equilibrium value. Monthly mean mixing ratios for January and July of year 20 for experiment \( N1 \) are shown in Figure 2. Seasonality of the mixing ratios will be assessed later, we note here that the January-to-July differences evident in Figure 2 are relatively weak, so that we may first address the spatial structure independent of seasonality. The impact of different values of \( \lambda \) on the overall structure is illustrated in Figure 3, which shows results for experiments \( N2 \) and \( N3 \) for January of the final year of these experiments. Despite some gross differences, the overall shape of the mixing ratio surfaces is very similar in the three runs, suggesting that this is controlled by transport acting on time scales shorter than the 6 months of the tracer in experiment \( N2 \). The most obvious differences are evident in the stratospheric mixing ratios, which are relatively smaller in experiment \( N1 \) and much smaller in \( N2 \) than in \( N3 \), indicating that the time scale (when defined in an appropriate way, an issue to be addressed later) for transport from the surface up to the stratosphere is comparable with 5 years and certainly longer than 6 months. In the troposphere, experiments \( N1 \) and \( N3 \) show very similar structure, but \( N2 \) has relatively much weaker mixing ratios in high southern latitudes.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Source</th>
<th>Sink</th>
<th>Duration, years</th>
</tr>
</thead>
<tbody>
<tr>
<td>N1</td>
<td>N</td>
<td>GS, ( \lambda^{-1} = 5 ) years</td>
<td>20</td>
</tr>
<tr>
<td>N2</td>
<td>N</td>
<td>GS, ( \lambda^{-1} = 0.5 ) year</td>
<td>5</td>
</tr>
<tr>
<td>N3</td>
<td>N</td>
<td>GS, ( \lambda^{-1} = 50 ) years</td>
<td>100</td>
</tr>
<tr>
<td>N4</td>
<td>N</td>
<td>SS</td>
<td>100</td>
</tr>
<tr>
<td>M1</td>
<td>M</td>
<td>GS, ( \lambda^{-1} = 50 ) years</td>
<td>100</td>
</tr>
<tr>
<td>E2</td>
<td>E</td>
<td>GS, ( \lambda^{-1} = 0.5 ) year</td>
<td>5</td>
</tr>
</tbody>
</table>

GS indicates global sink; SS indicates stratospheric sink. The sources and sinks are given in equations (7)-(9).
Jan Yr 20 NI a

Source results both from proximity to the source and from the weakness of vertical transport in the mid-latitude lower troposphere (see earlier note on this problem in the model). Both advective and diffusive processes are responsible for transport away from the source region, though with quite different consequences. As illustrated in Figure 1 (and shown in more detail in Figures 5-8 of PM), lower tropospheric diffusion occurs along sloping (upward-poleward) surfaces which in fact (as noted by PM) approximately mirror the mean isentropic surfaces. The consequence of this is that the Earth's surface forms a partial barrier to diffusive transport toward the equator, but not toward the pole. Since the transport circulation is weak poleward of the mid-latitude source, this diffusive transport dominates the tropospheric tracer structure in high lati-

Fig. 2. Monthly mean mixing ratios (MRU) for (a) January and (b) July for year 20 of experiment N1.

The common tropospheric features are as follows: (1) strong vertical gradient in the northern hemisphere lower troposphere, with very weak surface horizontal gradient northward of the source; (2) mixing ratio decreasing upward and equatorward in the northern hemisphere midtroposphere, except in the lowest layer, the tracer mixing ratio is greater at the north pole than at the latitude of the source maximum; (3) almost no vertical gradient in the tropics up to a "tracer tropopause" between 110 and 65 mbar (which is a little too high, for reasons noted earlier); (4) a reversed vertical gradient in the southern hemisphere; and (5) a very weak surface gradient south of about 30°S.

Most of these features are easily explicable in terms of the structure of tropospheric transport processes, as represented in the model. The strong vertical gradient in the region of the source results from proximity to the source and from the weakness of vertical transport in the mid-latitude lower troposphere (see earlier note on this problem in the model). Both advective and diffusive processes are responsible for transport away from the source region, though with quite different

Fig. 3. Monthly mean mixing ratios (MRU) for (a) January, year 5 of experiment N2 and (b) January, year 100 of experiment N3.
The southern hemisphere structure is a response to a virtual source and northward, almost all tracer entering the southern hemisphere does so via the upper troposphere. Thus the southern hemisphere structure of a tracer of northern hemisphere origin, however, the Hadley flow is diminished, leading to the strong mixing in the tropical upper troposphere. This appears to be the reason for the reversed vertical gradient in the southern hemisphere; it is also a factor, in addition to the moderately strong quasi-horizontal mixing, contributing to the weakness of the surface gradient in high southern latitudes.

If these statements concerning the southern hemisphere structure are correct, then this structure will be insensitive to the location of the northern hemisphere source. To illustrate this, an extreme case was chosen (experiment E2), in which the mid-latitude northern hemisphere source was replaced by a tropical source, in fact source $E$ given by (9), with a global sink of 6-month time constant (i.e., the same as experiment N2). Results for January of the fifth year of this experiment are shown in Figure 4. As expected, the shapes of the southern hemisphere mixing ratio isopleths in this experiment are very similar to those of experiment N2, although, not surprisingly, the mixing ratios there are larger than in experiment E2 when the source is closer to the southern hemisphere. Again, because the tropical transport is upward into the upper troposphere and then outward, the virtual source for the southern (and, indeed, the northern) hemisphere is elevated, and the response in both hemispheres is similar to the southern hemisphere response in experiment N2.

While it is not the point of this paper to make any detailed comparison with observations of atmospheric constituents, we note that observations confirm the concentration of surface gradients in low latitudes (for example, CH$_3$CCl$_3$; Fraser et al. [1986]) and the reversed vertical gradient in the southern hemisphere (e.g., CCl$_3$F, Fraser and Pearman [1978]; CH$_4$, Fraser et al. [1986]). However, observations of a number of constituents (e.g., CO$_2$ [Kohnmyr et al., 1985] and CH$_4$ [Fraser et al., 1986]) seem to show maximum mixing ratios at very high northern latitudes, even at the surface, a feature not reproduced in the model, though, as we have seen, it is apparent at all other lower tropospheric levels.

5. EQUILIBRIUM STRUCTURE AND TRACER LIFETIMES

Results of experiments N1, N2, and N3 appear to suggest that the shapes of mixing ratio isopleths, and even mixing ratio gradients, are to some degree reproducible for constituents with different lifetimes. In order to assess this, we consider here some general properties of the structure of long-lived tracers. The arguments are based entirely on simple considerations independent of our particular model, which is used here solely to illustrate the results.

