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Sensitivity Study of Advection and Diffusion Coefficients in a Two-Dimensional Stratospheric Model Using Excess Carbon 14 Data

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Using the California Institute of Technology/Jet Propulsion Laboratory two-dimensional transport model, with transport coefficients taken from Yang and Tung (1989), we study the time evolution of excess carbon 14 in the stratosphere and the troposphere from October, 1963 to December, 1966. The model provides a satisfactory simulation of the observed data. Due to the impulsive nature of its source, initial distributions of excess carbon 14 exhibit large spatial gradients. This permits important constraints on the range of transport coefficients in the lower stratosphere to be derived. The standard model uses the circulation and eddy diffusivity of the year 1980. Large deviations (by factor of 2) from this standard transport are ruled out by our model. A self-consistently derived K_{yy} which is small ($\sim 10^9$ cm^2 s^{-1}) in tropical regions, but is larger ($\sim 10^{10}$ cm^2 s^{-1}) at higher latitudes is preferred. A K_{zz} as large as 1×10^4 cm^2 s^{-1} would be inconsistent with the data. Excess carbon 14 is removed from the atmosphere with surface deposition velocities $v_S = 3 \times 10^{-3}$ cm s^{-1} and $v_N = 5 \times 10^{-3}$ cm s^{-1} in the southern and northern hemispheres, respectively. The last result is contrary to the current understanding that the oceans are the dominant sink for excess ^{14}C.

INTRODUCTION

Excess carbon 14 in the stratosphere, collected during the period 1963–1966, has been used by *Johnston et al.* [1976] to calibrate one-dimensional models of the stratosphere. Therefore, it is natural to ask whether this data set is also useful for testing the transport coefficients in two-dimensional models. Recently, H. Johnston (Use of excess carbon 14 data to test two-dimensional stratospheric models, submitted to *Journal of Geophysical Research*, 1988; hereinafter Johnston (1988)) advocated the usefulness of this exercise, and carried through considerable data processing to make the data convenient for use in a two-dimensional model. Figure 1 presents Johnston's (1988) initial data in October, 1963 in pressure coordinates (Johnston used altitude). This is several months after the last nuclear bomb test, so that the excess ^{14}C has attained approximate zonal symmetry. Subsequently, the evolution of excess ^{14}C was documented at 70°N, 31°N, 9°N, and 42°S for several years until July, 1966, with data extending to about 30 km. We must emphasize that there are no data above 33 km or southward of 42°S. Hence, much of Figure 1 consists of "hand-drawn data", which are marked by dashed lines. The units used for the contours in the figure are 10^5 ^{14}C atoms per gram of dry air. These numbers multiplied by 4.82×10^{-18} give the volume mixing ratio of ^{14}C. Thus the maximum mixing ratio of ^{14}C in the stratosphere is 5.9×10^{-15}, and the maximum column abundance (from the ground to 56 km) is 2.3×10^{10} atoms cm^{-2}. The purpose of this paper is to use the California Institute of Technology/Jet Propulsion Laboratory (Caltech/JPL) two-dimensional model of the stratosphere to simulate the transport history of excess ^{14}C, using Figure 1 as the initial distribution.

There are three essential components of transport in "modern" two-dimensional stratospheric models [*Garcia and Solomon*, 1983; *Guthrie et al.*, 1984; *Ko et al.*, 1985; *Stordal et al.*, 1985; *Holton*, 1986; *Tung and Yang*, 1988]. First is the Brewer-Dobson circulation, which consists of upwelling at the equatorial tropopause and descending motion at high latitudes [*Brewer*, 1949; *Dobson*, 1956]. Second is the horizontal eddy diffusivity K_{yy}. The third component is the summer-pole to winter-pole Leovy circulation in the upper stratosphere and mesosphere [*Leovy*, 1964]. Vertical eddy diffusivity, K_{zz}, which plays a fundamental role as the only transport process in one-dimensional models, is not believed to be important in the bulk of the stratosphere. It is the triumph of this new generation of two-dimensional models to be able to account for the observed vertical, latitudinal, and seasonal distributions of source molecules such as N_2O, CH_4, $CFCl_3$, and CF_2Cl_2, as well as photochemically produced species such as O_3 (see *World Meteorological Organization (WMO)* [1985] for detailed comparison between theory and data). Indeed, there is sufficient confidence in the transport properties of the model that the discrepancy between observed and computed NO_y led to the postulation of a new NO_y source in the equatorial lower stratosphere [*Ko et al.*, 1986].

What then is unique about excess ^{14}C, and how can it lead to a refinement of the transport component of a two-dimensional model? First, excess ^{14}C (as $^{14}CO_2$) is a chemically inert tracer in the stratosphere. Its distribution, once we set the initial conditions using observations, is governed only by transport. There are no uncertainties due to chemical production or loss. Second, the source of excess ^{14}C is impulsive rather than quasi-steady. The ultimate disappearance of this radioactive material from the stratosphere provides an absolute calibration for mass exchange between the stratosphere and the troposphere. In fact, it was the study of the dissipation of stratospheric radioactive debris that first yielded estimates of the rate of mass exchange between the troposphere and the middle atmosphere [*Reed and*

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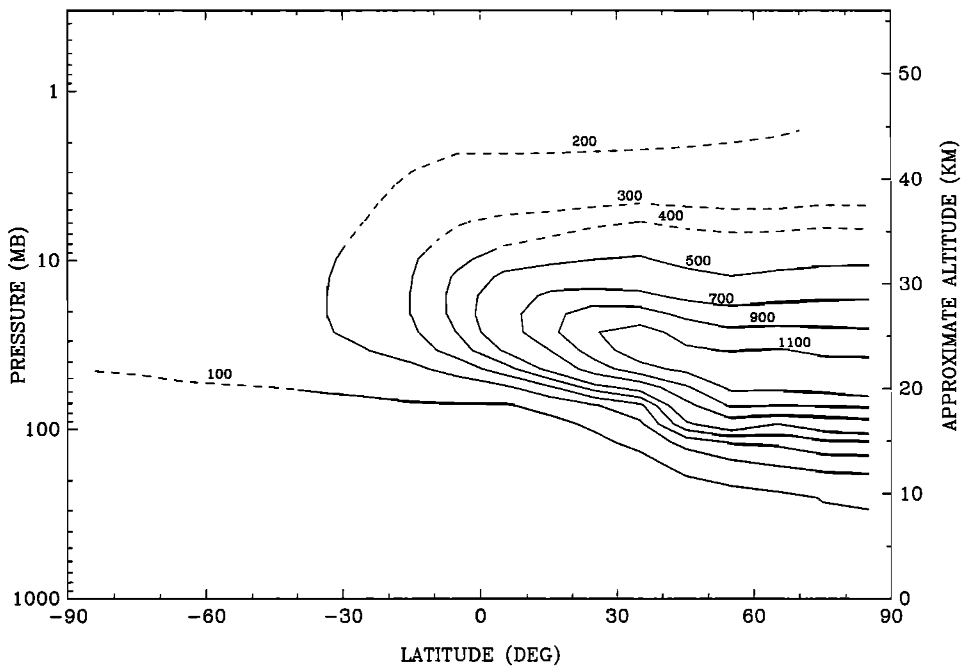

Fig. 1. Initial ^{14}C distribution in October, 1963 taken from Johnston (1988) and interpolated to our pressure coordinates. The units are in 10^5 ^{14}C atoms per gram of dry air. To convert to ^{14}C volume mixing ratios, the numbers must be multiplied by 4.82×10^{-18}. The dashed lines are not based on measured data (see text).

German, 1965; *Mahlman*, 1965; *List and Telegadas*, 1969; *Reiter*, 1975].

The advantages of excess ^{14}C as a tracer may be appreciated if we compare it with other chemically produced, downward transported species such as NO_y and O_3. The ultimate source of NO_y is upwelling of tropospheric N_2O, and the primary stratospheric sink of NO_y is downward flow into the troposphere. Thus the source and sink are related to the same circulation. Hence, the abundance of NO_y in the stratosphere would be rather insensitive to the exact strength of the Brewer-Dobson circulation. As for O_3, 99% of it is produced and destroyed chemically in the stratosphere. The observed distribution is the consequence of a competition between chemistry and transport. Given the current uncertainties in the chemistry of O_3 (see for example *Froidevaux et al.* [1985]), it is hard to see how O_3 can be considered a well-defined tracer. In other words, even complete agreement between model results and observations may be due to a fortuitous cancellation of errors between chemistry and dynamics.

A third advantage in using excess ^{14}C is that, due to its impulsive nature, excess ^{14}C has very steep initial spatial gradients. In October, 1963 near the peak at midlatitudes, the mixing ratio decreases in the vertical by more than a factor of two per atmospheric scale height away from the peak. At this time there is also a large gradient in the horizontal dimension. The column densities above 10 km at the north pole, equator, and south pole are in the ratio 10:3:1.3 (see Figure 9). Thus, the degree of latitudinal contrast in the northern hemisphere is between those of O_3 and HNO_3 [*Ko et al.*, 1985], but the interhemisphere asymmetry is unique. Such large gradients would allow us to explore the sensitivity of our transport model to values of K_{yy} and K_{zz}, especially in the lower stratosphere. The fact that our numerical scheme has very little dispersion gives us confidence in the values of the eddy diffusion coefficients thus derived [*Prather*, 1986; R.-L. Shia et al., Two-dimensional atmospheric transport and chemistry model: Numerical experiments with a new advection algorithm, submitted to *Journal of Geophysical Research*, 1988 (hereinafter Shia et al., (1988))]. Unfortunately, we can learn almost nothing about transport in the upper stratosphere due to the limited altitude of the data base (mostly confined to below 30 km). Also, most of the steep gradients have disappeared after the first six months so that we cannot derive any useful information on K_{yy} and K_{zz} after this time.

For our first attempt to reproduce the excess ^{14}C distribution, for the period from October, 1963 to July, 1966, we use the transport coefficients of *Yang and Tung* [1989]. The authors diagnostically calculated the diabatic circulation from the National Meteorological Center (NMC) temperature field and self-consistently determined the isentropic mixing coefficients by solving the momentum equation (see discussion in next section). These transport coefficients have been successfully used to simulate O_3 distribution. A number of runs will be made to test the sensitivity of our results to variations in advection and horizontal eddy diffusivity (K_{yy}). Upper limits on K_{zz} can be set. Finally, the interaction between ^{14}C and the surface will be explored in our model. As we shall see, our major contributions will be sensitivity studies of two-dimensional transport modeling in the lower stratosphere. We find that simulation of the removal of excess ^{14}C from the stratosphere requires K_{yy} values which are larger in the extratropics than in the tropics, and that $K_{zz} < 10^4$ cm^2 s^{-1} in the lower stratosphere. Furthermore, our sensitivity studies indicate that tropospheric removal (at the surface) must be significantly faster in the northern hemisphere than in the south.

