ON EDDY DIFFUSION COEFFICIENTS

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Abstract

This paper presents a review concerning the transport by eddies in the stratosphere and its parameterization in atmospheric models. The eddy diffusion concept is very convenient for aeronomical calculations since its leads to satisfactory distributions of minor constituents but it is not theoretically demonstrated. Therefore the eddy diffusion coefficients are usually deduced from the distribution of several trace species and have to be considered as phenomenological parameters.

1. INTRODUCTION: The behavior of minor constituents in the atmosphere is determined by a combination of chemical and photochemical reactions and transport processes. The relative importance of these two effects varies considerably from one species to another and for each of them is a function of the altitude, latitude and time. When the residence time characterising a region of the atmosphere becomes of the same order of magnitude, or smaller, than the chemical half time of a constituent its transport has to be taken into account.

Gaseous and particulate trace species suspended in the atmosphere are transported quasi- horizontally by motion systems of widely varying space and time scales. In fact, the transport of atmospheric trace substances can be represented by mean motions associated with the zonal and meridional circulation and by a broad spectrum of

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wave motions. These include in particular the tropospheric systems of wavenumbers about 3 to 9 which die out in the lower stratosphere and the large wavenumbers 1-2 which may increase in amplitude with height in winter in the middle stratosphere.

In most two-dimensional stratospheric models, the transport of minor constituents will be parametrized by a combination of mean and turbulent motions. If one considers a small volume of particles suspended in the atmosphere, mean motions will displace the center of mass of the volume without deforming it and without modifying the particle concentrations; turbulent motions will distort the volume and the particles will be spread out. Therefore, from a macroscopic point of view, the eddy motions act very much as diffusion processes.

The purpose of this paper is to survey how the fluctuating component of the atmospheric dynamics can be mathematically modeled in the homosphere (below 100 km). The problem of assessing mean motions in relation to the thermal structure of the atmosphere is treated in other lectures of this Advanced Study Institute (see Murgatroyd, 1979; Pyle, 1979). It should be noted, however, that the distinction between mean motion and eddy diffusion is not unique and will, thus, depend upon the model. Therefore, in most cases, when both types of data are not consistent, the methods used to derive exchange coefficients (also called eddy diffusion coefficients) will lead to approximate values which will have to be tested and adjusted by making numerical experiments. Also, when deriving a transport model, a distinction should be made between two-dimensional models, where meridional exchanges are considered, and one-dimensional representations where horizontal stratification is assumed and only vertical transport is considered. In both cases, however, the definition of eddy diffusion coefficients for the transport of heat or minor constituents, such as ozone and water vapor, cannot be fully justified by fluid dynamics theory. However, since it leads to results (heat or particle concentration, fluxes,...) in rather good agreement with observation and since the formalism of such complicated mechanisms is rather simple, these coefficients are readily used by aeronomers while their use is widely criticised by meteorologists.

2. MEAN MOTIONS AND FLUCTUATIONS: Since many sporadic phenomena appear in the atmosphere, one assumes that the general circulation can be described by the average value of atmospheric quantities and by correlations between the fluctuations of these quantities about their average. Therefore, one introduces the temporal local mean

$$\chi(T) = \frac{1}{T}\int_0^T \chi(t)\, dt \qquad (1)$$

of any atmospheric quantity $\chi(t)$ (e.g. the concentration, the temperature or the wind velocity), so that

$$\chi(t) = \overline{\chi} + \chi'(t) \qquad (2)$$

where $\chi'(t)$ represents the departure of χ from $\overline{\chi}$. The time interval T is generally chosen so that the mean motion can be considered as stationary. Zonal means $[\chi]$ i.e. averages round latitude circles can also be introduced and are of particular interest in two-dimensional models. If λ represents the longitude, one writes

$$[\chi] = \frac{1}{2\pi}\int_0^{2\pi} \chi(\lambda)\, d\lambda \qquad (3)$$

and any atmospheric variable can be expressed as

$$\chi(\lambda) = [\chi] + \chi^*(\lambda) \qquad (4)$$

where $\chi^*(\lambda)$ is the departure of χ from its zonal average.
Further mean quantities can be defined, for example averages over all longitudes and latitudes which are useful in one-dimensional (vertical) models. Finally, one can also introduce an average both in time and longitude called $[\overline{\chi}]$ and write for any quantity varying with longitude and time

$$\chi(\lambda,t) = [\overline{\chi}] + [\chi'] + \overline{\chi}^* + \chi'^* \quad (5)$$