Structure of Long-Lived and/or Slowly Growing Constituents

We begin by writing (1) in the form

\[ \frac{\partial q}{\partial t} + \mathcal{L}(q) = S - \lambda q \]  

where $\mathcal{L}(q)$ represents the transport terms. For our model, $-\mathcal{L}(q)$ specifically represents the first two terms on the right-hand side of (1); however, for the purposes of the following arguments we need only specify the following properties: first, $\mathcal{L}$ is a linear operator; second, $\mathcal{L}(Q) = 0$, if $Q$ is independent of position, and third, $\langle \mathcal{L}(q) \rangle = 0$, where the angle brackets denote the global average:

\[ \langle X \rangle = (2Hp_a)^{-1} \int_{-\pi/2}^{\pi/2} pX \cos \phi \, d\phi \, dz \]  

where $H_p$ and $a$ are the tropospheric height, being large in the lower troposphere but weak in the upper troposphere. Again, the reason is a simple one. In the lower troposphere, where horizontal diffusion is relatively weak, the gradients are concentrated by the confluence of the Hadley cell inflow; this maximum gradient follows the annual migration of the tropical convergence. In the upper troposphere, however, the Hadley flow is diffusive, leading to weakening of the horizontal gradient; this is compounded by the strong mixing in the tropical upper troposphere.

These properties of tropical transport are crucial in fixing the southern hemisphere structure of a tracer of northern hemisphere origin. Because the lower tropospheric transport to the south of the Hadley cell confluence is primarily advective and northward, almost all tracer entering the southern hemisphere does so via the upper troposphere. Thus the southern hemisphere structure is a response to a virtual source in the tropical upper troposphere.
Clearly, these three properties are satisfied by the transport operator in (1); in fact, these criteria would be satisfied by any conceivable operator representing material exchange, so the comments to follow which are based on (11) are restricted neither to our particular model, nor to the "K-theory" description of atmospheric transport nor, indeed, to a two-dimensional formulation.

The effect of transport is, of course, to tend to homogenize the constituent mixing ratio. In the absence of other processes, transport would destroy the mixing ratio gradients on some time scale which characterizes the intensity of the transport in the region of interest (which may be the entire atmosphere). In the presence of internal atmospheric sinks, nonzero gradients may be sustained; however, if the characteristic time scale $\lambda^{-1}$ of the sink is much longer than the relevant transport time scale, we anticipate that in steady state, the constituent will be well-mixed, so that $q \approx \langle q \rangle$ everywhere. We adopt this criterion as our definition of "long-lived" constituents; how the criterion is quantified will become apparent in what follows.

We first consider the steady state. Taking the global average of (11), given the third property above, we have $\langle \lambda q \rangle = \langle S \rangle$, or, since $q \approx \langle q \rangle$,

$$\langle q \rangle \approx \langle S \rangle / \langle \lambda \rangle \quad (13)$$

Then (11) may be written as

$$\mathcal{L}(q) = S - \frac{\lambda}{\langle \lambda \rangle} \langle S \rangle \quad (14)$$

at equilibrium for small $\lambda$. Since, by the second property, $\mathcal{L}(q)$ and the boundary conditions are functions of the gradients of $q$ but not of $\langle q \rangle$, it follows that the gradients depend on the nature and strength of the source and on the structure but not the magnitude of the sink rate $\lambda$, provided that the latter is sufficiently weak for the approximation of uniform $q$ to be valid. The gradient of tracer mixing ratio therefore remains finite as $\lambda \to 0$; this simply reflects the constraint that the sink $\lambda q$ must be finite in order to balance the source, and therefore the transport from source to sink must be finite. However, from (13), the magnitude of $\langle q \rangle$ varies as $\langle \lambda \rangle^{-1}$, so that the fractional gradients, $\nabla q / \langle q \rangle$, will vary as $\langle \lambda \rangle$ and therefore vanish as $\langle \lambda \rangle \to 0$; we shall return to this in section 6.

These results for weakly nonconserved tracers in steady state can readily be extended to tracers with slowly growing mixing ratios, which is the case for many pollutants of industrial origin. The global average of (11) gives

$$\frac{\partial}{\partial t} \langle q \rangle = \langle S \rangle - \langle \lambda q \rangle \quad (15)$$

If the time scale of growth is much longer than the time scale for transport to destroy gradients, then we again anticipate that $q$ will be approximately uniform. Further, if $\langle q \rangle(t)$ is growing at a rate $\gamma(t)$, such that

$$\frac{\partial}{\partial t} \langle q \rangle = \gamma \langle q \rangle \quad (16)$$

then

$$(\langle \lambda \rangle + \gamma) \langle q \rangle = \langle S \rangle \quad (17)$$

Note that (16) does not require the source to be growing in time also, though this is undoubtedly true for many atmospheric constituents of interest. In all experiments described here, the source is constant, but $\langle q \rangle$ grows in time as the system evolves toward equilibrium. Indeed, for $\langle q(t=0) \rangle = 0$, as in these experiments,

$$\langle q \rangle = \frac{S}{\lambda} [1 - e^{-\lambda t}]$$

whence

$$\gamma(t) = \lambda e^{-\lambda t} / [1 - e^{-\lambda t}]$$

We may use (17) to define a "dynamic lifetime" $\tau = (\langle \lambda \rangle + \gamma)^{-1}$:

$$\tau = \langle q \rangle / \langle S \rangle \quad (18)$$

In general, since if $M$ is the mass of the atmosphere, $M \langle q \rangle$ is the total atmospheric loading, and $M \langle S \rangle$ the total source, $\tau$ is just equal to the ratio of these two quantities. This contrasts with the conventional definition of atmospheric lifetime as $M \langle q \rangle / (\text{total sink})$, which is just $\langle \lambda \rangle^{-1}$ under present assumptions. For a steady tracer, $\tau = \langle \lambda \rangle^{-1}$, but the two are not the same for a time-dependent tracer; if the tracer mixing ratio is growing on a time scale much shorter than $\langle \lambda \rangle^{-1}$, $\tau$ is just the growth time.

For a weakly growing constituent, then, (1) may be approximated

$$\mathcal{L}(q) = S - \left[ \frac{\lambda + \gamma}{\langle \lambda \rangle + \gamma} \right] \langle S \rangle \quad (19)$$

Thus (19) is identical to the steady result (14) with $\lambda$ replaced by $(\lambda + \gamma)$. If therefore either: (1) the sink rate is spatially uniform, or (2) $\lambda \ll \gamma$, (19) simply reduces to

$$\mathcal{L}(q) = S - \langle S \rangle \quad (20)$$

implying that the mixing ratio gradients are independent of both $\lambda$ and $\gamma$, and therefore of $\tau$, even when the constituent mixing ratio is not steady.