MODEL DESCRIPTION

The meridional coordinate that has been adopted in the two-dimensional model is $y = a\theta$, where a is planetary radius, and θ is latitude defined to be $-90°$ at the south pole and $90°$ at the north pole. For vertical coordinate we choose $z = H \ln(p_s/p)$, where p is pressure, p_s is surface pressure, and H is a fixed constant. The constants p_s and H are set equal to 1000 mbar and 7 km, respectively, in this paper. We also use the dimensionless coordinate $\xi = z/H$. For the studies reported here, the transport code (Shia et al., 1988) uses a grid having 18 latitudinal boxes from pole to pole, and 32 vertical layers from $z = 0$ to $z = 56$ km. The transport circulation consists of a zonal mean stream function ψ and eddy diffusivities K_{yy} and K_{zz}. The two-dimensional model is modular in form, so that the transport circulation can be derived from either observed or modeled (for example, by a general circulation model (GCM)) fields (R. W. Zurek et al., Derivation of the stream function and eddy diffusivity from monthly mean fields of the Geophysical Fluid Dynamics Laboratory stratospheric model within the framework of a two-dimensional model, to be submitted to *Journal of Geophysical Research*, 1989; hereinafter Zurek et al. (1989)). That circulation can also be independently specified, which is the option taken in this paper.

From the stream function we can readily derive velocities for meridional motion, v, and vertical motion, w,

$$v = -\frac{1}{\cos\theta} e^{\xi} \frac{\partial}{\partial z} \left(e^{-\xi}\psi\right) \quad (1)$$

$$w = \frac{1}{\cos\theta} \frac{\partial \psi}{\partial y} \quad (2)$$

Note that the quantities ψ, v, and w refer to the Eulerian residual circulation, and are usually denoted by ψ^*, v^*, and w^*. However, we drop the asterisks for simplicity. This velocity field $\mathbf{u} = (v, w)$ is non-divergent in the following sense,

$$e^{\xi}\nabla \cdot \left(e^{-\xi}\mathbf{u}\right) = \frac{1}{\cos\theta}\frac{\partial}{\partial y}(\cos\theta v) + e^{\xi}\frac{\partial}{\partial z}\left(e^{-\xi}w\right) = 0 \quad (3)$$

The eddy diffusive fluxes are parameterized by

$$F_y = -\left(K_{yy}\frac{\partial \chi}{\partial y} + K_{yz}\frac{\partial \chi}{\partial z}\right) \quad (4)$$

$$F_z = -\left(K_{zy}\frac{\partial \chi}{\partial y} + K_{zz}\frac{\partial \chi}{\partial z}\right) \quad (5)$$

where χ is the mixing ratio of the tracer under consideration. The horizontal eddy diffusivity K_{yy} cannot be arbitrarily specified, but is derived here from the relation

$$K_{yy}\frac{\partial q}{\partial y} = -\nabla \cdot \mathbf{E} \quad (6)$$

where q is the zonal mean potential vorticity and $\mathbf{E}$ is the Eliassen-Palm flux [*Edmon et al.*, 1980; *Tung*, 1986, 1987; *Plumb and Mahlman*, 1987; *Newman et al.*, 1986, 1988]. According to *Tung* [1982], $K_{yz} \cong K_{zy} \cong 0$ in isentropic coordinates. However, in transforming from this coordinate to log pressure coordinate (used in our model), K_{yy} may have a nonzero projection into K_{yz}. The impact of this K_{yz} derived from coordinate transformation is, however, small due to the small angle of inclination of the isentropes with respect to the isobaric surfaces in the stratosphere, and can therefore be ignored. In our standard model we adopt an arbitrarily small value of $K_{zz} = 1 \times 10^2$ cm^2 s^{-1} in the stratosphere.

In the troposphere, the diabatic circulation is not very effective for mass transport. Rather than trying to develop a correct stream function for the troposphere, we choose to parameterize the transport by eddy diffusion. This region of strong diffusive mixing extends to the model tropopause, defined in each month by the minimum z such that,

$$\frac{dT}{dz} > -2.5 \frac{\text{K}}{\text{km}} \quad (7)$$

Based on our experience with one-dimensional and three-dimensional modeling [*Froidevaux et al.*, 1985; *Pinto et al.*, 1983], we adopt the following values,

$$K_{zz} = 1 \times 10^5 \,\text{cm}^2\,\text{s}^{-1} \quad (8)$$

$$K_{yy} = 1 \times 10^{10} \,\text{cm}^2\,\text{s}^{-1} \quad (9)$$

The continuity equation for the mixing ratio of an inert tracer is

$$\frac{\partial \chi}{\partial t} + v\frac{\partial \chi}{\partial y} + w\frac{\partial \chi}{\partial z} - \frac{1}{\cos\theta}\frac{\partial}{\partial y}\left\{\cos\theta K_{yy}\frac{\partial \chi}{\partial y}\right\} - e^{\xi}\frac{\partial}{\partial z}\left\{e^{-\xi}K_{zz}\frac{\partial \chi}{\partial z}\right\} = 0 \quad (10)$$

where the left-hand side of (10), respectively, denotes the time rate of change of χ, the divergence of advective fluxes, and the divergence of diffusive fluxes (F_i). The transport coefficients for each month (the midmonth values) are entered into the tracer model.

Figures 2*a*–2*d* give the mass-weighted stream function, Ψ_m, for the months of January, April, July, and October. Ψ_m is related to ψ by

$$\Psi_m = 2\pi a \rho_s e^{-\xi}\psi \quad (11)$$

where ρ_s is the density of air at the ground $\rho_s = p_s/gH = 1.46 \times 10^{-3}$ g cm^{-3}, and a is the radius of the Earth. The stream function (Figure 2), and the associated velocities (Figures 3 and 4), and the horizontal eddy diffusivities K_{yy} (Figure 5) are taken from *Yang and Tung* [1989] for the year 1980. They are derived from NMC observational analyses of temperature of the troposphere and stratosphere (up to 8 scale heights [see *Geller and Wu*, 1987]). The data are available from the end of 1978 to the present. The first complete year of data in the series, 1979, is abnormally active when compared with the climatological mean based on all the years in the series. Data from the second year, 1980, are chosen here because they are more "normal." The diabatic velocities and isentropic mixing coefficients were computed in isentropic coordinates and then transformed to pressure coordinates in the present study.

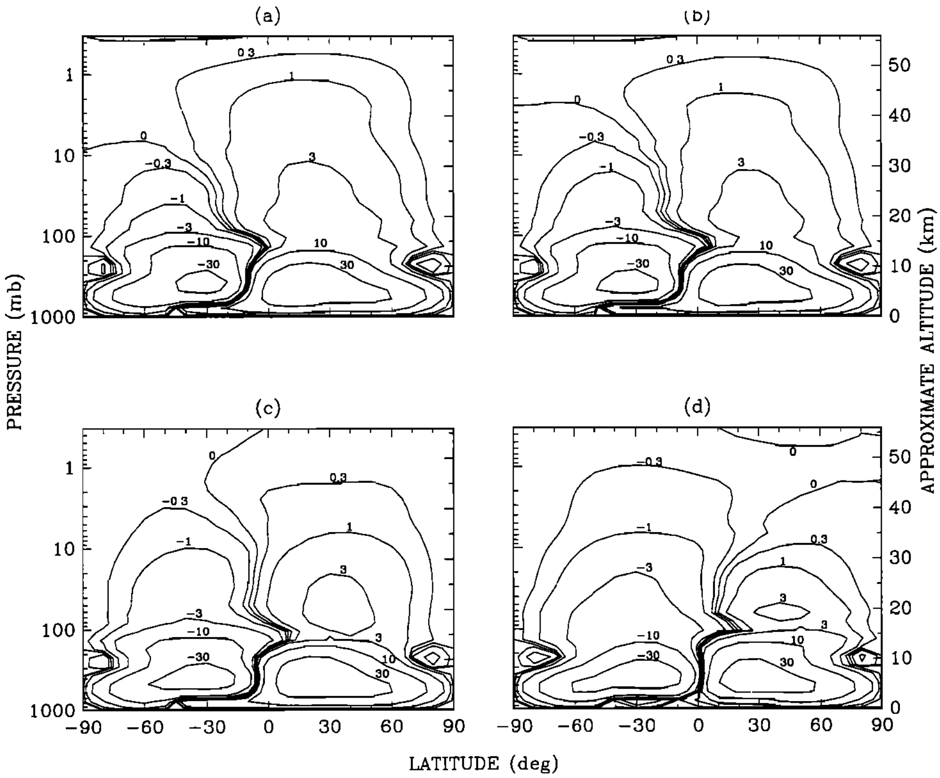

Fig. 2. Mass-weighted stream function Ψ_m in units of 10^{12} g s^{-1}. (*a*) January, (*b*) April, (*c*) July, (*d*) October. Positive (negative) values imply clockwise (anticlockwise) flow.

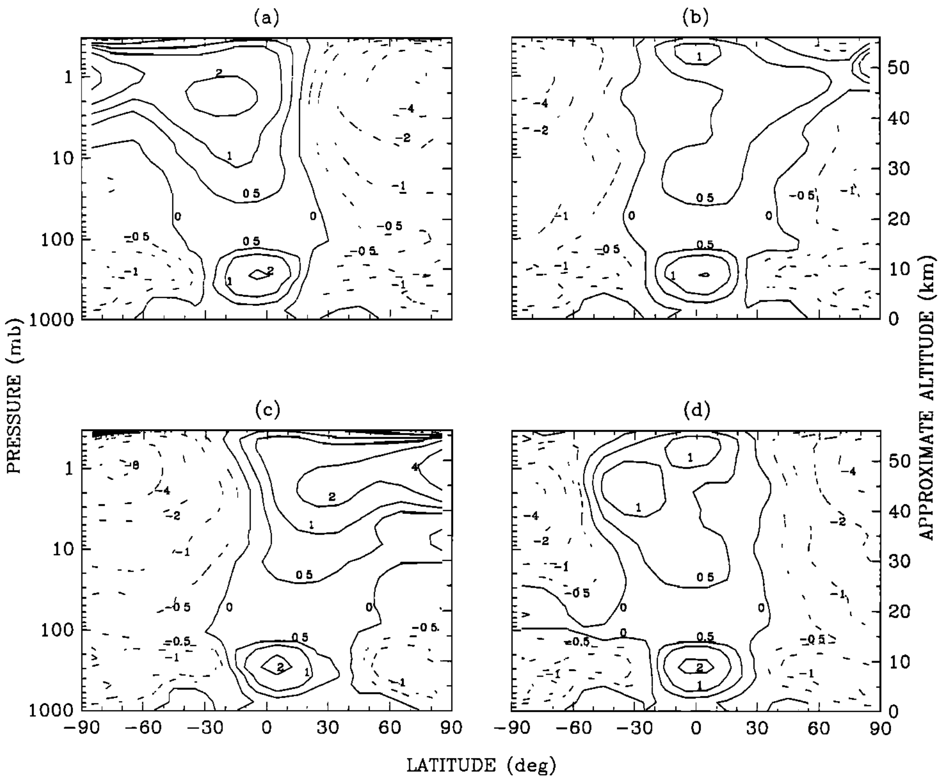

Fig. 3. Vertical velocity *w* computed for the diabatic circulation. The units are millimeters per second. The dotted lines refer to negative values (downward velocity). (*a*) January, (*b*) April, (*c*) July, (*d*) October.

