

## A Model for Diffusion of Radioactive Debris in the Troposphere

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### ABSTRACT

A theoretical model is constructed and tested for the analysis and prediction of radioactive concentration in the troposphere. It is found that turbulent motion near the jet core plays the major role in the transport of radioactive debris from the stratosphere into the troposphere, whereas the mean motion of the jet core contributes to the spring maximum and autumn minimum of the concentration. A semiannual period in the variation of concentration exists, resulting from the interaction between the meridional gradient of the mean concentration and the mean motion of the jet core. It is also found that the average value of the vertical component of the eddy diffusivity in the troposphere is about  $10^7 \text{ cm}^2 \text{ sec}^{-1}$ , and that the time required for diffusing radioactive particles from the tropopause level to the surface of the earth is about 11 hours.

### 1. Introduction

It is generally believed that the tropopause in the atmosphere acts as a barrier to the vertical transport of radioactive debris and that the tropopause-break, which coincides with the core of the jet stream, represents the gateway for the injection of radioactive debris from the stratosphere into the troposphere. However, little study has been made of the role played by the tropopause-break on the distribution of radioactive concentration in the troposphere. It has been known for sometime that radioactive concentration in the troposphere has its maximum in spring and its minimum in autumn, yet little is known of the transport processes which contribute to the time variation in the radioactive concentration in the troposphere. To investigate these problems, knowledge of the mechanism of turbulence and the processes of transport in the atmosphere is needed. The characteristics of large-scale horizontal turbulence and diffusion in the troposphere have recently been studied (Kao, 1962, 1964, 1965; Kao and Bullock, 1964). Our understanding, however, of the large-scale turbulence and diffusion along the vertical in the troposphere is very meager. The purposes of this investigation are to construct and test a diffusion model, which may be used for the analysis of the mechanism of turbulence and for the prediction of the distribution of radioactive concentration in the troposphere.

### 2. Diffusion of radioactive particles in the troposphere

We assume that the mean tropopause-break is oriented in the east-west direction and acts as a line source. The diffusion equation in such a system may be

written as

$$\frac{\partial \bar{C}}{\partial t} = \frac{\partial}{\partial y} \left( K_{yy} \frac{\partial \bar{C}}{\partial y} \right) + \frac{\partial}{\partial y} \left( K_{yz} \frac{\partial \bar{C}}{\partial z} \right) + \frac{\partial}{\partial z} \left( K_{zy} \frac{\partial \bar{C}}{\partial y} \right) + \frac{\partial}{\partial z} \left( K_{zz} \frac{\partial \bar{C}}{\partial z} \right) - \bar{v} \frac{\partial \bar{C}}{\partial y} - \bar{w} \frac{\partial \bar{C}}{\partial z}, \quad (1)$$

where the positive  $y$  and  $z$  axes are directed toward the north and zenith respectively, with the origin of the coordinate system located at mean sea level right beneath the mean tropopause-break.  $K_{ij}$  is the eddy diffusion tensor, and  $\bar{C}$ ,  $\bar{v}$  and  $\bar{w}$  are, respectively, the longitudinal mean of the concentration, and the  $y$ - and  $z$ -components of the air velocity averaged over a time interval  $T$ . Thus,

$$\bar{C}(y, z, T) = \frac{1}{2\pi T} \int_0^T \int_0^{2\pi} C(x, y, z, t) dx dt, \quad (2)$$

where  $x$  is the longitude.

For a comparatively long time interval, say a year or more, the values of  $\bar{v}$  and  $\bar{w}$  are generally very small as compared with the turbulent velocity, and the terms containing these quantities in (1) may be considered as second order terms and neglected as a first approximation. The effect of these mean advective terms on the mean concentration in the troposphere will be considered in a later section. Within the period of time mean, the mean concentration is a constant and is, therefore, independent of time. Furthermore, if we assume that the principal axes of the eddy diffusion are parallel to the  $y$  and  $z$  axes, and that  $K_{yy}$  and  $K_{zz}$  are independent

of the coordinates, the diffusion equation (1) becomes

$$K_y \frac{\partial^2 \bar{C}}{\partial y^2} + K_z \frac{\partial^2 \bar{C}}{\partial z^2} = 0, \tag{3}$$

where  $K_y$  and  $K_z$  are the abbreviations for  $K_{yy}$  and  $K_{zz}$ , respectively.