These predictions receive strong confirmation from the experiments described here. Figure 5 shows the evolution of $\langle q \rangle$ and $\tau$ (annually averaged values) against time for the 100-year duration of experiment N3. Since the source is independent of time and $\lambda$ is uniform, (20) predicts that apart from an increasing global mean, the structure of $q$ will become independent of $\tau$ once $\tau$ becomes sufficiently large. This is shown in Figure 6.
in which the quantity \( q - \langle q \rangle \) is contoured for January of year 25 (when \( \tau = 20.1 \) year) and year 100 (when \( \tau = 44.2 \) year). Despite the differing values of \( \tau \), Figures 6a and 6b are indistinguishable, and it seems clear that they are showing the long-dynamic-lifetime limit structure of \( q - \langle q \rangle \) for this source. This quantity is also shown in Figure 6c for the final January of experiment N1, at which time \( \tau = 5.0 \) years. At this value of \( \tau \), some differences from the large \( \tau \) limit become apparent. In the troposphere, however, the differences are small (typically 2% or less); more substantial differences are to be found in the stratosphere, where the vertical gradients for \( \tau = 5 \) years are only a little over one half of these for large \( \tau \). For the very short lifetime case, experiment N2, \( q - \langle q \rangle \) is quite different from the structures shown in Figures 6a and 6b, even in the troposphere (not shown here, but this statement is obvious from a comparison of Figures 3a and 3b). The significance of these comparisons will be discussed later.

**Exchange Times**

Consider now the full, three-dimensional transport of tracer from a point source \( S_0 \) to the location \( P \) at \((\varphi, p)\). We focus attention on a microscopic element of air, small enough that we may assume that it maintains its integrity during transit. Since, following each such element,

\[
\frac{dq}{dt} = -\lambda q
\]

we know that

\[
q(\varphi, p, t) = q_0(t - 2\pi) e^{-2\pi} \tag{22}
\]

where \( \beta(\varphi, p) \) is the time taken for that element to travel from the source (where its mixing ratio at time of departure was \( q_0(t - 2\pi) \)) to \( P \). Since \( q_0 \) is growing as \( e^{2\pi} \),

\[
q(\varphi, p, t) = q_0(t)e^{-2\pi} \tag{23}
\]

Now, any macroscopic parcel \( \Phi \) of air at \( P \) consists of many such elements, which will, in general, have taken different paths from source to \( P \). Let us suppose that a fraction \( \mu \) of the air within \( \Phi \) has a transit time to \( P \) of less than \( T(\mu) \). Then within \( \Phi \), the average mixing ratio is

\[
q = q_0 \int_0^1 e^{-T(\mu)t} d\mu \tag{24}
\]

We now define the exchange time, \( \Gamma \), from source to sink, such that

\[
q(\varphi, p, t) = q_0(t - \Gamma) e^{-\Gamma t} \tag{25}
\]

which means that the mixing ratio at \((\varphi, p)\) is the same as if the entire air parcel had traveled intact to \( P \) in a time \( \Gamma \). Then from (24),

\[
\Gamma = -\tau \ln \int_0^1 e^{-T(\mu)t} d\mu \tag{26}
\]

Now, for long-lived tracers, \( T \ll \tau \) for all significant paths. (\( T \) will, in fact, be very large for some paths, for example, via the upper atmosphere, but these will make a negligible contri-
TABLE 2. Conversion From $q$ to Exchange Time $\Gamma$ for Experiment N3 (Figure 3b)

<table>
<thead>
<tr>
<th>$q$, MRU</th>
<th>$\Gamma$, years</th>
</tr>
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<tbody>
<tr>
<td>3280</td>
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<tr>
<td>3240</td>
<td>0.94</td>
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<tr>
<td>3200</td>
<td>1.49</td>
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<td>3040</td>
<td>3.76</td>
</tr>
<tr>
<td>3000</td>
<td>4.35</td>
</tr>
<tr>
<td>2960</td>
<td>4.94</td>
</tr>
</tbody>
</table>

MRU is mixing ratio units.

The prediction of this particular two-dimensional model therefore is that a long-lived tropospheric constituent is one whose dynamic lifetime is longer than about 5 years; in the middle stratosphere, $\tau$ must exceed about 10 years.

6. Isopleth Slopes

Results of the short-lifetime experiment N2 ($\tau = 0.5$ year) do not fit these earlier predictions; nevertheless, it is apparent from Figures 2 and 3 that while the mixing ratio gradients differ in N2 from those of the longer-lived tracers of N1 and N3, the shapes of the mixing ratio isopleths remain essentially the same in the three experiments. This brings to mind the “equilibrium slopes” hypothesis of Mahlman et al. [1986] and Holton [1968]; they argued that the mixing ratio isopleths of sufficiently long-lived stratospheric tracers will possess characteristic slopes which are determined by a balance between the slope-steepening effects of mean advection and the slope-flattening effects of quasi-isentropic diffusion. It appears that in some ways, a similar balance may hold throughout much of the atmosphere, including the troposphere, in the present results.

Some of the assumptions made by Mahlman et al. and by Holton are not applicable in our case, and so we briefly consider here the more general balance of isopleth slopes. We define the tracer isopleth slope as

$$\sigma = \left[ \frac{\partial q}{\partial y} \right] / \left[ \frac{\partial q}{\partial x} \right]$$

where, as before, $y = a \sin \phi$. It is shown in Appendix B that, from (1), the slope budget equation may be written

$$\frac{\partial \sigma}{\partial t} = E_{\text{ADV}} + E_{\text{DIFF}} + E_{\text{SOURCE}} + E_{\text{SINK}}$$

where the terms on the right-hand side represent the contributions from diffusive transport, advective transport, source, and sink terms, respectively. (These contributions are derived in equations (B5)-(B8) of Appendix B).

In general, the sink term becomes independent of $\langle \lambda \rangle$ as $\langle \lambda \rangle \to 0$. However, if $\lambda$ is spatially uniform or, if not, locally small, this term makes no contribution to the slope budget. Therefore if this is so, in regions away from the source the
equilibrium isopleth slopes are determined purely by a local balance between advective and diffusive transport processes, in agreement with the arguments of Mahlman et al. [1986] and Holton [1986]. This does not necessarily mean in general that $\sigma$ is determined by purely local characteristics, however, since both the advective and diffusive effects involve spatial derivatives of $\sigma$.

Under certain circumstances, however, the steady balance of (32) does yield simple local solutions for $\sigma$. Suppose, for example, that the diffusion occurs solely in the $y$ direction with spatially constant coefficient $D_{yy}$. If diffusion occurs solely in a nonhorizontal plane, we may rotate coordinates so that the $y$ axis coincides with the axis of diffusion. If, further, $|\chi| \ll D_{yy}$ it is shown in Appendix B that balance may be achieved in (32) with a small slope

$$\sigma \approx -\chi/D_{yy} \tag{33}$$

This is (in a slightly different form) the relation obtained by Mahlman et al. [1986] and Holton [1986] as representing the balance between the slope-steepening effects of advection and the flattening effects of quasi-horizontal diffusion. We have made the assumption here that the diffusion is uniplanar (though not necessarily isentropic) and $\sigma$ in (33) should be interpreted as the slope relative to the principal axis of diffusion. (The principal axes of diffusion for this model for each season are shown in Figures 5d–8d of PM.) It should be noted that the existence of this simple solution apparently depends on the assumptions of uniplanar diffusion and weak advection. There may, in general, be no purely local solution for $\sigma$. Moreover, even given these assumptions, there is no guarantee that the solution (33) is unique.