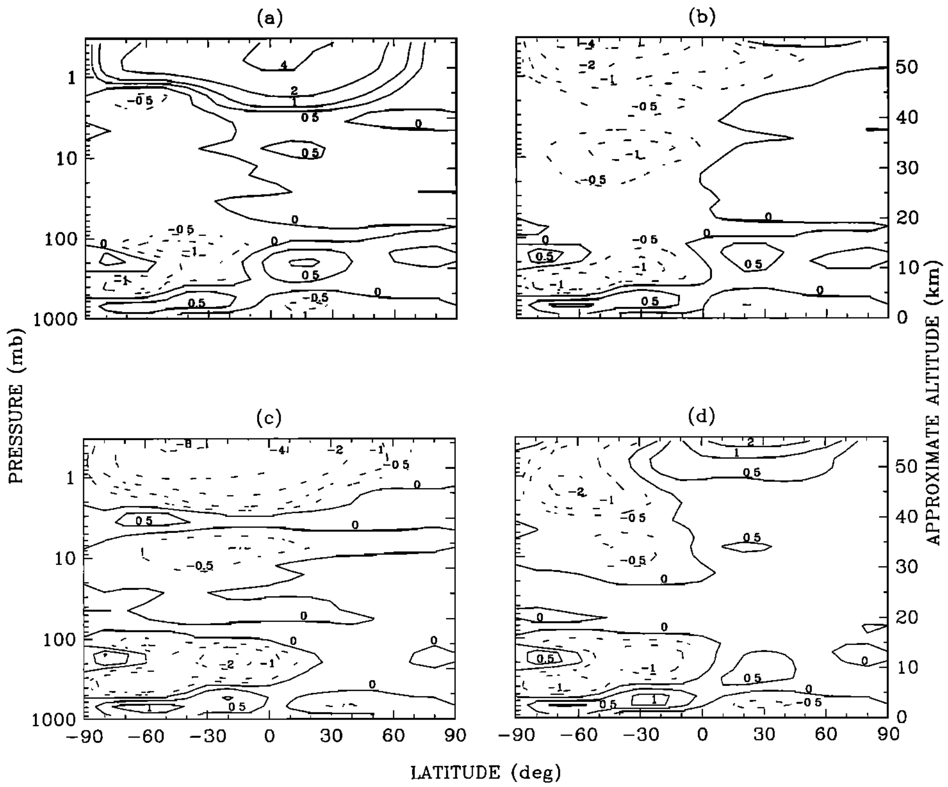

Fig. 4. Same as Figure 3 for meridional velocity v in units of meters per second.

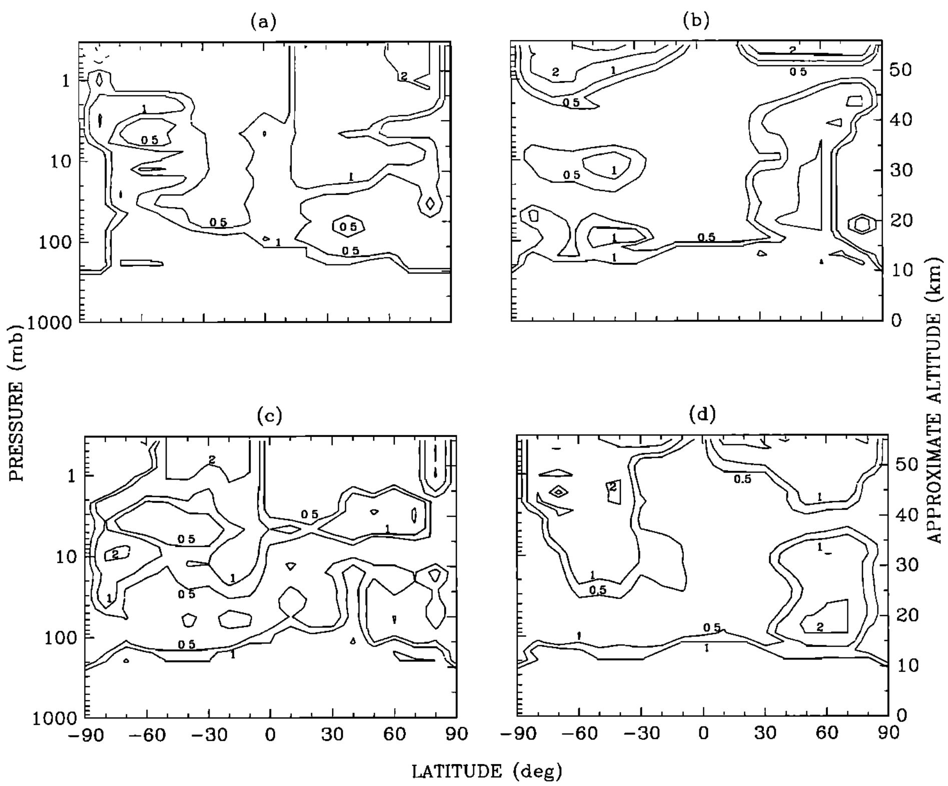

Fig. 5. Same as Figure 3 for K_{yy} in units of 10^{10} cm^2 s^{-1} computed using conservation of potential vorticity.

Ψ_m has a simple physical meaning: it is the integrated mass flux,

$$\Psi_m(\theta, z) = -2\pi a \rho_s \cos\theta \int_0^z e^{-\xi}\, v\, dz$$
$$= 2\pi a^2 \rho_s e^{-\xi} \int_{-\pi/2}^{\theta} \cos\theta\, w\, d\theta \qquad (12)$$

The first expression is the vertically integrated (0 to z) mass flux across latitude θ; the second expression is the horizontally integrated ($-\pi/2$ to θ) mass flux across pressure surface z. Inspection of Figure 2 suggests that the integrated mass flux in the region of equatorial upwelling is about 9.3×10^{12} g s^{-1}. Since the stratospheric mass above 100 mbar is 5.2×10^{20} g, this implies that the entire stratosphere is ventilated in about 1.8 years in our model. Above 10 mbar ($\sim$ 30 km), the stream functions clearly reveal the single-cell summer pole to winter pole circulation (Figures 2*a* and 2*c*). Figures 3*a*–3*d*, 4*a*–4*d*, and 5*a*–5*d* show corresponding values of the vertical velocity w, meridional velocity v, and K_{yy}. Figures 3*a*–3*d* show that the upwelling portion of the Brewer-Dobson circulation extends to ±30° for all seasons. Descending motion at the tropopause occurs at higher latitudes. The horizontal velocities (Figures 4*a*–4*d*) are related to the vertical velocities through the nondivergence relation (3). Note that the order of magnitude of stratospheric K_{yy} is 10^{10} cm^2 s^{-1}, with characteristic horizontal diffusion time over a planetary radius given by

$$\tau_{yy} = \frac{a^2}{K_{yy}} = 4.1 \times 10^7 \text{ s} = 1.3 \text{ years} \qquad (13)$$

This time scale is comparable to that due to advection. Therefore, we expect both advection and diffusion to be important for tracer transport. There are regions of the atmosphere during certain seasons (Figures 5*a* and *c*) with large negative values of K_{yy}. This is due to the breakdown of the approximate relationship (6) between potential vorticity and Eliassen-Palm flux in the upper stratosphere, where gravity waves may become important. Where the computed K_{yy} is negative, we replace it by a small positive value 1×10^8 cm^2 s^{-1}. The values of K_{zz} are given by a small constant (10^2 cm^2 s^{-1}) above the tropopause, and a larger constant (10^5 cm^2 s^{-1}) below the tropopause and are not shown graphically. The essential aspects of our model are summarized in Table 1.

The model was initialized with the distribution of excess ^{14}C as shown in Figure 1. The boundary conditions at the top and at the poles are zero fluxes. After some experimentation, we find a standard deposition velocity at the surface $v_S = 3 \times 10^{-3}$ cm s^{-1} for the southern hemisphere, and $v_N = 5 \times 10^{-3}$ cm s^{-1} for the northern hemisphere. The choice is close to *Liss'* [1988] value of 5.6×10^{-3} cm s^{-1} for ^{14}C takeup by oceans (the stated probable error is 25%; for a more detailed discussion see *Broecker and Peng* [1982]). The sensitivity of our model results to the choice of v_S and v_N will be discussed later.

Having prescribed the transport coefficients, the initial and the boundary conditions, we solve (10) by marching forward in time using the numerical algorithms described by *Prather* [1986] and Shia et al. (1988). The time step is 12 hours, and the stream functions and K_{yy} are updated each month, repeating themselves after 12 months. The run is stopped after 3.25 years in December, 1966.

STANDARD MODEL

The time evolution of the excess ^{14}C cloud in the stratosphere in 1964 as simulated by the standard model is shown in Figures 6*a*–6*d*. Initially excess ^{14}C is transported into the southern stratosphere, and later into the upper stratosphere and the troposphere. Note that in October, 1963 (Figure 1) the 300 units contour line barely crosses the equator, but a year later (Figure 6*d*) it has nearly reached the south pole. By this time the 100 units contour has penetrated into the troposphere at northern high latitudes. Figures 7*a*–7*d* and 8*a*–8*d* follow the subsequent evolution in 1965 and 1966. Motion of material is generally upward near the equator, and downward toward the poles, consistent with the circulation given in Figures 2–4. The effect of K_{yy} in the model is to cause a steady smoothing of the horizontal gradient. To summarize this information more compactly we perform vertical integrals of excess ^{14}C abundance above 10 km. The results, as a function of latitude, for October, 1963 and January 1964–1966, are shown in Figure 9. Note the rapid initial decay of the large horizontal gradient in column excess ^{14}C, followed by a slower rate of decay as the distributions become smoother. Toward 1965 and 1966, the column abundances display an equatorial minimum, reminiscent of that for O_3. The reason is, of course, common to both phenomena, i.e., equatorial upwelling, as is also evident from Figure 8*d*. Starting in January, 1966 (Figure 8*a*), the excess ^{14}C distribution in the lower stratosphere gradually as-

TABLE 1. Summary of the Basic Structure and the Transport Coefficients in the Caltech-JPL Two-Dimensional Model

Model Feature	Specification
Grid	Horizontal coordinate $a\theta$ θ from −85° (south pole) to 85° (north pole) in steps of 10° Vertical coordinate $z = H \ln(p_s/p)$ ($H = 7$ km, $p_s = 1000$ mbar) z from 0 ($p = p_s$) to 56 km ($p = 0.34$ mbar) in steps of 1.75 km
Tropopause	Defined by (7) using GFDL SKYHI GCM data for temperature.
Advection	Residual circulation input from *Yang and Tung* [1989]. Updated every month for middle of the month.
Horizontal diffusion	Self-consistent stratospheric K_{yy} computed using (6) supplied by *Yang and Tung* [1989]. Updated every month. Tropospheric $K_{yy} = 1 \times 10^{10}$ cm^2 s^{-1}.
Vertical diffusion	Stratospheric $K_{zz} = 1 \times 10^2$ cm^2 s^{-1}. Tropospheric $K_{zz} = 1 \times 10^5$ cm^2 s^{-1}.
Boundary conditions	Zero fluxes at the upper boundary and at the poles. Deposition velocities at the lower boundary (see Table 3).