It can be shown that the solution of the above differential equation takes the form

$$\bar{C}(y,z,T) = A + B \ln \left[ \frac{y^2}{K_y} + \frac{(z-H)^2}{K_z} \right], \tag{4}$$

where  $A$  and  $B$  are constants and  $H$  is the mean height of the jet core. Eq. (4) applies to the region in the troposphere, except in the region of the tropopause-break.

Eq. (4) may further be expressed in terms of dimensionless variables  $y/H$  and  $z/H$ , i.e.,

$$\bar{C}(y,z,T) = R + S \ln \left[ \frac{K_y}{K_z} \left( \frac{z}{H} - 1 \right)^2 + \left( \frac{y}{H} \right)^2 \right], \tag{5}$$

where  $R = A + 0.5 B \ln(H^2/K_y)$  and  $S = 0.5 B$ .

For convenience in comparing data for different periods with the solution derived from the model, we introduce a normalized concentration,  $\bar{C}_N$ , defined as the ratio of concentration to its maximum value at the same level, i.e., at  $y=0$ . Thus,

$$\bar{C}_N(y,z,T) = \frac{\bar{C}(y,z,T)}{\bar{C}(0,z,T)}. \tag{6}$$

Substitution of (5) into the above equation gives

$$\bar{C}_N(y,z,T) = \frac{R + S \ln \left[ \frac{K_y}{K_z} \left( \frac{z}{H} - 1 \right)^2 + \left( \frac{y}{H} \right)^2 \right]}{R + S \ln \left[ \frac{K_y}{K_z} \left( \frac{z}{H} - 1 \right)^2 \right]}. \tag{7}$$

It is convenient to express the constants  $R$  and  $S$  in (7) in terms of quantities which can readily be determined from the values of concentration measured at the surface. The normalized mean concentration at the surface may be written

$$\bar{C}_N(y,0,T) = \frac{\bar{C}(y,0,T)}{\bar{C}(0,0,T)} = N + P \ln \left[ \frac{K_y}{K_z} + \left( \frac{y}{H} \right)^2 \right], \tag{8}$$

where

$$N = \frac{R}{\bar{C}(0,0,T)}, \quad P = \frac{S}{\bar{C}(0,0,T)} \tag{9}$$

can easily be determined from the observed mean concentration at the surface.

Since  $N$  and  $P$  are dimensionless quantities, their values determined from measured surface concentration averaged over a long time interval may be considered as constants. Using (9), the normalized mean concentration (7) may then be expressed as

$$\bar{C}_N(y,z,T) = \frac{N + P \ln \left[ \frac{K_y}{K_z} \left( \frac{z}{H} - 1 \right)^2 + \left( \frac{y}{H} \right)^2 \right]}{N + P \ln \left[ \frac{K_y}{K_z} \left( \frac{z}{H} - 1 \right)^2 \right]}. \tag{10}$$

This equation will be used for the construction of a nomogram of the normalized mean concentration expressed in terms of the dimensionless lengths  $y/H$  and  $z/H$ .

The mean concentration may then be expressed in terms of the mean surface concentration right beneath the jet core, i.e.,

$$\bar{C}(y,z,T) = \bar{C}(0,0,T) \left\{ N + P \ln \left[ \frac{K_y}{K_z} \left( \frac{z}{H} - 1 \right)^2 + \left( \frac{y}{H} \right)^2 \right] \right\}. \tag{11}$$

To determine the values of  $N$  and  $P$  in (8), (10) and (11), the value of the mean surface concentration right beneath the tropopause-break is needed. In practice, this value is not easily determined, since the position of the jet core generally varies with time, whereas the positions of observation stations are fixed. For practical reason, we define a new dimensionless mean concentration,

$$\bar{C}_L(y,z,T) = \frac{\bar{C}(y,z,T)}{\langle \bar{C}(L,0,T) \rangle}, \tag{12}$$

where

$$\langle \bar{C}(L,0,T) \rangle = \frac{1}{L} \int_{-1/2L}^{1/2L} \bar{C}(y,0,T) dy \tag{13}$$

is the area mean of the surface mean concentration.