The assumptions are, in fact, reasonably well satisfied in mid-latitudes of the model, the diffusion being mostly directed along a quasi-horizontal (in fact quasi-isentropic) principal axis, and advection being weak outside the tropical Hadley cell (cf. Figures 5–8 of PM). Equilibrium slopes calculated for January from (33) are shown in Figure 7. Comparing these slopes with the January mixing ratio isopleths from the results of the experiments with the mid-latitude northern hemisphere source (Figures 2a and 3), it is evident that the equilibrium slope has been attained in high northern latitudes throughout the troposphere and stratosphere, around the tropical tropopause, and in the upper troposphere and stratosphere of the southern hemisphere. The most obvious discrepancies are in the subtropical northern hemisphere troposphere and the southern hemisphere lower troposphere. Considering the equilibrium slopes in these regions in Figure 7, it is perhaps not surprising, given the overall requirement of a flux of constituent directed from source to sink, that these discrepancies occur. In the southern hemisphere lower troposphere, the boundary condition of zero flux through the Earth’s surface is inconsistent with the almost-horizontal slopes of Figure 7, although, on the basis of the northern hemisphere results in these experiments, it seems likely that these slopes would be achieved for a mid-latitude southern hemisphere source. In the northern hemisphere subtropics, it is also difficult to see how the steep equilibrium slope of Figure 7 could be attained by a constituent with the source used in these experiments. It is interesting to note that the mixing ratio isopleths for experiment E2 (Figure 3a) in this region do match the equilibrium slopes quite well, so it seems unlikely that the discrepancy is simply an indication of the breakdown of the assumptions involved in the derivation of $\sigma$. This result suggests either that the equilibrium slope is attained only when the overall constraints of the source/sink structure allow, or that there exist equilibrium solutions other than those obtained here, and the selection of the appropriate solution in any particular case is determined by such constraints.

7. INTERHEMISPHERIC GRADIENTS

We have seen that for tracers with small, uniform $\tau$, the gradients of $q$ depend only on the structure and intensity of the source. In fact, from (29) we obtain

$$\nabla q = -\nabla \nabla \tau$$

for uniform $\tau$, where, recall, $\Gamma(\phi, \rho)$ is the exchange time appropriate to the source in question. As in (28) and (29), we obtain a relation for the fractional gradient of mixing ratio

$$\nabla q/\langle q \rangle = -\nabla \tau/\tau$$

which, while independent of the source strength, is (through $\Gamma$) a function of its structure. In regions sufficiently remote from the source, however (and what “sufficiently remote” means will need to be assessed case by case), $\nabla \Gamma$ should be relatively insensitive to the details of the source structure, and so the fractional atmospheric mixing ratio gradients of long-lived constituents in such regions will depend solely on the dynamic lifetime $\tau$.

In order first to illustrate the dependence of gradients on $\tau$ for a given source, we concentrate on results from experiment N3. The evolution of the annually averaged, global-mean mixing ratio for this run was shown in Figure 5. The value of $\langle q \rangle$ increases rapidly in the early years, reaching a value of about 3200 MRU after 100 years; since $\langle S \rangle = 2.20 \times 10^{-6}$ MRU s⁻¹, the value of $\tau$ in years is, from (18), simply 0.0138$\langle q \rangle$. At the end of the 100-year integration, $\tau$ has the value 44.2 years, still some 12% below its steady value.

![Fig. 7. Equilibrium slopes for January determined from equation (33). See text for discussion.](image-url)
As a measure of the fractional gradient between conjugate latitudes at the same level, we adopt the quantity

$$\Delta(\varphi, p) = \frac{q(\varphi, p) - q(-\varphi, p)}{q(\varphi, p) + q(-\varphi, p)}$$

(36)

**Fig. 8.** Fractional gradient $\Delta(30^\circ, p)$ at $p$ equal to (a) 190 mbar, (b) 500 mbar, and (c) 1000 mbar for each year of experiment N3 (solid curve) and for experiment N1 after 20 years (crosses), plotted against $\tau^{-1}$, the inverse of the dynamic time scale.

**TABLE 3.** Differential Exchange Times for Experiment N3

<table>
<thead>
<tr>
<th>Latitude, $\varphi$, deg</th>
<th>$p = 190$ mbar</th>
<th>$p = 500$ mbar</th>
<th>$p = 1000$ mbar</th>
</tr>
</thead>
<tbody>
<tr>
<td>30</td>
<td>0.38</td>
<td>0.73</td>
<td>1.92</td>
</tr>
<tr>
<td>60</td>
<td>0.65</td>
<td>1.30</td>
<td>2.27</td>
</tr>
</tbody>
</table>

Gross interhemispheric differential exchange time, $\Delta T$, is equal to 0.73 year; $p$ equals pressure.

where the mixing ratios used are annually averaged values. (We could equally well choose nonconjugate latitudes for the two points, for example, the latitudes of two monitoring sites, which of course may be more useful in practice). From (35) we anticipate (since $q(\varphi, p) + q(-\varphi, p) = 2q(\varphi)$)

$$\Delta(\varphi, p) \simeq \Delta \Gamma(\varphi, p) \tau^{-1}$$

(37)

where $\Delta \Gamma(\varphi, p) = \Gamma(-\varphi, p) - \Gamma(\varphi, p)$ is the differential exchange time between the two locations for the source in question. What this means, of course, is the difference between the local values of exchange times from the source and not the time for transport between the two locations.

The dependence of $\Delta$ on $\tau^{-1}$ through the 100 years of this run is shown in Figure 8 for $\varphi = 30^\circ$ in the upper, middle, and lower troposphere. In the middle and upper troposphere the dependence of $\Delta$ on $\tau^{-1}$ is very close to linear, except for short dynamic lifetimes ($\tau < 3$ years), in agreement with the preceding arguments. A linear fit to $\Delta(\tau^{-1})$ is less good at the surface, except for longer lifetimes ($\tau > 10$ years), perhaps because the local source is nonzero at the surface at $30^\circ N$.

Values of $\Delta \Gamma$ obtained from this experiment by taking the gradient of the linear part of the curves (that is, for the larger values of $\tau$) for $\varphi = 30^\circ$ shown in Figure 8 and others for $\varphi = 60^\circ$ (not shown) are listed in Table 3. Also listed in Table 3 is the gross interhemispheric differential exchange time

$$\Delta \Gamma = (q_N - q_S)/\langle S \rangle$$

(38)

where $q_N$, $q_S$ are the hemispheric average mixing ratios for the northern and southern hemisphere, respectively.

This experiment was chosen as a means of illustrating the dependence of $\Delta$ on $\tau$ because of the wide range of dynamic lifetimes obtaining during the course of the experiment. However, the arguments presented at the beginning of this section predict that other tracers of spatially uniform $\tau$ and with the same source should exhibit the same values of $\Delta$ for the same $\tau$, independent of whether $\tau$ is dominated by the chemical sink or by temporal growth. This is supported by results from experiment N1. Values of $\Delta$ from the almost steady mixing ratios at 20 years for this experiment are plotted as crosses on Figure 8; as expected, they lie almost exactly on the curves obtained from experiment N3 (and for which, at this value, $\tau$ is dominated almost entirely by the temporal growth term $y$).