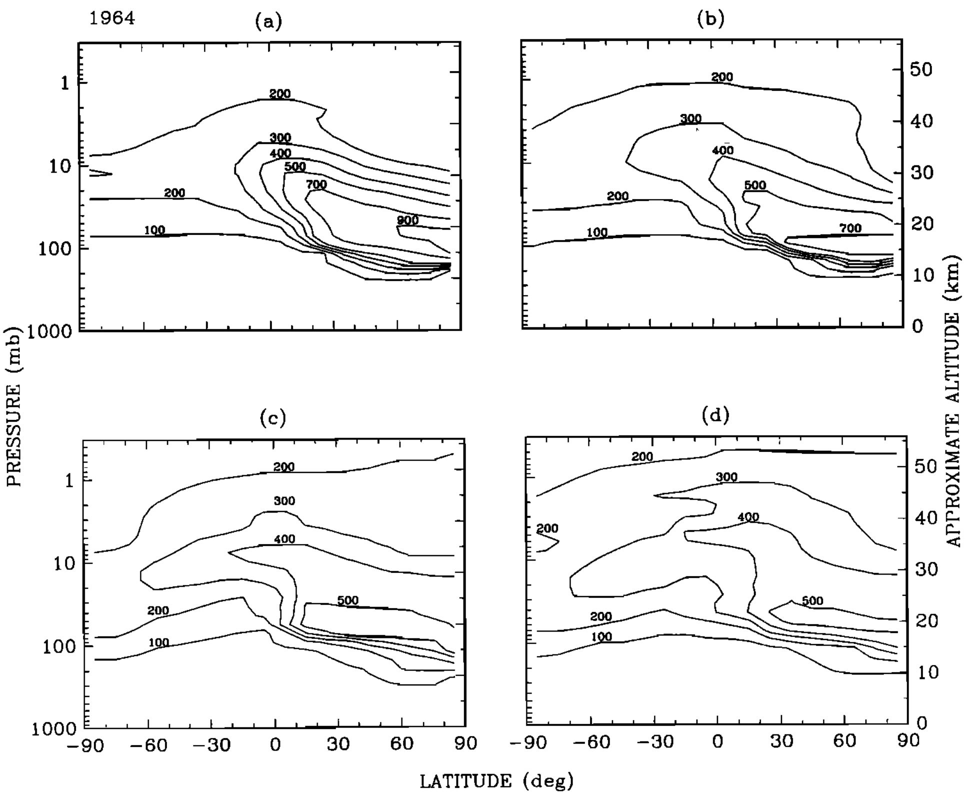

Fig. 6. Contours of excess ^{14}C computed in the standard model in 1964 in the same units as in Figure 1. (*a*) January, (*b*) April, (*c*) July, (*d*) October.

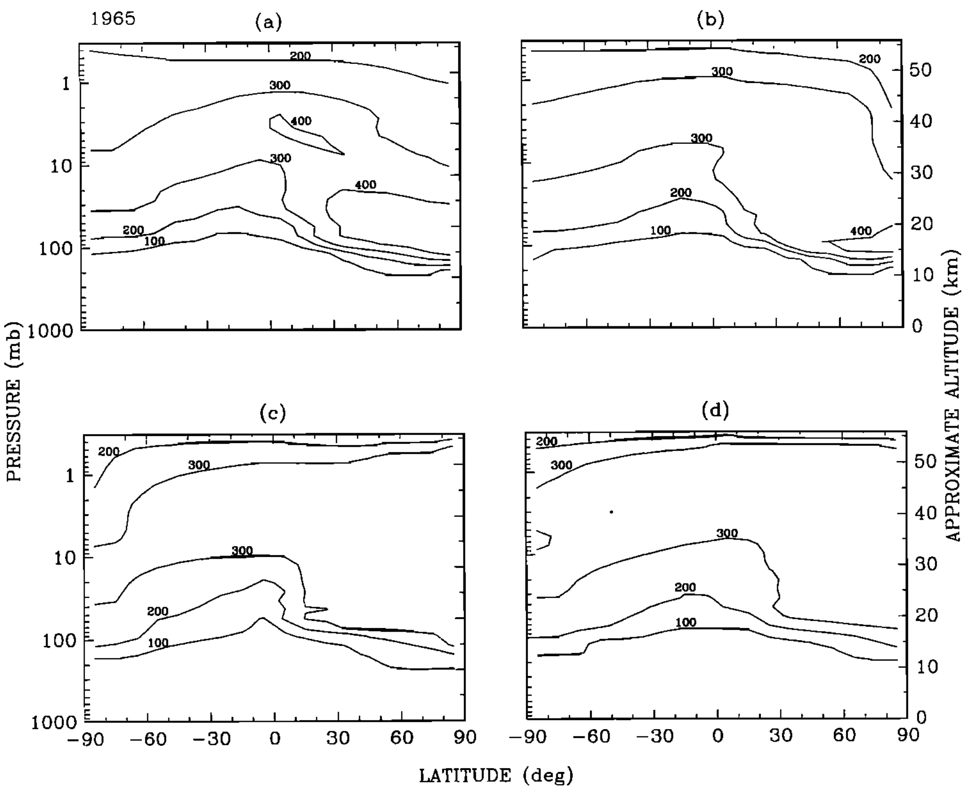

Fig. 7. Same as Figure 6 for 1965.

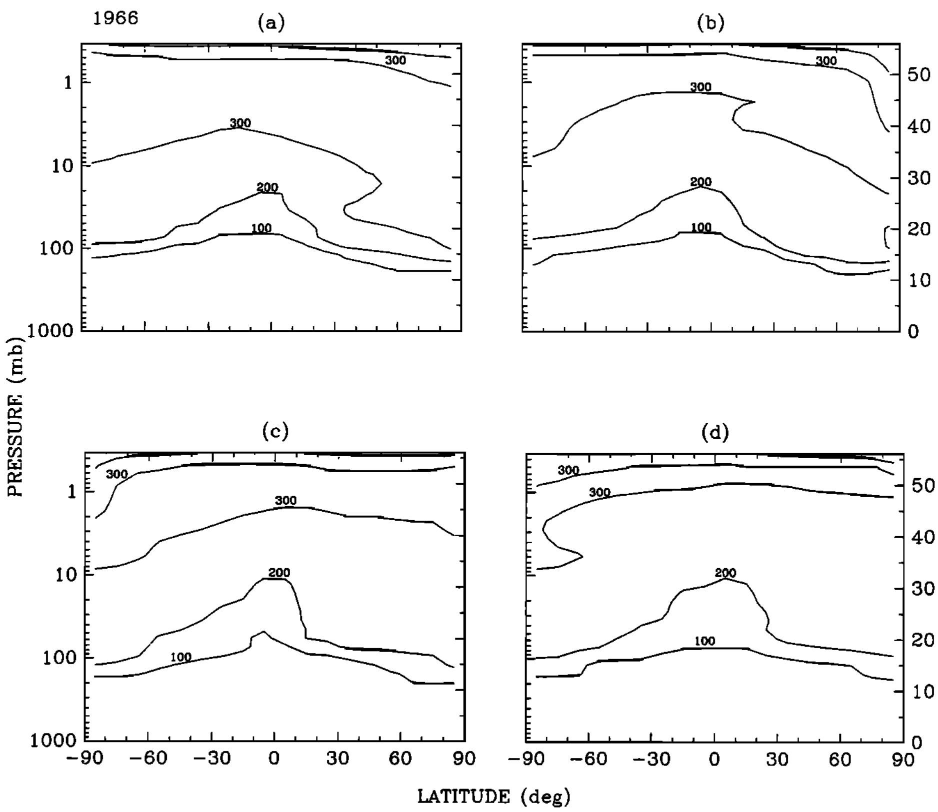

Fig. 8. Same as Figure 6 for 1966.

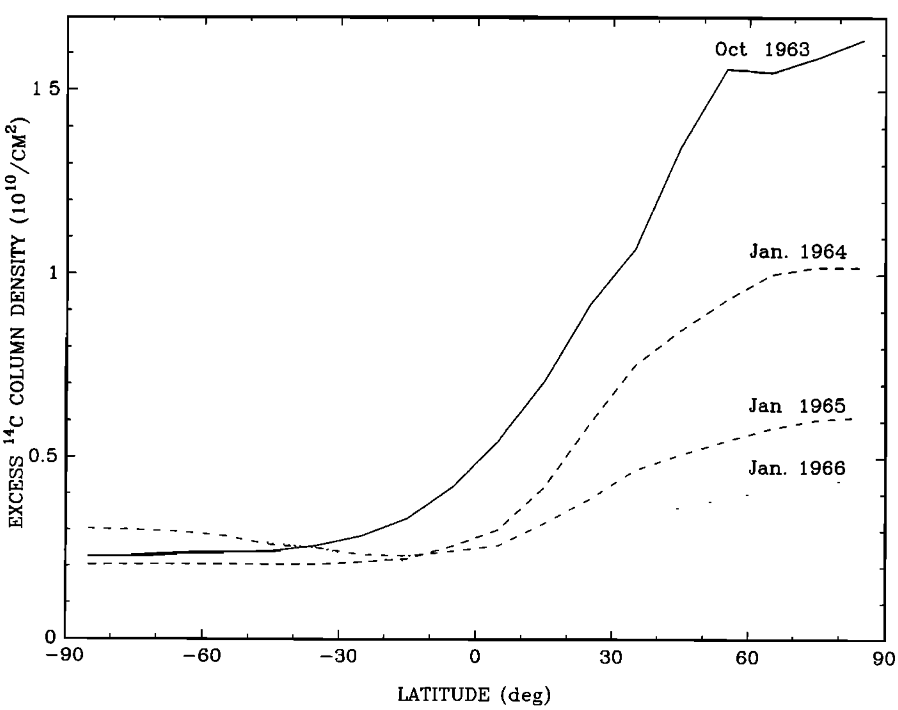

Fig. 9. Column density of excess ^{14}C above 10 km for October, 1963, and January, 1964–1966 in the standard model.

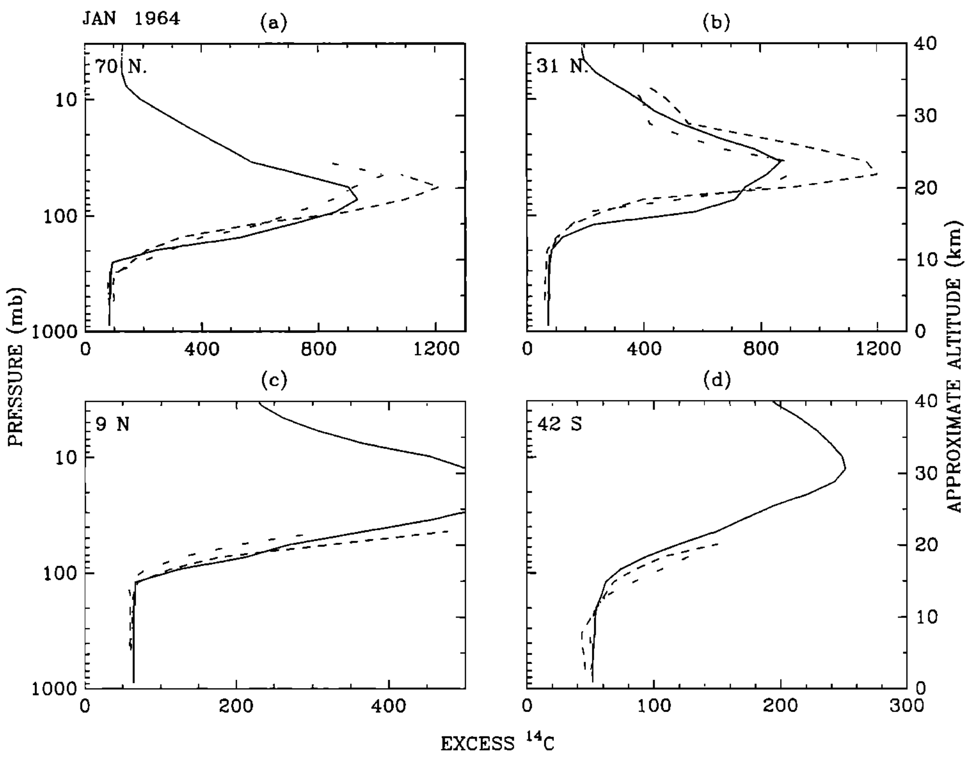

Fig. 10. Comparison of standard model and data for January, 1964 at 70°N, 31°N, 9°N, and 42°S. The units are same as in Figure 1. Model is represented by solid lines, initial data (October, 1963) by dashed lines, and January, 1964 data by dash-dot lines.