In view of (5), the new dimensionless mean concentration may be expressed as

$$\bar{C}_L(y,z,T) = G + M \ln \left[ \frac{K_y}{K_z} \left( \frac{z}{H} - 1 \right)^2 + \left( \frac{y}{H} \right)^2 \right], \tag{14}$$

where

$$G = \frac{R}{\langle \bar{C}(L,0,T) \rangle}, \quad M = \frac{S}{\langle \bar{C}(L,0,T) \rangle}. \tag{15}$$

Using the observed mean surface radioactive concentration over the United States for the years of 1963 and 1964, the two years' mean of the dimensionless surface concentration,  $\bar{C}(y,0,T) / \langle \bar{C}(L,0,T) \rangle$ , has been com-

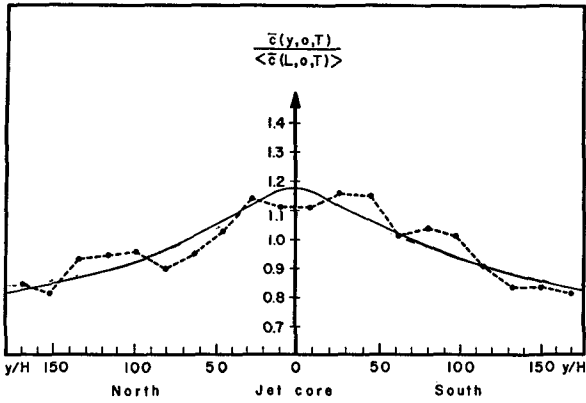


FIG. 1. Distribution of the dimensionless surface mean concentration relative to the jet core.

puted<sup>1</sup> and is shown by the dashed curve in Fig. 1. The solid curve in this figure represents the solution for diffusion from the line source located at the jet core, i.e.,

$$\bar{C}_L(y,0,T) = G + M \ln \left[ \frac{K_y}{K_z} + \left( \frac{y}{H} \right)^2 \right], \quad (16)$$

with  $G = 1.746$ ,  $M = -0.088$  and  $K_y/K_z = 800$ .

It may be noted in Fig. 1 that the model solution for the surface concentration shows a more or less bell-shape distribution with its maximum located right beneath the jet core. This theoretical curve gives general agreement between the observation and the model. However, deviations in the distribution of the mean surface concentration from the model exist as shown in the relative maxima and minima in Fig. 1. These deviations which are primarily the result of the mean advective motion will be explained in the following sections.

First, we consider the relative maximum concentration occurring approximately at  $y/H = 45$  south in Fig. 1, that is, about 400 km south of the jet core if an averaged jet core height of 9 km is assumed. This is the region of intersection between the frontal surface and the surface of the earth (Palmén and Newton, 1948). A study made by Staley (1962) indicates that frontal zones may be considered one of the gateways for the transport of radioactive debris from the lower stratosphere to the surface of the earth. This descending transport of radioactive debris gives a positive contribution to the term of  $-w\partial\bar{C}/\partial z$  in (1), which may account for the relative maximum concentration occurring about 400 km south of the jet core.

In his model of the mean meridional circulation, Palmén (1951) shows that the northern branch of the tropical cell descends to the surface of the earth around 800 km south of the jet core. This descending air current, which advects radioactive debris from the upper troposphere to the surface of the earth, may account

for the relative maximum concentration occurring at about  $y/H = 90$  south of the jet core in Fig. 1.