The sensitivity of these results was tested by determining $\Delta \Gamma$ from experiment M3, which (see Table 1) is identical to N3 in all respects except that the mid-latitude source $N$ was replaced by the high-latitude source $M$. The meridional structure of the mixing ratio isopleths from this experiment (not shown here) was very similar to that of N3 at the same time, except, not surprisingly, in the lower and middle troposphere at middle and high latitudes of the northern hemisphere (that is, in the region of the sources). The differential exchange times $\Delta \Gamma(\varphi, p)$ for experiment M3 are given in Table 4. In the upper tropo-
sphere for \( \phi = 60^\circ \) and at all tropospheric levels for \( \phi = 30^\circ \), the values found are quite similar to (within 10–15%) those for experiment N3 given in Table 3. In the middle and (especially) lower troposphere at \( \phi = 60^\circ \), however, \( \Delta \Gamma \) is much larger in M3; it appears that this is simply a reflection of the impact of the shift to 60°N of the source maximum. Overall, this comparison confirms the naive expectation that gradients in regions south of the source are not strongly dependent on the source structure. Nevertheless, the nonuniqueness of concepts such as interhemispheric exchange times is highlighted by the significantly differently values of \( \Delta \Gamma \) for the two runs.

8. Seasonal Variations

Thus far, almost all comments have been directed at the annual mean tracer structure; seasonal variability of the transport coefficients has been neglected, though in places this is quite substantial (see Figures 5–8 of PM). Nevertheless, as is evident from a comparison of Figure 1a and Figure 1b, seasonality of the tracer mixing ratio for experiment N1 is in fact quite small. If we define by an overbar the annual average and by a prime the seasonally varying component, we may write the transport coefficients as

\[
K(\phi, p, \theta) = \bar{K}(\phi, p) + \Delta K(\phi, p, \theta)
\]

where \( \theta \) is time of year, that is, time measured from, say, January 1 of the current year (there being no interannual variability in the transport coefficients). Similarly, we may separate the operator \( \mathcal{L} \) into time-independent and seasonally varying components, \( \mathcal{L}' \) and \( \mathcal{L}'' \), respectively, so that

\[
\mathcal{L}'(q) = \mathcal{L}'(q' + \bar{q}) + \mathcal{L}''(q' + \bar{q})
\]

Given that seasonal variability in \( q'(\phi, p, \theta) \) is small, we ignore \( \mathcal{L}''(q') \) and write the seasonally varying component of (1) as

\[
\frac{\partial q'}{\partial t} = \mathcal{L}'(q') + \lambda q' = \mathcal{L}''(q') = \mathcal{L}''(\bar{q} - \langle q' \rangle)
\]

where there is no explicit contribution from the source term, since, for the purposes of the present experiments, the latter is assumed constant in time. For real trace constituents, of course, there may be contributions to seasonality from variations in both sources and sinks; here we address solely those aspects of seasonal variability which can be attributed to seasonality in the atmospheric transport alone. The relation (40) expresses the fact that transport-induced seasonal variability arises solely from the effects of seasonally varying transport acting on mean gradients of the tracer mixing ratio.

Now, if \( \tau \) is much longer than 1 year (and it must be to satisfy our criterion of "long-lived") then the sink term in (40) is negligible. Moreover, although \( \langle q' \rangle \) may vary from year to year, \( q' - \langle q' \rangle \) does not (since its annual mean is independent of \( \tau \)) and the term on the right of (40) depends on time only as \( \theta \). Therefore \( q' = q'(\phi, p, \theta) \) and (40) may be written

\[
\frac{\partial q'}{\partial t} = \mathcal{L}'(q') = \mathcal{L}''(\bar{q} - \langle q' \rangle)
\]

Since, from (34), \( \bar{q} - \langle q' \rangle \) is purely a function of \( \Gamma \) and \( \langle S \rangle \), it follows that

\[
q'(\phi, p, \theta) = \Omega(\phi, p, \theta) \langle S \rangle
\]

where \( \Omega(\phi, p, \theta) \) is a function of \( \Gamma(p, \phi) \), \( \bar{K}(\phi, p) \) and \( K'(\phi, p, \theta) \). Therefore the absolute seasonal cycle is, in both magnitude and \( \theta \) dependence, source-dependent but, in the large \( \Gamma \) limit, independent of \( \tau \); this simply reflects the same characteristic of the mixing ratio gradients. However, like the fractional gradients, the fractional seasonality is independent of \( \langle S \rangle \) and inversely proportional to \( \tau \):

\[
\frac{q'}{\bar{q}} = \frac{\Omega}{\tau}
\]

Like the fractional gradients, it is anticipated that sufficiently far from the source, the fractional seasonality will be insensitive to details of source structure. Note that this result is in some ways a special case of the relation given by Junge [1974] for the temporal variability of tracer mixing ratios. Junge was, in fact, considering synoptic, rather than seasonal, variations, but the principle (variability induced by fluctuating transport acting on background gradients) is the same.

Detrended time series of mixing ratio for a selection of locations during the last 5 years of experiment N3 are shown in Figure 9. Before assessing the validity of the predictions made earlier regarding the dependence of seasonal variability on \( \langle S \rangle \) and \( \tau \), we describe briefly the gross characteristics of the seasonal cycle produced in the model.

The magnitude of the seasonal variability for experiment N3 is in fact quite small. The greatest seasonal variability occurs in the tropical lower troposphere and in the mid-latitude middle troposphere and high-latitude upper troposphere of the northern hemisphere, with minimum variability in mid-latitudes of the southern hemisphere. Even at the equator, the annual range at the surface is less than 1% for year 100 of this experiment (see Figure 9 and note from Figure 5 that \( q \approx 3200 \) MRU at this time); this compares with a fractional surface gradient between 30°N and 30°S (Figure 9c) of about 4%. In the low levels of the tropics, maximum mixing ratios occur in late southern summer (see Figure 9) as the low-level flow in the Hadley cell brings tracer-rich northern hemisphere air southward toward a convergence at about 15°S (see Figure 5b of PM), south of which the amplitude of the seasonal cycle falls off markedly. In the northern hemisphere middle troposphere, maximum values occur in northern summer, when, as noted in section 3, upward transport from the source is at its most efficient (the circulation being upward there at this time of year). The high-latitude seasonality in the upper troposphere (not shown) is dominated by a sharp minimum in the upper troposphere in June and July, which appears to correspond to a slowing of the effective tropopause; the reality of this feature is uncertain.

It is evident from Figure 9 that even though in most locations shown here the annual cycle appears to be the dominant component, the seasonal cycle cannot be described solely by a single harmonic. Moreover, in southern hemisphere mid-latitudes a significant semiannual cycle is evident, which appears to be indicative of the strong semiannual component in cross-equatorial transport; there are suggestions of this feature in observations [Fraser et al., 1986].