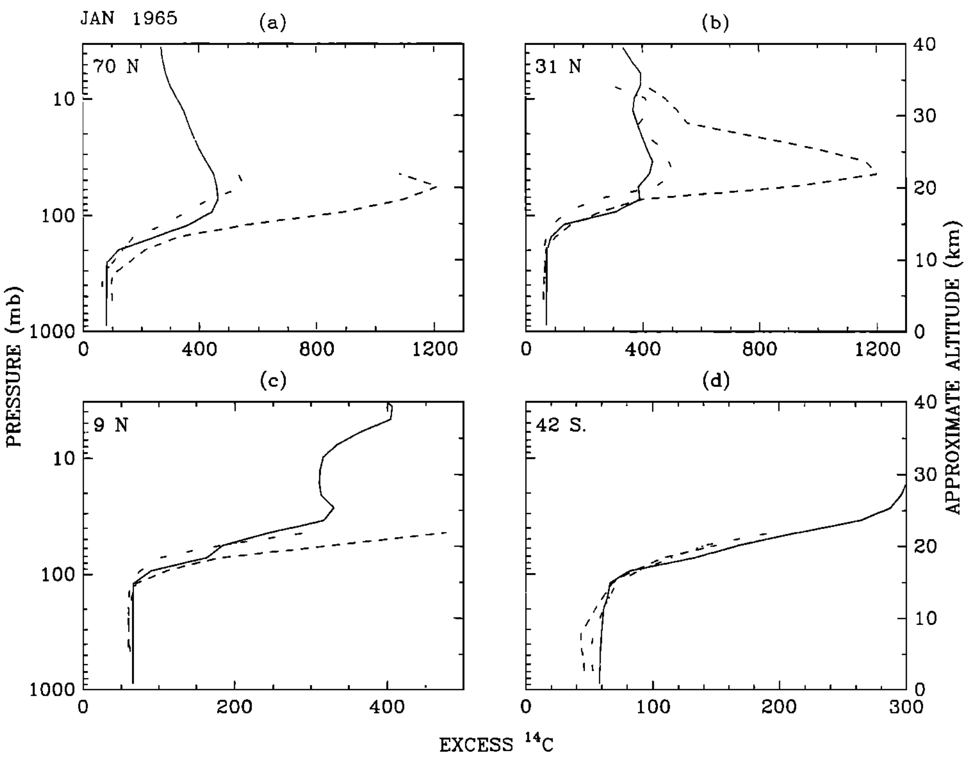

Fig. 11. Same as Figure 10 for January, 1965.

sumes a horizontal structure resembling those of NO_y and O_3 with contour lines of constant mixing ratio lying parallel to the tropopause. The original hemispheric asymmetry has largely disappeared.

Quantitative comparison between modeling and data is shown for January, 1964 for selected latitudes 70°N, 31°N, 9°N, and 42°S in Figures 10*a*–10*d*. The dashed lines represent the initial distribution (October, 1963). The dash-dot lines represent the observed data (Johnston, 1988), and the solid lines are model computations. The comparison for later years, January, 1965–1966 is shown in Figures 11*a*–11*d* and 12*a*–12*d*. In general, the agreement is good. This is significant because the transport coefficients used in the model have been previously validated by *Tung and Yang* [1988] and *Yang and Tung* [1989] for modeling stratospheric O_3. We have not attempted any "tuning" except for adjusting the surface deposition velocity. Unfortunately, the lack of a complete data base for excess ^{14}C does not permit a critical test of our model transport in the upper stratosphere ($z > 30$ km) or at high latitudes in the southern hemisphere.

Sensitivity Tests

A large number of model runs have been performed to test the sensitivity of our results to the transport coefficients. We start with the standard model as described in the previous section, and repeat the runs by varying one set of transport coefficients at a time. The different test cases are summarized in Table 2, along with brief comments on how the results compare with the observations.

In model A1, we increase the stream function uniformly in the atmosphere by a factor of 2 to produce a faster circulation. This run clearly shows that the advection is too fast, as can be seen from the comparison between model and observation in 1964 presented in Figures 13*a*–13*d*. The loss of material from the northern hemisphere is too great (see Figures 13*a* and 13*b*). The results of the later years (not shown) reinforce this conclusion. For instance, in 1966 the model has generally half the observed excess ^{14}C. The stream function is halved in model A2 to create a weaker circulation. The results for 1964 are shown in Figures 14*a*–14*d*. Note that in this case the results are not in serious conflict with observations, except perhaps near the tropopause at 31°N, where the model profile does not have enough upwelling. The results of the later years (not shown) give values that are about 30% too high when compared with observations, suggesting that the reduced circulation cannot satisfactorily account for the loss of excess ^{14}C from the stratosphere.

What is the implication of these runs for other two-dimensional models? We shall make a crude assessment. In our standard model the maximum upwelling velocity at 100 mbar is $w_{max} = 2.9 \times 10^{-2}$ cm s^{-1}. By comparison, this velocity is 0.1 cm s^{-1} in the work by *Murgatroyd and Singleton* [1961]. But most models [*Guthrie et al.*, 1984; *Ko et al.*, 1985; *Stordal et al.*, 1985] use the heating rates of Murgatroyd and Singleton, scaled down by a factor of 0.4. Thus, the corresponding w_{max} is about 4×10^{-2} cm s^{-1}. *Garcia and Solomon* [1983] prescribe vertical velocities at 100 mbar in their model, with $w_{max} = 7 \times 10^{-2}$ cm s^{-1}. Thus, in model A1 we have $w_{max} = 5.8 \times 10^{-2}$ cm s^{-1}, a value intermediate between most groups and Garcia and Solomon. Our work suggests that the advection used by Garcia and Solomon may be too large. However, the exchange of mass between the troposphere and the stratosphere depends on the width of the Brewer-Dobson circulation and seasonal variations as well. Therefore, no definitive statements can

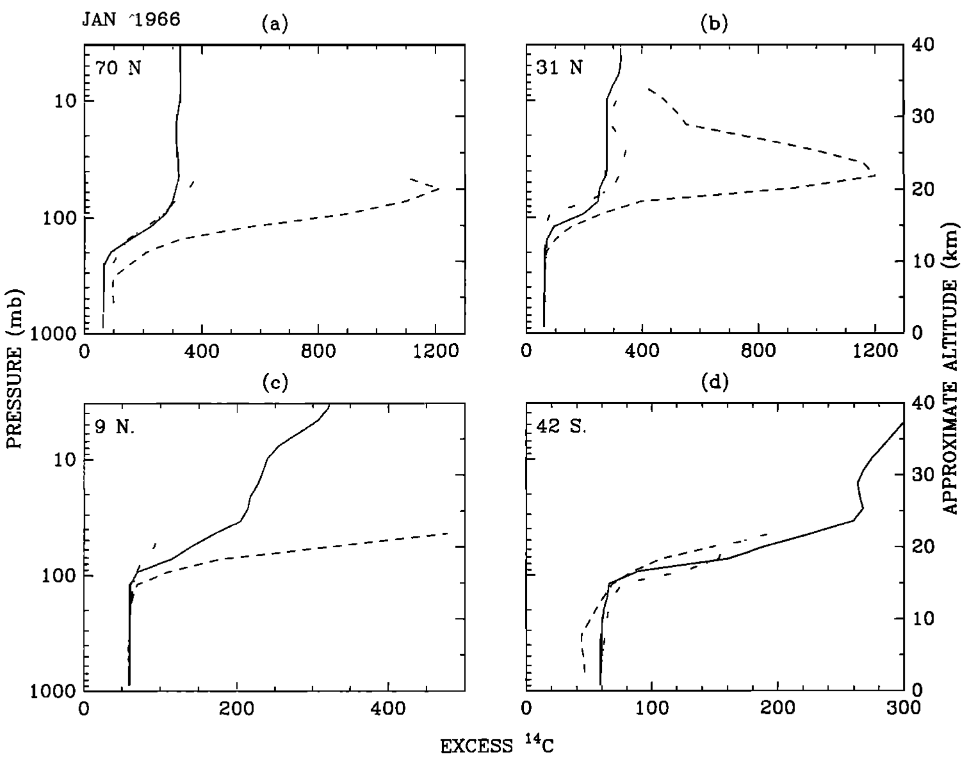

Fig. 12. Same as Figure 10 for January, 1966.

TABLE 2. Summary of Runs Performed to Test the Sensitivity of our Model to Variations in the Transport Coefficients

Model	Change Made*	Remarks
A1	$\psi \times 2$	too fast (Figure 13)
A2	$\psi \div 2$	marginal (Figure 14)
B1	stratospheric $K_{yy} = 1 \times 10^{10}$ cm^2 s^{-1}	too diffusive (Figure 15)
B2	stratospheric $K_{yy} = 1 \times 10^9$ cm^2 s^{-1}	not enough diffusion (Figure 16)
B3	stratospheric $K_{yy} = 3 \times 10^9$ cm^2 s^{-1}	cannot reproduce standard model
B4	stratospheric $K_{yy} = 1 \times 10^9$ cm^2 s^{-1} $\lvert\theta\rvert < 30°$ $K_{yy} = 1 \times 10^{10}$ cm^2 s^{-1} $\lvert\theta\rvert > 30°$	reproduces standard model (Figure 17)
C1	stratospheric $K_{zz} = 1 \times 10^3$ cm^2 s^{-1}	little impact
C2	stratospheric $K_{zz} = 1 \times 10^4$ cm^2 s^{-1}	too diffusive (Figure 18)
C3	stratospheric $K_{zz} = 1 \times 10^4$ cm^2 s^{-1} $\lvert\theta\rvert < 20°$ $K_{zz} = 1 \times 10^2$ cm^2 s^{-1} $\lvert\theta\rvert > 20°$	too diffusive
D1	ψ and K_{yy} derived for 1979	a little too strong (Figures 19–21)

*Everything remains the same as in the standard model unless otherwise stated.