Furthermore, Palmén's (1951) mean meridional circulation model shows that another descending motion occurs north of the polar front at about 1000 km north of the jet core. This descending motion would also advect radioactive debris to the surface of the earth, which may account for the relative maximum concentration occurring about  $y/H = 120$  north of the jet core in Fig. 1. The relative minimum concentration occurring right south of the relative maximum may be the effect of the ascending motion resulting from the descending motion near 1000 km north of the jet core.

Let us now return to solution (16) and the solid curve in Fig. 1. It has been estimated in a previous paper (Kao and Bullock, 1964) that the lateral component of the eddy diffusivity  $K_y \approx 10^{10} \text{ cm}^2 \text{ sec}^{-1}$  at 500 mb, which represents an averaged value of the lateral eddy diffusivity in the troposphere. Using this value of  $K_y$  and the aforementioned value of 800 for the ratio between the lateral and vertical components of the eddy diffusivity, we find that in the troposphere the average vertical component of the eddy diffusivity is  $K_z \approx 10^7 \text{ cm}^2 \text{ sec}^{-1}$ .

The time required for radioactive particles diffusing from the jet core to the surface of the earth is then  $t \doteq H^2/2K_z \approx 11 \text{ hr}$ , where it is assumed that the mean height of the tropopause-break  $H = 9 \text{ km}$ . The time for radioactive debris transported from the jet core to the earth's surface through the frontal zone is of the order of 24 hr (Staley, 1962). Therefore, the process of the vertical turbulent diffusion in the troposphere is about twice as efficient as that of the isentropic transport of radioactive debris.

In view of the small time interval required for the vertical turbulent transport of radioactive debris from the jet core to the earth's surface, the previously mentioned values of  $G$ ,  $M$  and  $K_y/K_z$ , determined from the two years' mean of the observed radioactive concentration, may be considered constants. Using these constants the values of the mean dimensionless concentration  $\bar{C}_L$  in the troposphere have been computed from (14) for  $y/H$  at intervals of every 50 and for  $z/H$  at intervals of every 0.1, and are shown in the Appendix. It is found that the mean dimensionless concentration varies only about 2 per cent along the vertical in the troposphere, whereas it varies about 50 per cent in a distance of 4500 km along the horizontal.

It is convenient to express the normalized mean concentration (7) in terms of the dimensionless constants  $G$  and  $M$ , i.e.,

$$\bar{C}_N(y,z,T) = \frac{G + M \ln \left[ \frac{K_y}{K_z} \left( \frac{z}{H} - 1 \right)^2 + \left( \frac{y}{H} \right)^2 \right]}{G + M \ln \left[ \frac{K_y}{K_z} \left( \frac{z}{H} - 1 \right)^2 \right]}. \quad (17)$$

<sup>1</sup> Kao, S.-K., and W. J. Alder, 1966: Radioactive concentration in relation to the jet stream. Unpublished manuscript.

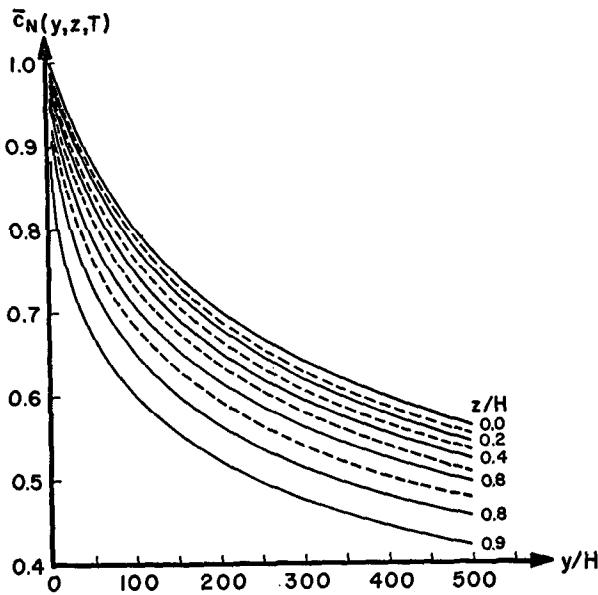


FIG. 2. Nomogram for the normalized mean concentration.