Here the first term $[\overline{\chi}]$ refers to the zonal-time mean, the second $[\chi']$ is the time fluctuation averaged over latitudinal circles, the third $\overline{\chi}^*$ is the departure from the zonal mean averaged over a period of time and the last term $\overline{\chi}'^*$ is the residual. If one now considers the product of two fluctuating quantities (e.g. the concentration and the meridional wind component), the mean value of this product can be written following the example of Newell (1966)

$$\overline{nv} = \overline{n}.\overline{v} + \overline{n'v'} \quad (6a)$$

$$[nv] = [n] . [v] + [n^*v^*] \quad (6b)$$

$$[\overline{nv}] = [\overline{n}] . [\overline{v}] + [\overline{n^*}.\overline{v^*}] + [\overline{n'v'}] \quad (6c)$$

The last expression shows that the mean south to north over the time T transport of a quantity (here the concentration) in the meridional plane can be represented by the sum of :

(i) a mean motion component $[\overline{n}].[\overline{v}]$

(ii) a standing eddies component (expressed as the correlation between $\overline{n^*}$ and $\overline{v^*}$ around the latitude circles)

(iii) A transient eddy component (expressed as the zonal average of the time correlation of n' and v').

Atmospheric motions of all scales contribute with different weights to the correlations between the fluctuations. The presence of these scale effects leads to serious difficulties in the treatment and interpretation of the equations of atmospheric dynamics.

3. CONTINUITY EQUATION AND TURBULENT TRANSPORT OF TRACE SPECIES: The instantaneous concentration n(t) of a trace constituent in the atmosphere can be derived, in the homosphere, from the continuity equation

$$\frac{\partial n}{\partial t} + \vec{\nabla} . (n.\vec{v}) = P - L \quad (7)$$

where P and L are, respectively, the local production and destruction rate of the species (e.g. chemical or photochemical reactions) and $\vec{v}$ the instantaneous wind velocity vector. If one wishes to derive the mean local concentration $\overline{n}$, one has to solve the following equation

$$\frac{\partial \overline{n}}{\partial t} + \vec{\nabla} . (\overline{n}\,\overline{\vec{v}} + \overline{n'\vec{v}'}) = \overline{P} - \overline{L} \quad (8)$$

while, if the zonal and time average concentration [n] is required, the continuity equation

$$\frac{\partial [\overline{n}]}{\partial t} + \vec{\nabla}.([\overline{n}][\overline{\vec{v}}] + [\overline{n^*}\overline{\vec{v}^*}] + [\overline{n'\vec{v}'}]) = [\overline{P}] - [\overline{L}] \quad (9)$$

It should be noted that the determination of the mean value of P and L generally requires the calculation of time/space correlation products between the concentration of different species (and also reaction rates which may vary with temperature) and, therefore, depends on the turbulent state of the atmosphere. However, in most models this effect is usually neglected and will not be considered here.

Even if the mean circulation $[\overline{\vec{v}}]$ is known, or is derived from other dynamical equations, equation (9) still needs a supplementary condition before it can be solved, namely an equation relating the turbulent and the mean motions terms. The K-theory provides the simplest turbulence closure approximation available for this purpose. It assumes that the eddy fluxes are proportional to the negative gradient of the mixing ratio $f = n/n(M)$, where $n(M)$ is the total atmospheric concentration. If one defines the time and zonal mean of the meridional (y) and vertical (z) turbulent flux components by

$$[\overline{\Phi_y}] = [\overline{n^* v^*}] + [\overline{n'v'}] \quad (10a)$$

$$[\overline{\Phi_z}] = [\overline{n^* w^*}] + [\overline{n'w'}] \quad (10b)$$

where v and w refer respectively to the meridional and vertical components of the wind velocity $\vec{v}$, the simplest assumption leads to the Fickian law

$$[\overline{\Phi_y}] = - K_y \, n(M) \frac{\partial f}{\partial y} \quad (11a)$$

$$[\overline{\Phi_z}] = - K_z \, n(M) \frac{\partial f}{\partial z} \quad (11b)$$