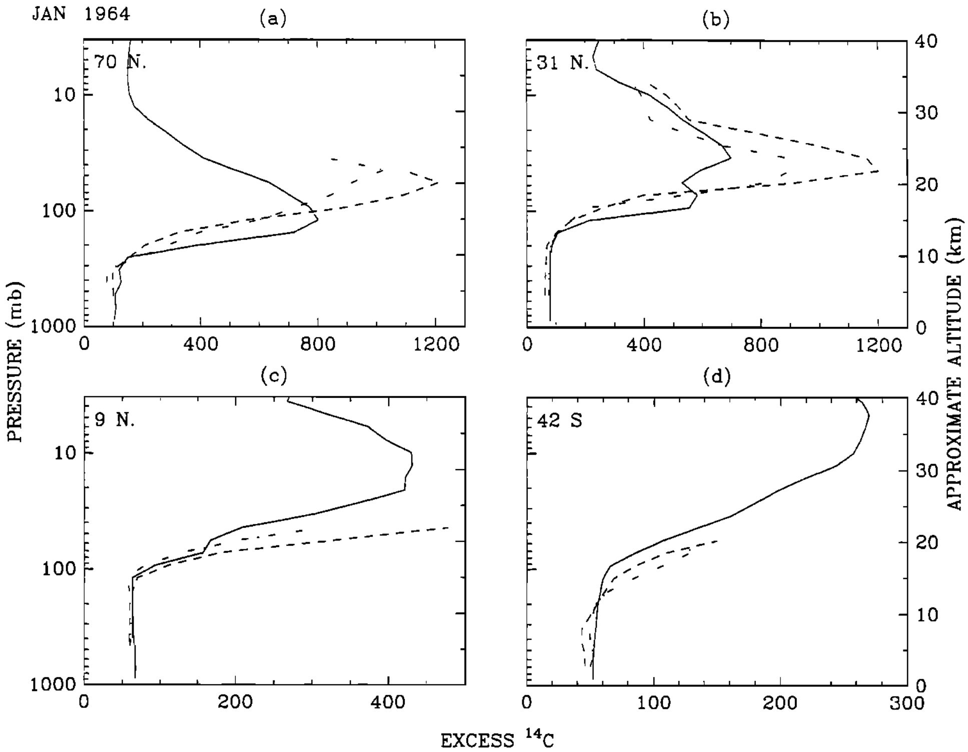

Fig. 13. Same as Figure 10 for model A1 using $\psi \times 2$.

be made until these model groups have carried out their own simulations of the excess ^{14}C data. The implication of model A2 is well known. A circulation that is half as strong as our standard model is obviously too sluggish because it would not be able to simulate the equatorial ozone minimum (see the detailed discussion of this constraint by *Tung and Yang* [1988]).

In model B1, the values of K_{yy} in the stratosphere are set equal to 1×10^{10} cm^2 s^{-1}. The results for January, 1964 are shown in Figures 15*a*–15*d*. The rate of loss of material from the peak region is clearly too fast (see Figure 15*b* for 31°N). The results for the later years (not shown) show generally poor agreement with the observations. Model B2 is similar to model B1 except that $K_{yy} = 1 \times 10^9$ cm^2 s^{-1} in the stratosphere. The results for January, 1964 are given in Figures 16*a*–16*d*. In this case, it is clear that the downward air motion (advection) dominates the lateral diffusion. As illustrated in Figure 16*a*, the excess ^{14}C peak, while having the correct magnitude, is displaced downward by as much as 3 km. The results of these models are consistent with those of *Ko et al.* [1985], who conclude that a smaller K_{yy} allows more HNO_3 to descend deeper into the lower polar

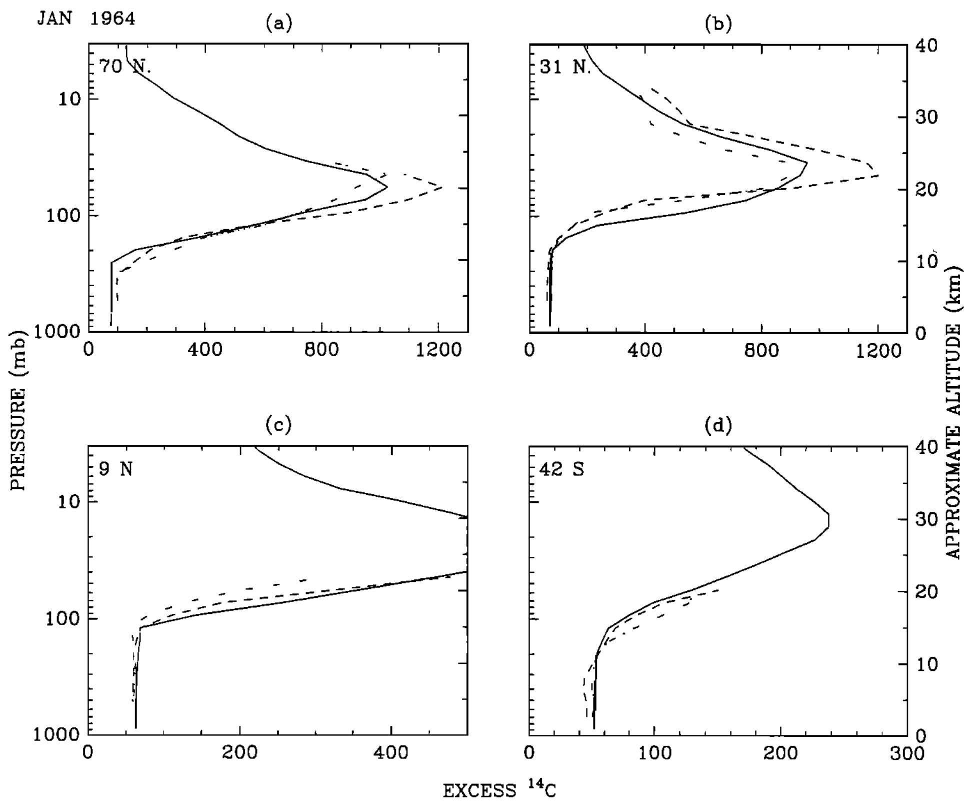

Fig. 14. Same as Figure 10 for model A2 using $\psi \div 2$.

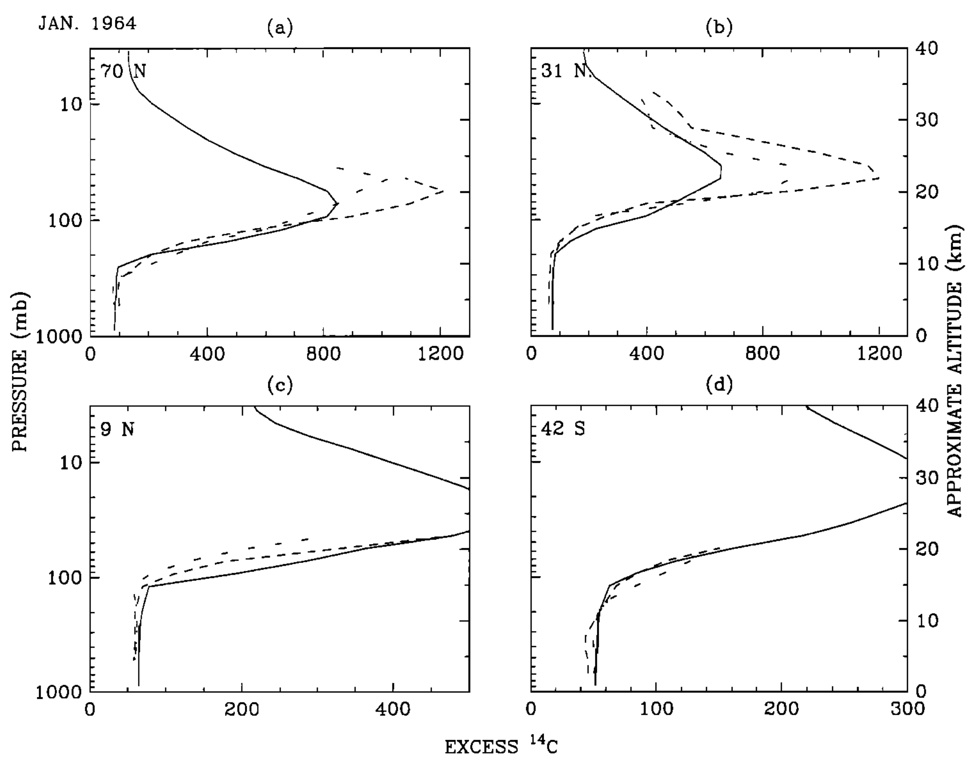

Fig. 15. Same as Figure 10 for model B1 with stratospheric $K_{yy} = 1 \times 10^{10}$ cm^2 s^{-1}.

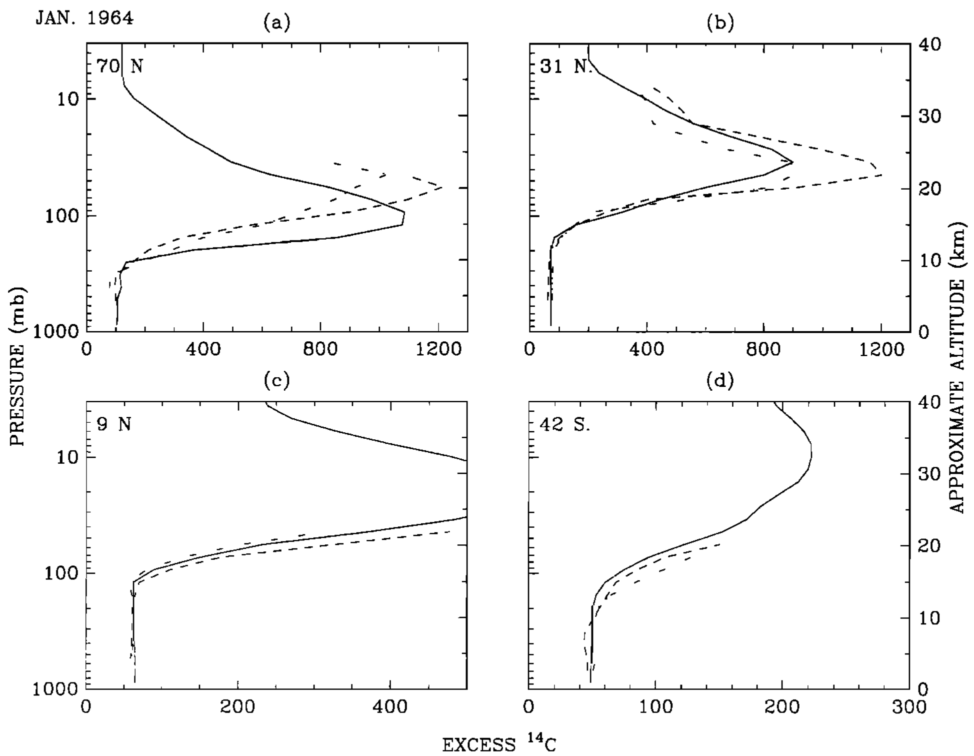

Fig. 16. Same as Figure 10 for model B2 with stratospheric $K_{yy} = 1 \times 10^9$ cm^2 s^{-1}.

stratosphere, thus creating a greater latitudinal contrast in the column abundances. No single constant value of stratospheric K_{yy} in the range 1–10 $\times 10^9$ cm^2 s^{-1} can reproduce the overall good agreement between our standard model and the observations. The reason is obvious from models B1 and B2. The low latitude data require a low K_{yy} and the high latitude data require a high K_{yy}. A model such as model B3, with $K_{yy} = 3 \times 10^9$ cm^2 s^{-1}, cannot satisfy both demands. This provides support for the nonconstant K_{yy} derived by the self-consistent theory [*Tung*, 1984, 1986; *Plumb and Mahlman*, 1987; *Newman et al.*, 1988; *Yang and Tung*, 1989], a result independently tested here using a very sensitive tracer.