Turning now to the verification of (42) and (43) in these experiments, time series (not shown here) of \( q' \) for years 25, 50,
and 75 of experiment N3 (when $\tau = 20.1, 32.2, \text{and} 39.8$ years, respectively) were compared with those shown in Figure 9 for year 100 (when $\tau = 44.2$ years) and found to be indistinguishable. Time series for the last 5 years of experiment N1 ($\tau \approx 5$ years) are shown in Figure 10; while there are some small differences between these time series and those shown in Figure 9, the seasonality for this run at these tropospheric locations is almost identical to that found in experiment N3. This is consistent with our previous comments on the close similarity of tropospheric structure in experiments N1 and N3 and confirms that the seasonal variations shown in Figure 9 represent the long-dynamic-lifetime limit for this model.

Some idea of the sensitivity of seasonal variability to small changes in source structure can be gleaned from a comparison of Figure 9 with Figure 11, the latter showing the mixing ratio time series for years 95–100 of experiment M3. In the southern hemisphere the time series are indistinguishable, confirming that the differences in source structure are eliminated in the transport to these regions, that is, the virtual source for the southern hemisphere is insensitive to details of the northern hemisphere source. Differences in the equatorial time series are detectable but extremely small. More significant differences are, not surprisingly, evident in the northern hemisphere, although, even there, they are largely confined to the lower troposphere. At 30°N (and, indeed, throughout the northern hemisphere) there are major differences at 1000 mbar; while the time of maximum mixing ratio is much the same, the shape of the seasonal variation is different and the magnitude of the variability is almost twice as large in experiment M3 as in N3. Despite these large surface differences, however, the seasonal variations are very similar in the middle and upper troposphere, even in latitudes where the local surface source is substantial. Thus it appears, at least in the context of the model used here, that the seasonality in mixing ratios of tropospheric constituents induced by seasonal variability in transport processes is relatively insensitive to changes in source structure, except in the immediate vicinity of the source.

9. Long-Lived Constituents With Stratospheric Sinks

The results obtained earlier indicate that there are structural and seasonal characteristics which are universal to all long-lived constituents that satisfy one of the conditions that $\lambda$ be spatially uniform or that $\lambda \ll \gamma$ everywhere. While the first of these is unlikely to be satisfied for any real tracer, the second may be valid for some which are accumulating in the atmosphere. On the face of it, the results are not applicable to those tracers (for example, the chlorofluorocarbons) which have long chemical lifetimes but which may not satisfy the criterion $\lambda \ll \gamma$ within the stratosphere. However, it will be demonstrated later that such constituents do exhibit the general properties of long-lived tracers within the troposphere, provided that their sinks are confined to the stratosphere and that $\langle \lambda \rangle \ll \gamma$. In fact, such a result is suggested by the observation from the results of experiment N1 (which does not satisfy the criterion $\tau \gg \Gamma$ in the stratosphere) that the tropospheric con-
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Detrended time series were used to analyze the structure of long-lived tropospheric constituents. The constituent structure was almost identical to that of the longer-lived tracers, while the stratospheric structure was not. This implies that those transport paths from the source to any tropospheric location via the stratosphere do not contribute significantly to the tropospheric mixing ratio and therefore that tropospheric structure may not be sensitive to the details of stratospheric sinks.

Consider a constituent with a nonconstant sink \( \lambda \) which is nonzero only within the stratosphere, where it may be locally large. The global mean of (11) then gives

\[
\gamma \langle q \rangle + \langle \lambda q \rangle = \langle S \rangle
\]

whence

\[
\tau = \frac{\langle q \rangle}{\langle \lambda q \rangle} = \frac{\gamma + \langle \lambda q \rangle}{\langle q \rangle}^{-1}
\]

Unlike the constant-\( \lambda \) case, \( \tau \) depends on the instantaneous structure of \( q \). However, within the region of the sink, it may be assumed that \( q \ll \langle q \rangle \), so that \( \langle \lambda q \rangle \ll \langle \lambda \rangle \langle q \rangle \). Therefore if \( \gamma \gg \langle \lambda \rangle \), (45) simply tells us that \( \tau \approx \gamma^{-1} \), just as in the constant-\( \lambda \) case when \( \gamma \gg \langle \lambda \rangle \). Moreover, the implication of the results noted earlier from experiment N1 is that the dependence of the structure of \( q \) within the lower atmosphere should be independent of the stratospheric sink under these conditions.

This is apparent from the results of experiment N4, in which (see Table 1) the sink is confined to the stratosphere; the global chemical lifetime \( \langle q \rangle / \langle \lambda q \rangle \) at equilibrium in this experiment is about 30 years. To illustrate this, the evolving fractional interhemispheric gradients \( \Delta q(p, \rho) \) are shown for each of years 3–100 of experiment N4 in Figure 12 for (\( \rho, p \)) = (30°, 1000 mbar), (30°, 500 mbar), and (30°, 190 mbar). Recall that for a uniform sink, \( \Delta q(p, \rho) \) approaches a linear dependence on \( \tau \) when the latter is sufficiently large (cf. Figure 8), but the relationship will not apply in general when \( \tau \) is comparable with the exchange times from the source across the region of interest. In the present case we also anticipate that the relation will break down for large \( \tau \), specifically, before \( \tau \) becomes comparable with \( \langle q \rangle / \langle \lambda q \rangle \), that is, about 30 years. This behavior is evident in Figure 12. At (\( \rho, p \)) = (30°, 190 mbar) and (30°, 500 mbar), a linear relationship holds good for \( \tau < 6 \) years; at larger \( \tau \) the gradients are a little weaker than predicted, in fact appearing to asymptote toward a negative gradient for large \( \tau \). At 1000 mbar (Figure 12c) a linear relationship is not apparent, except for large \( \tau \), but the same was found to be the case in Figure 8, when it appeared that proximity to the source undermined the theoretical prediction. In fact, at all three levels, \( \Delta q \) as shown in Figure 12 matches Figure 8 very closely until \( \tau \) exceeds 10 years; thus the expectation that the tropospheric structure of experiment N4 will match that of the constant-sink experiments is confirmed by these results.

10. SUMMARY AND DISCUSSION

Levy et al. [1982] drew attention to the distinction between dynamical and chemical adjustment of atmospheric constituent distributions; for long-lived constituents the former proceeds more rapidly than the latter. In the experiments described here, it has become apparent that there are in fact three time scales involved, one chemical and two dynamical: adjustment to slope equilibrium, adjustmentment to gradient equilibrium, and adjustment to chemical equilibrium.
Adjustment to Slope Equilibrium

In much, though not all, of the model domain, the isopleth slopes rapidly attain their equilibrium values, corresponding to the local dynamical equilibrium described by Mahlman et al. [1986] and Holton [1986]. In the current model this occurs on a time scale of less than 6 months.

Adjustment to Gradient Equilibrium

Adjustment to gradient equilibrium is reached when all significant paths from the source to the location of interest have come to equilibrium; the relevant time scale is thus a global one. In this model, mixing ratio gradients attain equilibrium in the troposphere in a time which is less than 5 years; stratospheric adjustment takes longer.