where K_y and K_z are (positive) exchange coefficients.
These expressions have been used by Machta and List (1959), Prabhakara (1963) and Jessen (1973) but it has been recognized, after an analysis of heat fluxes (White, 1954; Murakami, 1962 and Peng, 1963) and ozone transport (Newell, 1961; Hering and Borden, 1964), that horizontal eddy fluxes could clearly be countergradient above the tropopause. In his study on heat transport in the lower stratosphere, White (1954) points out that "up to the 200 mb level (12 km), the eddy flux of sensible heat is poleward from regions of high to regions of low temperature as might normally expected. At and above this level, the reverse is true". White notes that "above the tropopause level, the eddy processes are acting to build up rather than dissipate the existing temperature gradient". Newell (1964) has given a physical explanation for such an horizontal countergradient flux. He considers (figure 1) an air parcel A in the lower stratosphere moving poleward and downward at a slope exceeding that of the potential temperature. Such trajectories are common as shown by dispersion studies of radioactive tracers. Arriving in A', the air parcel will be warmer than its environment. Consequently it will be buoyant and tend to go back up unless forces are available to keep this from happening. Newell suggests that the kinetic energy of the motions themselves can do this, provided that the energy is replaced by upward transport from the lower portions of the westerly wind core. Figure 2 illustrate the slope of the maximum concentration level associated with various tracers injected into the stratosphere and shows that the inclination is steeper than the slopes of the isentropic surfaces. It can be seen that the motion AA' is up the horizontal gradient although it is down the vertical gradient.

4. THE CLASSICAL THEORY OF LARGE SCALE MERIDIONAL EDDY DIFFUSION: Demazure and Saïssac (1962) and Reed and German (1965) have developed a concept in 2 dimensions for eddy diffusion of conservative trace constituents taking into account possible countergradient transport in the meridional plane. The authors approach is based on the mixing length concept of the turbulence theory. For reasons of simplicity, transient and standing eddies are not distinguished and the flux is given by the following expressions

$$\phi_y = \overline{n'v'} \quad (12a)$$

$$\phi_z = \overline{n'w'} \quad (12b)$$

In this theory, it is assumed (figure 3) that an air parcel located at P_1 and representative of its local environment, moves a distance $\vec{l}(l_y, l_z)$, called the displacement vector or the mixing length, before it mixes suddenly and completely with its new environmental air at P_o. It is also assumed that during the displacement the mixing ratio f in the air parcel is conserved. If the vector $\vec{l}$ is allowed to have any orientation in space, the deviation of the conservative quantity f is given, to a first order approximation, by