Figures 5*a*–5*d* suggest that K_{yy} is larger in the tropics. A bimodal value of stratospheric K_{yy} equal to 1×10^9 cm^2 s^{-1} for latitudes below 30° and 1×10^{10} cm^2 s^{-1} for higher latitudes (model B4) can closely reproduce our standard model for all years. The results for January, 1964 are shown in Figures 17*a*–17*d*. In the model of *Ko et al.* [1985] a constant stratospheric K_{yy} is used. A bimodal K_{yy} like that of model B4 can probably improve the agreement of their latitudinal distribution of HNO_3. However, we must caution the reader not to regard this as a substitute for the self-consistent K_{yy}, because of the sparse data base on which this bimodal K_{yy} rests. Any other K_{yy} with a small value in the equatorial lower stratosphere, and a higher value at high latitudes, where the Brewer-Dobson circulation is descending, will probably reproduce model B4.

Although there are no theoretical reasons for a large K_{zz} in the stratosphere, it is of interest to set an upper limit for K_{zz}. In model C1 we set the stratospheric $K_{zz} = 1 \times 10^3$ cm^2 s^{-1}, which is 10 times the value in the standard model. The results are virtually the same as in the standard model, and hence are not shown. A higher value $K_{zz} = 1 \times 10^4$ cm^2 s^{-1} is used in model C2. The results for January, 1964 are presented in Figure 18. The 70°N and 31°N (Figures 18*a* and 18*b*) comparisons clearly show that this model is too diffusive. The loss of excess ^{14}C in later years (not shown) is also very severe, and by 1966 the computed excess ^{14}C is only about half the observed value. Thus we conclude that K_{zz} must be less than 5×10^3 cm^2 s^{-1} in the stratosphere. Most "modern" two-dimensional models have K_{zz} less than 3×10^3 cm^2 s^{-1}, with the exception of two models. *Garcia and Solomon* [1983] use a K_{zz} that increases steeply with altitude. Below 26 km $K_{zz} < 1 \times 10^4$ cm^2 s^{-1}, but rises to $> 1 \times 10^5$ cm^2 s^{-1} above 40 km. This choice of K_{zz} may be too high to be consistent with our data. *Pitari and Visconti* [1985] derive their residual mean circulation and eddy diffusion coefficients from the MIT–GIT GCM. They report values of $K_{zz} > 1 \times 10^4$ cm^2 s^{-1} in the equatorial lower stratosphere. In model C3 we attempt to represent their K_{zz} by a bimodal value, $K_{zz} = 1 \times 10^4$ cm^2 s^{-1} for $|\theta| < 20°$ and $K_{zz} = 1 \times 10^2$ cm^2 s^{-1} for $|\theta| > 20°$. The results suggest that K_{zz} as large as 1×10^4 cm^2 s^{-1} concentrated only in the tropics is too diffusive.

As mentioned earlier, the choice of using the circulation and K_{yy} of 1980 to simulate the transport of excess ^{14}C in the years 1963–1966 is arbitrary and is dictated mostly by the availability of stratospheric temperature data in the NMC series. In fact, 1980 corresponds to a year of relatively weak stratospheric wave activity. In model D1 we repeat the calculations with the circulation and K_{yy} of 1979

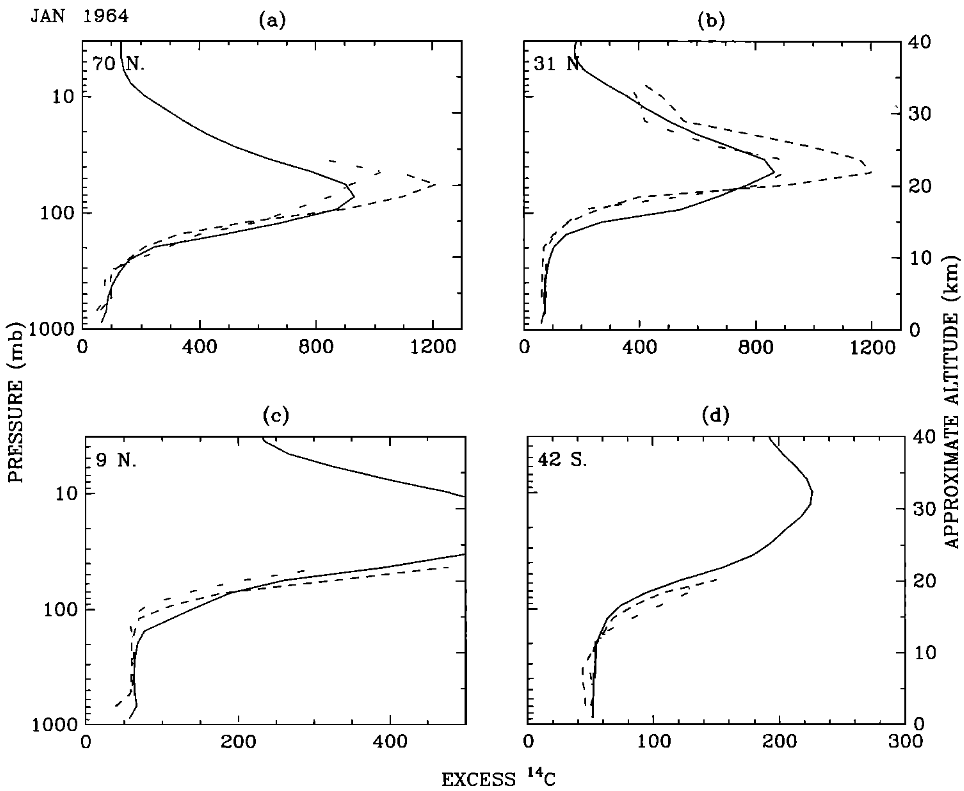

Fig. 17. Same as Figure 10 for model B4, with stratospheric $K_{yy} = 1 \times 10^9$ cm^2 s^{-1} for $|\theta| < 30°$, and $K_{yy} = 1 \times 10^{10}$ cm^2 s^{-1} for $|\theta| > 30°$.

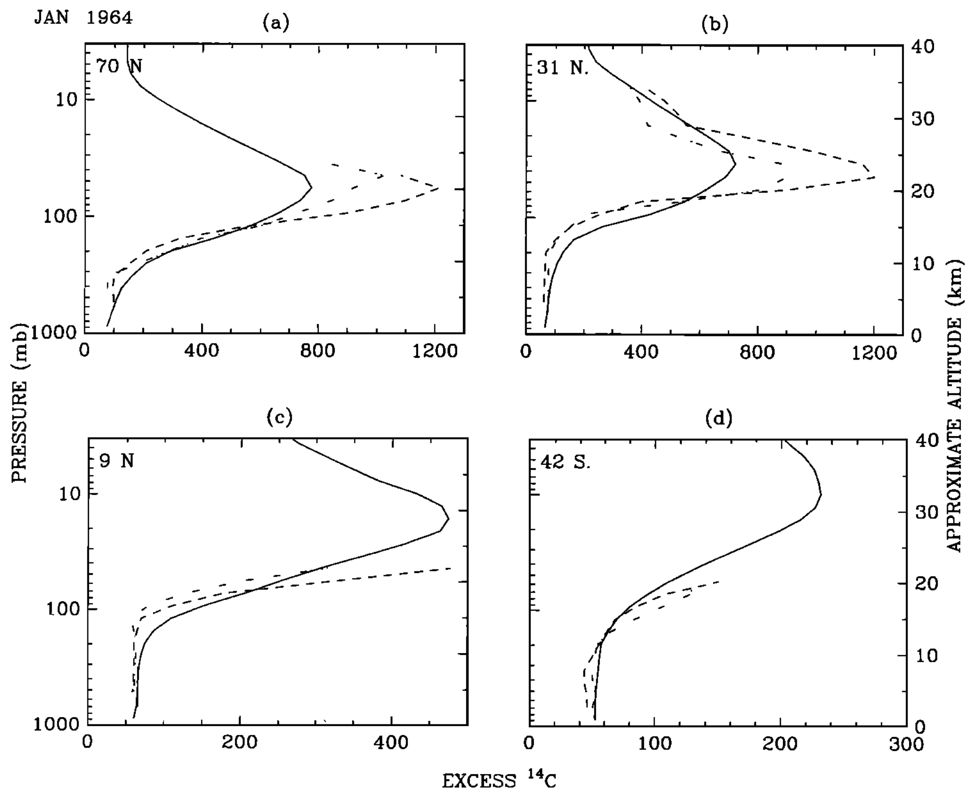

Fig. 18. Same as Figure 10 for model C2, with stratospheric $K_{zz} = 1 \times 10^4$ cm^2 s^{-1}.

[*Tung and Yang*, 1988], a year of very strong stratospheric wave activity. The results, summarized in Figures 19*a*–19*d*, 20*a*–20*d*, and 21*a*–21*d* for January, 1964, 1965, and 1966, respectively, suggest that although there are differences in the simulations using 1979 and 1980 transport coefficients, these cannot be distinguished using the current data base. We have also carried out simulations using transport coefficients derived from the GFDL three-dimensional model (Zurek et al., 1989). The results are not as good as those based on *Yang and Tung* [1989], again demonstrating the sensitivity of our model to different input parameters. However, a detailed comparison of this model with other models will be referred to Zurek et al. (1989).

TROPOSPHERE

The main thrust of this work is modeling of the stratosphere. However, correct modeling of tropospheric excess ^{14}C is crucial for a correct treatment of its stratospheric abundances. We believe that this is the first time that excess ^{14}C has been traced from its source in the stratosphere to its ultimate sink on the surface. The question arises as to what determines the abundance of excess ^{14}C near the surface. Clearly the essential elements in this question are the surface deposition velocity and the vertical transport and the interhemispheric exchange (parameterized here by a tropospheric K_{zz} and K_{yy}, respectively).