Adjustment to Chemical Equilibrium

The global-mean mixing ratio \( \langle q \rangle \) adjusts on a time scale which is determined by the chemical lifetime of the constituent.

The focus of this paper is on those constituents which are in slope and gradient equilibrium. A key parameter is the "dynamic lifetime" \( \tau = \langle q \rangle / \langle S \rangle \), the inverse of which is just the sum of the inverse of the chemical lifetime and the growth rate of the global mean mixing ratio. On very general grounds, it has been argued that constituents of sufficiently large dynamic lifetime will exhibit certain universal characteristics. Under conditions that (1) \( \tau \) is much longer than the exchange time \( \Gamma(q, p) \) within the region of interest, and either (2) the rate of removal of the constituent, \( \lambda \), is spatially uniform within the atmosphere, (3) the growth rate \( \gamma \) of the constituent is much greater than \( \lambda \) everywhere, or (4) the sink is confined to the stratosphere and \( \gamma > \langle \lambda \rangle \) (in which case the following statements are applicable only to the troposphere), then the following statements apply.

1. To a first approximation, the constituent is well mixed (that is, spatial variations are small) and \( q \approx \langle q \rangle \) everywhere. The \( \langle q \rangle \) may be time-dependent but is independent of transport processes, being a balance between the constituent source and its sink (and/or the growth of its atmospheric loading).

2. The structure of the constituent mixing ratio, \( q(q, p) = \langle q \rangle \), depends only on the magnitude and geographical distribution of the source and not on \( \tau \). Isosepts of constant mixing ratio are simply isopleths of the exchange time \( \Gamma \); this quantity is not the time taken for transport from the source via a single direct route, but reflects an aggregate time scale for transport via an infinite number of paths.

3. Gradients of mixing ratio between two locations are proportional to the difference in exchange times to those locations from the source and are not related to any measure of direct transport times between the two locations.

4. Fractional gradients of mixing ratio are inversely proportional to \( \tau \) but independent of the source strength and, in regions sufficiently remote from the source, of the distribution of the source.

5. If, in addition to conditions 1–4 in the itemized list above, both the source and \( \lambda \) are independent of time of year, seasonal variability of constituent mixing ratio is induced by seasonality in atmospheric transport acting on the background gradients. The magnitude and the spatial and seasonal structure of this variability is also independent of \( \tau \). The frac-

Fig. 11. (Opposite) As in Figure 9, but for years 95–100 of experiment M3.
Fig. 12. Fractional gradient $\Delta(30^\circ, p)$ at $p$ equal to (a) 190 mbar, (b) 500 mbar, and (c) 1000 mbar for each year of experiment N4, plotted against $\tau^{-1}$, the inverse of the dynamic time scale.

The characteristic meridional structure of long-lived constituents in the model described in section 4 shows a number of distinct features (for example, strong horizontal gradient in the equatorial lower troposphere, reversed vertical gradient in the southern hemisphere troposphere, weak surface gradients at high latitudes of both hemispheres), all of which have simple explanations in terms of the transport processes represented in the model. At high latitudes, quasi-horizontal diffusion is the dominant process; this leads to equilibrium slopes which are quasi-horizontal (in fact, quasi-isentropic) and produces, for example, the result that in experiments with a mid-latitude northern hemisphere source, the maximum mixing ratio above the lowest model layer was found to be at the north pole rather than in mid-latitudes. In low latitudes of the model, vertical diffusion is very efficient, almost destroying any vertical gradients there. Except in high latitudes, advection by the Hadley cell is apparently the most effective mechanism for latitudinal transport. The tight low-level gradients in the tropics are produced by the confluence of this cell. Cross-equatorial exchange takes place in the upper troposphere, where both the Hadley cell outflow and the strongest quasi-horizontal mixing are located; one consequence of this is the fact that for a constituent generated in the northern hemisphere, the effective source for the southern hemisphere is located in the tropical upper troposphere, resulting in a reversed vertical gradient there. Amongst other things, the rich meridional structure evident in the model results and in what observations are available for long-lived constituents serves to emphasize the importance of upper-air measurements (that is, at least in the middle and upper troposphere) in a global monitoring network.

The generality of the earlier statements on the meridional and seasonal structure makes possible an approach to the synthesis of observations of long-lived constituents which is largely independent of model assessments. According to these predictions, constituents which have similar geographical source distributions (which includes most constituents of industrial origin) should exhibit common properties. Given knowledge of the dynamic lifetime for any constituent, it will be possible to determine atmospheric exchange times $\Gamma(\phi, p)$ and/or differential exchange times $\Delta^{\Gamma}(\phi, p)$ (through equations (30) and (37)) and the “universal” seasonality $\Gamma(\phi, p, \theta)$ (through equation (42)). These quantities are, according to this

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theory, universal to all such long-lived tracers which satisfy conditions 1-4 and represent the impact of transport alone on constituent distributions. Failure of any constituent to satisfy this would then be an indication that these assumptions are not valid for that constituent and might help to highlight factors other than transport which are important in the determination of its meridional structure or seasonal variability. Similarly, knowledge of the dynamic lifetime of one constituent allows estimates to be made of the lifetimes of other constituents with similar source structures (such as most constituents of industrial origin) and weak sinks without resort to models, simply by comparing observed gradients between two locations or even by comparing seasonal cycles at a single observing site (though the seasonal variation of mixing ratios of long-lived constituents is necessarily very weak).

Such an approach might also simplify and strengthen the procedure of model validation. Interpretation of model predictions for a specific constituent with observations is clouded by the multiplicity of possible causes (errors in the representation of transport processes, of sources and sinks) of any discrepancy. If atmospheric exchange times or seasonal cycles can be identified from observations for long-lived constituents (such as the chlorofluorocarbons) with mid-latitude northern hemisphere sources, then the ability of a model to reproduce these can be tested in a single experiment. As we have seen, such an exercise would be rather insensitive to errors in the specification of sources and sinks.

**APPENDIX A: EXCHANGE TIMES FOR AN EXTENDED SOURCE**

The global structure of \( q(\phi, p) \) for a constituent with large and spatially uniform \( T \) is given by the solution of (20). Consider first a surface source \( S_L \) localized at latitude \( \phi_L \):

\[
S_L(\phi) = 2(S_L) \delta(\phi - \phi_L) \cos \phi_L
\]

where \( \delta(\cdot) \) is the delta function. The solution to (20) for this source may be written

\[
q_L(\phi, p) - q_L(\phi, L, p) = -\Gamma(\phi, p) \langle S_L \rangle
\]

where \( \Gamma \) is a Green's function for this problem, is independent of \( \tau \). By the arguments given in the text following (27) and (30), \( \Gamma \) may also be regarded as the exchange time from the source at \( (\phi_L, p_L) \) to \( (\phi, p) \). Now, for a general, extended, surface source \( S(\phi) \),

\[
S(\phi) = \int_{-\pi/2}^{\pi/2} \left[ \delta(\phi - \phi') \cos \phi' \right] S(\phi') \cos \phi' d\phi'
\]

and the response may be written

\[
q(\phi, p) = \int_{-\pi/2}^{\pi/2} \left[ q_L(\phi, 0) - G(\phi, \phi_L, p) \right] S(\phi_L) \cos \phi_L \cos \phi' d\phi'
\]

Now, the maximum mixing ratio must be located where the source is nonzero, that is, at the surface. If the latitude at which \( q \) is maximum is \( \phi_m \), then (A4) tells us that the maximum value is

\[
q(\phi, p) = \int_{-\pi/2}^{\pi/2} \left[ q_L(\phi_L, p_L) - G(\phi_m, \phi_L, p_L) \right] \langle S(\phi_L) \rangle \cos \phi_L \cos \phi' d\phi'
\]

whence

\[
\Gamma(\phi, p) = \langle S_L \rangle^{-1} \left[ \int_{-\pi/2}^{\pi/2} \left[ G(\phi, \phi_L, p) - G(\phi_m, \phi_L, p) \right] S(\phi_L) \cos \phi_L \cos \phi' d\phi' \right]
\]

is a function only of the source structure.