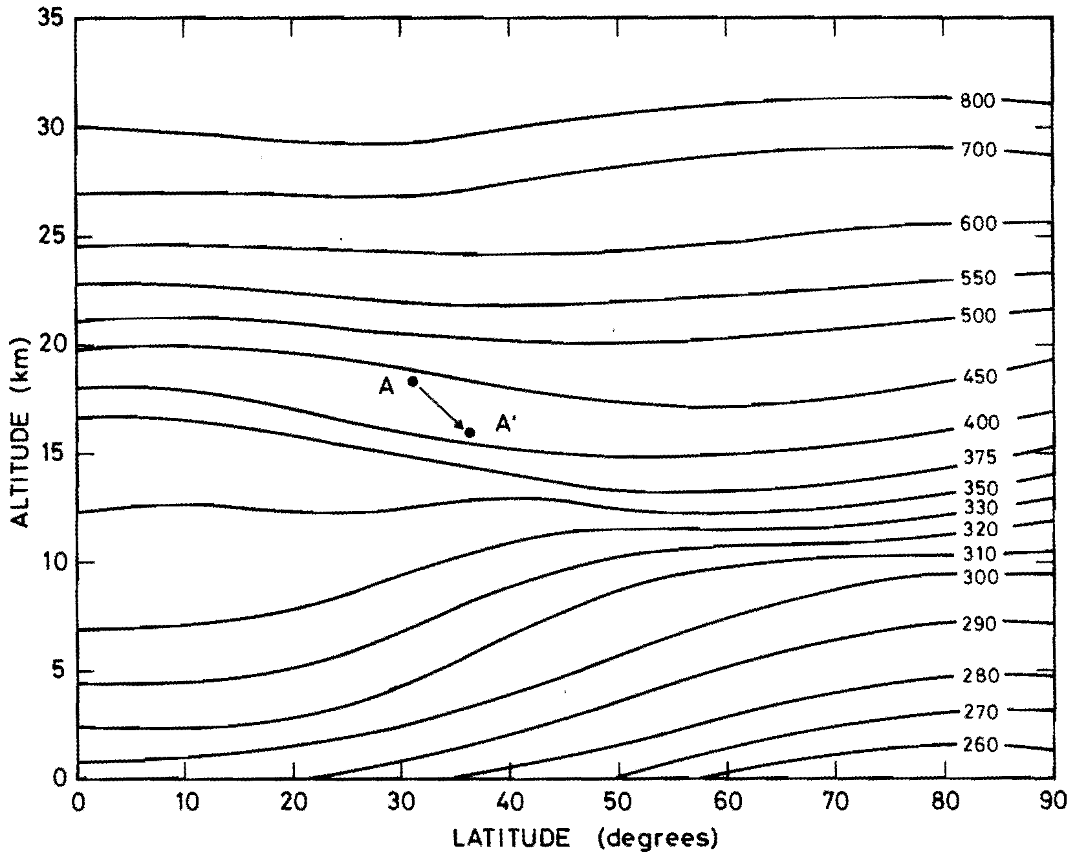

Fig. 1.- Potential temperature surfaces shown for one hemisphere in late winter. Temperature is given in degrees. Kelvin. In the lower stratosphere, poleward-moving parcels (A,A') descend more steeply than the potential temperature surfaces do. After Newell (1964).

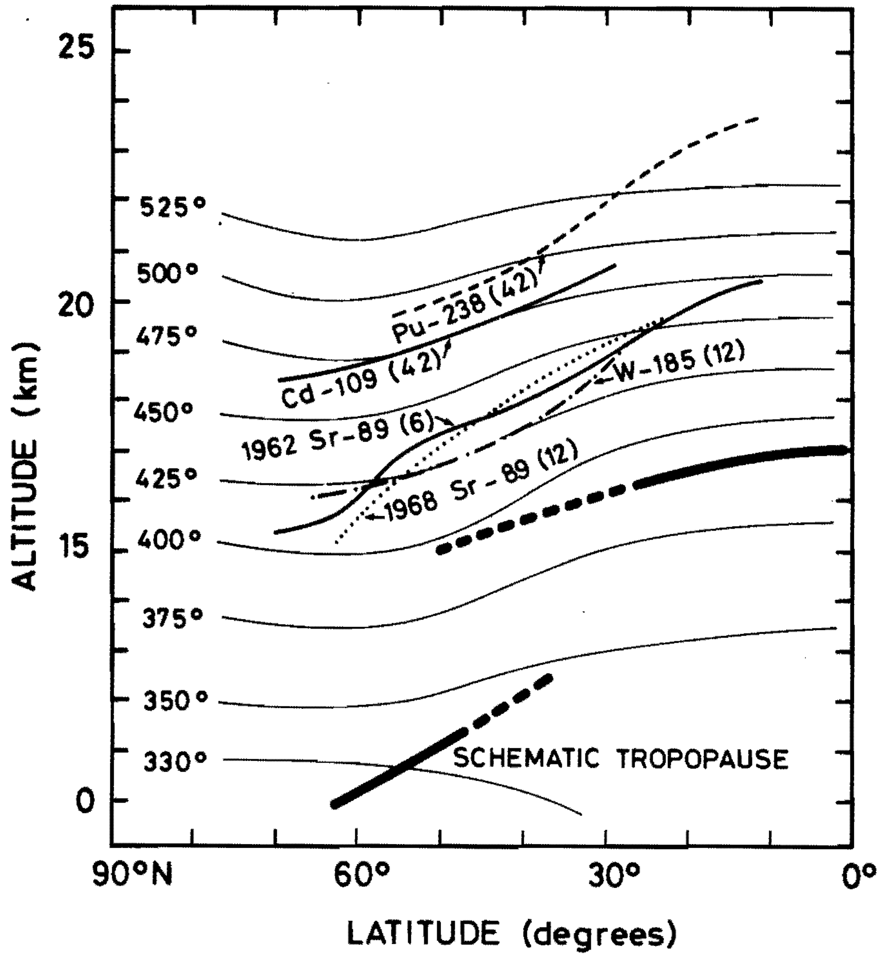

Fig. 2.- Altitude of the maximum concentration level versus latitude associated with various tracers injected into the stratosphere by the explosion of thermonuclear weapons in the early 60's. Potential temperature surfaces are also shown.