Sensitivity runs are summarized in Table 3. We choose to examine the results in January, 1966, by which time the model has reached a quasi-steady state between the stratosphere and the troposphere. In model T1, the surface deposition velocities v_S and v_N are set to zero. The computed excess ^{14}C at 5 km at 70°N, 31°N, 9°N, and 42°S are 98, 91, 89, and 84 units, respectively. Therefore, the prediction is too high compared with observations (see Table 3). In model T2, we raise the deposition velocities to $v_S = v_N = 1 \times 10^{-2}$ cm s^{-1}. In this case, the predicted values for excess ^{14}C are too low when compared with observations. In model T3, we set $v_S = v_N = 3 \times 10^{-3}$ cm s^{-1}. This case is fairly close to the observations, but there is apparently too much excess ^{14}C in the northern hemisphere. In model T4, we test the sensitivity of our results to a smaller equatorial $K_{yy} = 3 \times 10^9$ cm^2 s^{-1} in the troposphere. This run is not as good as the standard run and suggests that an interhemispheric barrier in the transport will enhance the excess ^{14}C gradient [see *Pinto et al.*, 1983]. Model T5 tests the sensitivity of our results to a weaker vertical eddy diffusivity $K_{zz} = 3 \times 10^4$ cm^2 s^{-1} in the southern hemisphere. Again, this results in a greater hemispheric asymmetry in excess ^{14}C. The need for asymmetric v_S and v_N can obviously be removed by a large K_{yy}. In model T6, we set $K_{yy} = 3 \times 10^{10}$ cm^2 s^{-1} and $v_S = v_N = 4 \times 10^{-3}$ cm s^{-1}. The results for January, 1966 (see Table 3) are marginally acceptable. However, there are two flaws with this model. First, the time constant for diffusion over global scale as computed by (13) is now only 5.2 months. This is much shorter than the known interhemispheric exchange time. Second, the modeled tropospheric excess ^{14}C in 1964 and 1965 at 42°S are much larger than the observed values. The results summarized in Table 3 are not intended to be a definitive study of tropospheric excess ^{14}C, but rather to provide some insight into how the transport parameters

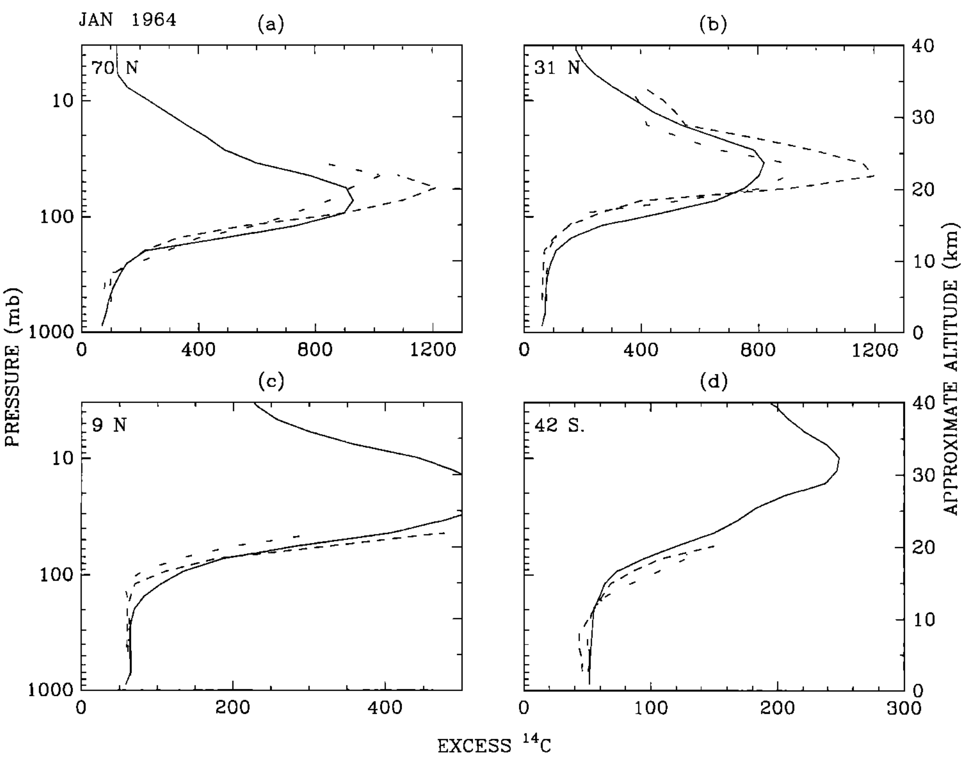

Fig. 19. Same as Figure 10, for model D1, and January, 1964 using 1979 ψ and K_{yy}.

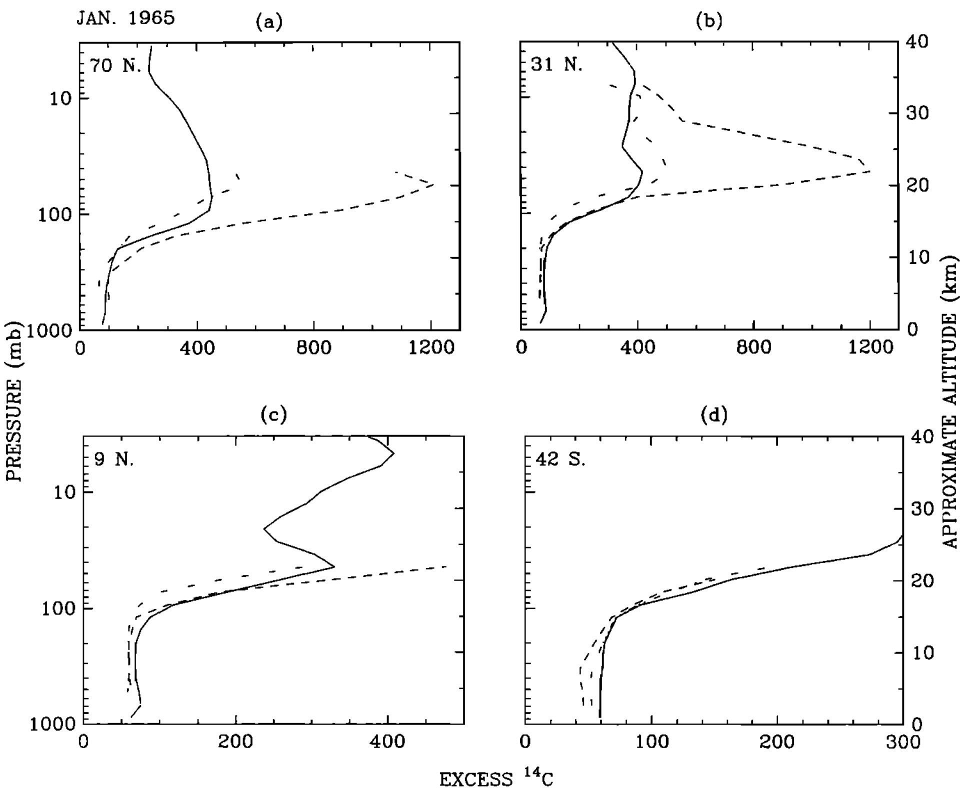

Fig. 20. Same as Figure 19, for January, 1965.

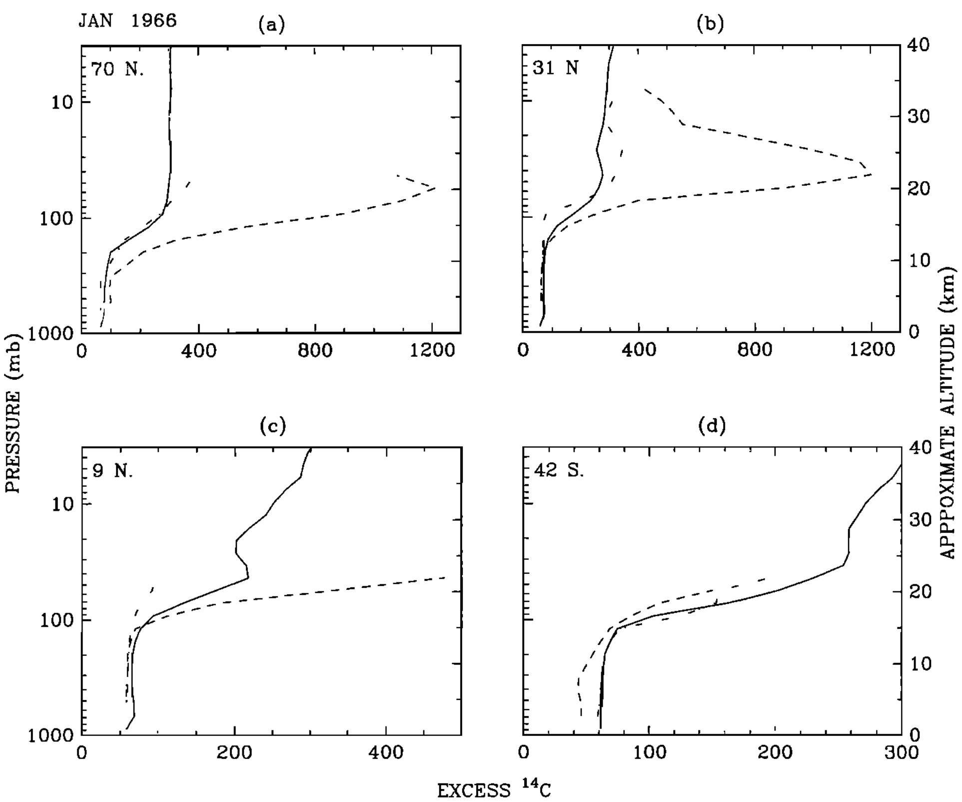

Fig. 21. Same as Figure 19, for January, 1966.

TABLE 3. Summary of Model Runs Performed to Test the Sensitivity of Our Model to Surface Deposition Velocity and Tropospheric K_{yy} and K_{zz}

Model	Change	Excess ^{14}C 70°N	31°N	9°N	42°S	Remarks
T1	$v_S = v_N = 0$	98	91	89	84	Too high
T2	$v_S = v_N = 1 \times 10^{-2}$ cm s^{-1}	56	56	49	40	Too low
T3	$v_S = v_N = 3 \times 10^{-3}$ cm s^{-1}	84	77	70	65	Too much hemispheric asymmetry, too high
T4	Tropospheric $K_{yy} = 3 \times 10^9$ cm^2 s^{-1} $\|\theta\| < 20°$	84	84	76	56	Too much hemispheric gradient, too high
T5	Tropospheric $K_{zz} = 3 \times 10^4$ cm^2 s^{-1} $\theta < 0°$	84	70	70	63	Too much hemispheric gradient, too high
T6	$v_S = v_N = 4 \times 10^{-3}$ cm^2 s^{-1} Tropospheric $K_{yy} = 3 \times 10^{10}$ cm s^{-1}	70	69	65	61	Marginal
Standard Model		67	67	59	58	
Observation		65	66	59	59	Johnston (1988)

All model parameters are the same as in the standard model except otherwise stated. For comparison, the deposition velocities in the standard model are $v_S = 3 \times 10^{-3}$ cm s^{-1} and $v_N = 5 \times 10^{-3}$ cm s^{-1} in the southern and northern hemisphere, respectively. Tropospheric values of excess ^{14}C at 5 km are given for 70°N, 31°N, 9°N, and 42°S in January, 1966. For comparison, the observed values at this time, and the results of the standard model are presented in the last rows.

are chosen in the standard model. The most puzzling conclusion is the hemispheric asymmetry of v_S and v_N, which is opposite to what is expected if the oceans are the major sink for ^{14}C [*Lal and Suess*, 1968]. If our conclusion is verified, it will have a profound implication for the global carbon budget. Further work is obviously necessary to assess the quality of the tropospheric data, and to improve the tropospheric transport in our model.

CONCLUSIONS

We have studied the distribution of excess ^{14}C from October, 1963 to December, 1966 using our two-dimensional model with transport coefficients taken from *Yang and Tung* [1989]. The model successfully accounts for the observations (Johnston, 1988). Excess ^{14}C is a unique tracer of atmospheric motion because it is derived from an impulsive source, and because it has initially large spatial gradients. Useful limits on the stream function and horizontal eddy diffusivity are obtained for the lower stratosphere as summarized in Table 2. We show that the data require a variable K_{yy} which is smaller in the tropics and larger at higher latitudes, consistent with the predictions of *Yang and Tung* [1989]. This is the first time that excess ^{14}C has been modeled from its source in the stratosphere to its ultimate sink on the surface. The deposition velocities, empirically derived in this study, are generally consistent with the rate of exchange between the atmosphere and the ocean, although a slight asymmetry between the hemispheres is puzzling (see Table 3). This opens up an exciting possibility of tracing the ultimate fate of excess ^{14}C to its biospheric sinks on land and in the oceans.

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