Using the above equation, the values of the normalized mean concentration have been computed for  $y/H$  at intervals of every 50 and for  $z/H$  at intervals of every 0.1, and are shown in Fig. 2. It is found that the normalized mean concentration generally decreases with increasing  $y/H$  and  $z/H$ .

3. Variation of radioactive concentration

For a time mean interval longer than that required for diffusing particles from the tropopause-break to the earth's surface, the mean concentration can be expressed as

$$\bar{C}(y,z,t) = \langle \bar{C}(L,0,t) \rangle \left\{ G + M \ln \left[ \frac{K_y}{K_z} \left( \frac{z}{H} - 1 \right)^2 + \left( \frac{y}{H} \right)^2 \right] \right\}, \tag{18}$$

where  $t$  is time mean interval.

Prediction of the mean concentration in the troposphere with the use of (18), involves the prediction of the area mean surface concentration  $\langle \bar{C}(L,0,t) \rangle$ . Using the observed monthly mean surface radioactive concentration over the United States for the years 1963 and 1964, the monthly area mean surface concentration has been computed,<sup>2</sup> and plotted as dots in Fig. 3. In this computation,  $L=3000$  km is used. Fig. 3 indicates that the monthly area mean surface concentration generally decreases with time and that its relative maxima and minima occurred, respectively, in spring and late autumn for both 1963 and 1964. This agrees well with the findings of many scientists (Frey *et al.*, 1960; Machta *et al.*, 1961; Bierly,<sup>3</sup> 1965).

<sup>2</sup> Kao, S.-K., and W. J. Alder, *loc. cit.*

<sup>3</sup> The application of isotopes to some problems in atmospheric sciences. Paper presented at the Symposium on Isotope Techniques in the Hydrologic Cycle.

To construct a model for the prediction of the variation of radioactive concentration in the troposphere, we assume that the tropopause-break is the opening for the injection of radioactive debris from the stratosphere into the troposphere. Let the radioactive concentration in the tropopause-break at a given time be

$$C_J(t) = \bar{C}_J(t) + C'_J(t),$$

where the bar denotes the time mean and the prime the departure from the mean.

Likewise, let the wind velocity at and relative to the tropopause-break be

$$V_{Jr}(t) = \bar{V}_{Jr}(t) + V'_{Jr}(t).$$

The instantaneous source strength at, or the flux of radioactive debris through, the tropopause-break may be expressed as

$$Q_J(t) = C_J(t)V_{Jr}(t) = \bar{C}_J\bar{V}_{Jr} + \bar{C}_J V'_{Jr} + C'_J\bar{V}_{Jr} + C'_J V'_{Jr},$$

and the time mean of the source strength is, therefore,

$$\bar{Q}_J(t) = \bar{C}_J\bar{V}_{Jr} + \overline{C'_J V'_{Jr}}. \tag{19}$$

This equation shows that the mean flux of radioactive debris from the stratosphere into the troposphere through the tropopause-break is due to the transport by the mean motion relative to the jet core  $\bar{C}_J\bar{V}_{Jr}$  and transport by the turbulent motion  $\overline{C'_J V'_{Jr}}$ .

Applying the mixing-length hypothesis to the last term of the above equation, we have

$$\bar{Q}_J(t) = \bar{C}_J\bar{V}_{Jr} + \overline{V_{Jr}' \ell_n'} \frac{\partial \bar{C}_J}{\partial n}, \tag{20}$$

where  $\ell_n'$  is the mixing-length along  $n$ , perpendicular to the jet core. Since the space variation of the mean concentration from the source to its neighboring region in the troposphere is generally large and is difficult to determine, we assume that  $\partial \bar{C}_J / \partial n$  is proportional to the mean concentration in the jet core. Thus,