**APPENDIX B: BUDGET FOR MIXING RATIO ISOPLETH SLOPES**

Derivation of Equation (32)

With latitude coordinate \( y = a \sin \phi \), (1) may be written

\[
\frac{\partial q}{\partial t} = \frac{\partial}{\partial y} \left[ \chi \frac{\partial q}{\partial y} + \frac{\partial}{\partial z} \left( p \frac{\partial q}{\partial y} \right) \right] + \nabla \cdot \mathbf{F} + S - \lambda q
\]

where

\[
\mathbf{F} = \frac{\partial}{\partial y} \left[ D_y \frac{\partial q}{\partial y} + D_z \frac{\partial q}{\partial z} \right] + \frac{1}{p} \frac{\partial}{\partial z} \left[ p D_y \frac{\partial q}{\partial y} + p D_z \frac{\partial q}{\partial z} \right]
\]

is the diffusive contribution to (1). Now, the tangent of the angle to the horizontal of the mixing ratio isopleths is

\[
\sigma = -\frac{\partial q}{\partial y} \left[ \frac{\partial q}{\partial z} \right]^{-1}
\]

therefore

\[
\frac{\partial \sigma}{\partial t} = -\frac{\partial^2 q}{\partial y \partial t} - \sigma \frac{\partial^2 q}{\partial z^2} \left( \frac{\partial q}{\partial z} \right) \left( \frac{\partial q}{\partial y} \right) \left( \frac{\partial q}{\partial z} \right)
\]

Substituting for \( \partial q/\partial t \) from the right-hand side of (B1), we may identify four separate contributions to (B4). That associated with the source term is simply

\[
E_{SOURCE} = -\frac{\partial S}{\partial y} + \sigma \frac{\partial S}{\partial z} \left( \frac{\partial q}{\partial z} \right)
\]

The sink contribution is

\[
E_{SINK} = q \left[ \frac{\partial \sigma}{\partial y} + \sigma \frac{\partial \sigma}{\partial z} \right] \left( \frac{\partial q}{\partial z} \right)
\]

Substituting the advective terms in (B1) (the first two terms on the right-hand side) into (B4) gives

\[
E_{ADV} = \frac{\partial^2 q}{\partial y^2} \left[ \chi \frac{\partial q}{\partial y} - \frac{\partial^2}{p \partial y \partial z} \left( p \frac{\partial q}{\partial y} \right) + \sigma \frac{\partial^2 q}{\partial y \partial z} \left( \chi \frac{\partial q}{\partial z} \right) \right]
\]

With some manipulation, this becomes

\[
E_{ADV} = \left[ \frac{\partial^2 q}{\partial y^2} + \sigma \frac{\partial}{\partial y} \left( -\chi \frac{\partial q}{\partial y} \right) + \frac{\partial^2 q}{\partial z^2} \right] \frac{\partial q}{\partial z} \left( \frac{\partial q}{\partial y} \right) \left( \frac{\partial q}{\partial z} \right)
\]
Finally, the diffusion term may be written in the form

$$E_{\text{DIFF}} = -\left[ \frac{\partial}{\partial y} (V \cdot F) + \sigma \frac{\partial}{\partial z} (V \cdot F) \right]$$

(B8)

**Derivation of Equation (33)**

The diffusion term does not yield any apparent simplification in general. However, it does have a simple form for the case used in the text, where $D_{yy} = 0$, $D_{zz} = 0$, and $D_{yy}$ is constant. Then, $V \cdot F = D_{yy} \frac{\partial^2 q}{\partial y^2}$ and (B8) becomes

$$E_{\text{DIFF}} = D_{yy} \left[ \frac{\partial q}{\partial y} \right] \frac{\partial^2 q}{\partial y^2} = \left[ \frac{\partial q}{\partial y} \right]^2$$

(B9)

But, from (B3),

$$\frac{\partial \sigma}{\partial y} = \left[ \frac{\partial q}{\partial y} \frac{\partial^3 q}{\partial y^3} - \frac{\partial q}{\partial z} \frac{\partial^3 q}{\partial y^3} \right] \frac{\partial^2 q}{\partial y^2}$$

whence

$$\frac{\partial}{\partial y} \left[ \left( \frac{\partial q}{\partial y} \right)^2 \frac{\partial \sigma}{\partial y} \right] = \frac{\partial q}{\partial y} \frac{\partial^3 q}{\partial y^3} - \frac{\partial q}{\partial y} \frac{\partial^3 q}{\partial y^3}$$

and therefore

$$E_{\text{DIFF}} = D_{yy} \frac{\partial}{\partial y} \left[ \left( \frac{\partial q}{\partial y} \right)^2 \frac{\partial \sigma}{\partial y} \right] \frac{\partial^2 q}{\partial y^2}$$

(B10)

Now, let us assume that $|x| \ll D_{yy}$; this is a reasonable assumption in most extratropical locations (cf. Figures 5-8 of PM). Since (B10) scales as $|\sigma|$, the balance $E_{\text{ADV}} \approx E_{\text{DIFF}}$ can be achieved for small $\sigma$, when (B7) gives (35),

$$E_{\text{ADV}} \approx \frac{\partial^4 \chi}{\partial y^4}$$

provided $|\sigma| \ll h/l$, where $h$ and $l$ are respectively the height and length scales of $\chi$. Moreover, for small $\sigma$ we may neglect the $y$ derivative of $\partial q/\partial z$ in (B10) so that the steady state of (32) becomes

$$\frac{\partial^4 \chi}{\partial y^4} \approx -D_{yy} \frac{\partial^2 \sigma}{\partial y^2}$$

Since we have assumed constant $D_{yy}$, the solution consistent with zero flux through the lateral boundaries is

$$\sigma \approx -\sigma/D_{yy}$$

This result is discussed in the text, following (33).

**Acknowledgments.** We thank Paul Fraser for discussions during the progress of the work described here and David Andrews, Ian Enting, Peter Hyson, and anonymous referees for comments on an earlier draft of this paper.

**References**


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