$$\bar{Q}_J(t) = \bar{C}_J\bar{V}_{Jr} + a\sigma_s\bar{C}_J, \tag{21}$$

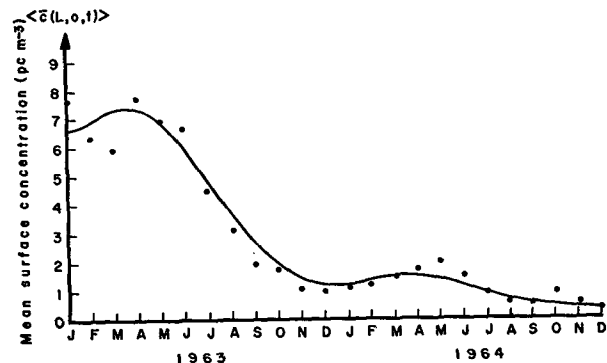


FIG. 3. Monthly variation of mean surface concentration for 1963 and 1964.

where  $a$  is a constant and  $\sigma_v$  is the standard deviation of the velocity in the jet core.

The position of the mean jet core in the atmosphere has a yearly period, reaching its lowest latitude in winter and its highest latitude in summer. Since the longitudinal mean meridional wind velocity in the jet core is generally small, the mean meridional wind velocity relative to the jet core may be approximated by the mean meridional velocity of the jet core. We assume that

$$\bar{V}_{Jr}(t) = A \sin \frac{2\pi}{12} t, \quad (22)$$

where  $A$  is the amplitude of the mean meridional velocity of the jet core, and  $t$  is in units of months.

Observation shows that a meridional gradient of radioactive concentration exists in the stratosphere near the tropopause (Machta *et al.*, 1961). In spring the jet core moves at its highest speed poleward into the region of high radioactive concentration. The flux of radioactive debris into the troposphere is, therefore, highest in spring, as is the radioactive concentration in the troposphere. In autumn the jet core moves at its highest speed equatorward into the region of small concentration. The concentration in the troposphere is, therefore, lowest in autumn. Furthermore, radioactive concentration generally decreases with time because of radioactive decay and fallout. We assume that the mean concentration in the jet core be expressed as

$$\bar{C}_J(t) = \left( \bar{C}_m - B \cos \frac{2\pi}{12} t \right) \exp(-bt), \quad (23)$$

where  $(\bar{C}_m - B)$  is the initial mean radioactive concentration in the jet core when the jet core is at its lowest latitude and  $b = \lambda + \beta$  is the residence constant where  $\lambda$  and  $\beta$  are the decay and fallout constants, respectively.

The mean source strength in the jet core therefore, becomes,

$$\bar{Q}_J(t) = \left( a\sigma_v + A \sin \frac{\pi}{6} t \right) \left( \bar{C}_m - B \cos \frac{\pi}{6} t \right) \exp(-bt). \quad (24)$$

Since the area mean surface concentration is proportional to the flux of radioactive debris from the stratosphere into the troposphere, the former may be written as

$$\begin{aligned} \langle \bar{C}(L, 0, t) \rangle &= m \bar{Q}_J(t) \\ &= m \left( a\sigma_v \bar{C}_m + A \bar{C}_m \sin \frac{\pi}{6} t - aB\sigma_v \cos \frac{\pi}{6} t \right. \\ &\quad \left. - 0.5AB \sin \frac{\pi}{3} t \right) \exp(-bt), \quad (25) \end{aligned}$$

where  $m$  is a constant of proportionality.

The solid curve in Fig. 3 represents the theoretical distribution of the monthly area mean surface concentration (25) for  $b = 0.135 \text{ month}^{-1}$ , and  $a\sigma_v \bar{C}_m = 10.20$ ,  $mA \bar{C}_m = 0.85$ ,  $aB\sigma_v = 3.40$  and  $mAB = 0.284$ , all in picocuries (pc)  $\text{m}^{-3}$ . The theoretical curve in Fig. 3 for the monthly area mean surface concentration agrees generally well with the observation.

We find from the above values that  $a\sigma_v \doteq 12A$  and  $B \doteq \frac{1}{3} \bar{C}_m$ . Thus, the source strength at the jet core for 1963 and 1964 may be expressed as

$$\begin{aligned} \bar{Q}_J(t) &= A \bar{C}_m \left( 12 + \sin \frac{\pi}{6} t \right) \left( 1 - \frac{1}{3} \cos \frac{\pi}{6} t \right) \\ &\quad \times \exp(-0.135t), \quad (26) \end{aligned}$$

where the amplitude of the mean meridional velocity of the jet core  $A$  and the mean concentration at the jet core at the initial time  $\bar{C}_m$  can be determined from observations. The source strength at the jet core can, therefore, be predicted.

The equation for the mean concentration in the troposphere may be expressed as

$$\begin{aligned} \bar{C}(y, z, t) &= m \left( a\sigma_v + A \sin \frac{\pi}{6} t \right) \left( \bar{C}_m - B \cos \frac{\pi}{6} t \right) \exp(-bt) \\ &\quad \times \left\{ G + M \ln \left[ \frac{K_y}{K_z} \left( \frac{z}{H} - 1 \right)^2 + \left( \frac{y}{H} \right)^2 \right] \right\}, \quad (27) \end{aligned}$$

and the corresponding equation for monthly mean radioactive concentration in the troposphere during 1963 and 1964 as

$$\begin{aligned} \bar{C}(y, z, t) &= \left( 10.2 + 0.85 \sin \frac{\pi}{6} t - 3.4 \cos \frac{\pi}{6} t \right. \\ &\quad \left. - 0.142 \sin \frac{\pi}{3} t \right) \exp(-0.135t) \left\{ 1.746 - 0.088 \right. \\ &\quad \left. \times \ln \left[ 800 \left( \frac{z}{H} - 1 \right)^2 + \left( \frac{y}{H} \right)^2 \right] \right\}, \quad (28) \end{aligned}$$

where  $\bar{C}(y, z, t)$  is in units of  $\text{pc m}^{-3}$  and can easily be computed with the use of Fig. 3 and the table in the Appendix.

It is seen from the magnitude of the terms in (26) and (28) that turbulent motion near the jet core plays the primary role in the transport of radioactive debris from the stratosphere into the troposphere. The spring maximum and autumn minimum are primarily the contribution of the mean motion of the jet core and the meridional gradient of the concentration near the tropopause in the stratosphere. A semiannual period in the variation of the radioactive concentration exists, resulting from the interaction between the meridional

concentration gradient and the mean motion of the jet core.

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APPENDIX

Values of  $\bar{C}_L(y,z,T)$  for various  $y/H$  and  $z/H$ .

$z/H$	$y/H$										
	0	50	100	150	200	250	300	350	400	450	500
1.0		1.057	0.935	0.864	0.813	0.774	0.742	0.715	0.692	0.671	0.652
0.9	1.563	1.057	0.935	0.864	0.813	0.774	0.742	0.715	0.691	0.671	0.652
0.8	1.441	1.056	0.935	0.864	0.813	0.774	0.742	0.715	0.691	0.671	0.652
0.7	1.370	1.055	0.935	0.864	0.813	0.774	0.742	0.715	0.691	0.671	0.652
0.6	1.319	1.053	0.934	0.864	0.813	0.774	0.742	0.715	0.691	0.671	0.652
0.5	1.280	1.051	0.934	0.863	0.813	0.774	0.742	0.715	0.691	0.671	0.652
0.4	1.248	1.048	0.933	0.863	0.813	0.774	0.742	0.715	0.691	0.671	0.652
0.3	1.221	1.045	0.932	0.863	0.813	0.774	0.742	0.715	0.691	0.671	0.652
0.2	1.197	1.041	0.931	0.862	0.812	0.774	0.742	0.715	0.691	0.671	0.652
0.1	1.176	1.037	0.930	0.862	0.812	0.773	0.742	0.715	0.691	0.670	0.652
0.0	1.158	1.033	0.929	0.861	0.812	0.773	0.741	0.714	0.691	0.670	0.652

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