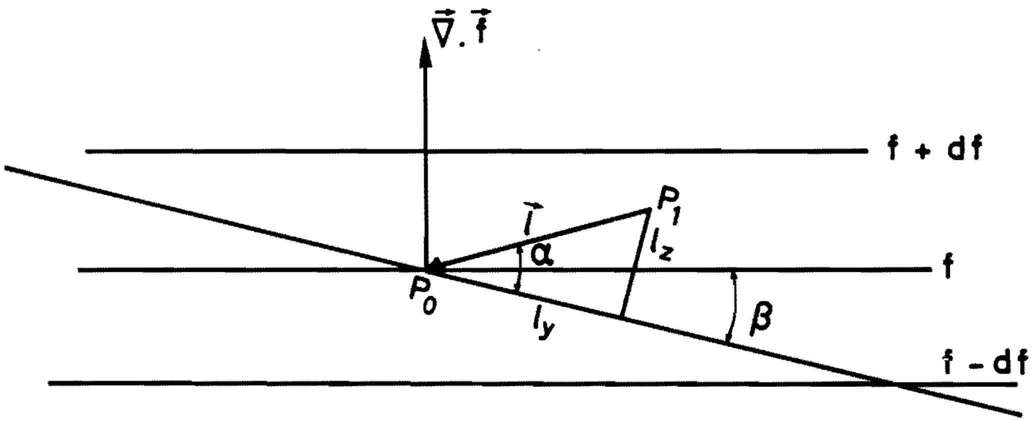

Fig. 3.- Model for the eddy flux of a property by exchange along a sloping mixing path. After Reed and German (1965).

$$f' = f_{P_1} - f_{P_o} = - \vec{l}.\vec{\nabla}f = - (l_y \frac{\partial f}{\partial y} + l_z \frac{\partial f}{\partial z}) \quad . \qquad (13)$$

Considering all the various parcel displacements to P_o during the time T substitution of (13) into (12a and b) leads to the time average flux components (f represents the mean mixing ratio)

$$\phi_y = - n(M) \; [K_{yy} \frac{\partial f}{\partial y} + K_{yz} \frac{\partial f}{\partial z}] \qquad (14a)$$

$$\phi_z = - n(M) \; [K_{zy} \frac{\partial f}{\partial y} + K_{zz} \frac{\partial f}{\partial z}] \qquad (14b)$$

where the K_{ij} coefficients are correlation products between the displacement and the velocity components :

$$K_{yy} = \overline{l_y v'} \qquad (15a)$$

$$K_{yz} = \overline{l_z v'} \qquad (15b)$$

$$K_{zy} = \overline{l_y w'} \qquad (15c)$$

$$K_{zz} = \overline{l_z w'} \qquad (15d)$$

Equations (14a and b) are reduced to the classical Fickian law (11a and b) only when the covariences between l_z and v' and l_y and w' are equal to zero. However, this is not the case since, as shown before, sinking motions in the stratosphere on the average coincide with polewards transport while rising motions are most frequently equatorwards. This was already established by Molla and Loisel in 1962. Accordingly, the introduction of K_{yz} and K_{zy} allows for the countergradient fluxes in the atmosphere.

Assuming that the mixing length ℓ (∿ 100 km) is small compared to the eddy sizes involved in the large scale mixing processes (∿ 1000 km), Reed and German have made the hypothesis that the velocity $\vec{V}$ and the displacement vector $\vec{\ell}$ are in the same direction. If α is the angle between $\vec{\ell}$ and the horizontal axis, one can write, since for large scale motions this angle is very small (< 1/1000),

$$v' = V \cos \alpha \cong V \qquad (16a)$$

$$l_y = \ell \cos \alpha \cong \ell \qquad (16b)$$

$$w' = V \sin \alpha \cong V\alpha \qquad (16c)$$

$$l_z = \ell \sin \alpha \cong \ell\alpha \qquad (16d)$$

Therefore, if α is divided into its mean value $\overline{\alpha}$ and its departure α' and if $\overline{\alpha}$ and $\overline{\alpha'^2}$ are assumed to be independent of V and ℓ, one obtains the relations

$$K_{yz} = K_{zy} = \overline{\alpha} \; K_{yy} \qquad (17)$$

$$K_{zz} = (\overline{\alpha}^2 + \overline{\alpha'^2}) \; K_{yy} \qquad (18)$$

Expression (17) shows that the diffusion matrix K_{ij} can be considered as symmetrical since the off diagonal terms K_{yz} and K_{zy} have the same value in this theory. Also, it appears that K_{yy} and K_{zz} necessarily have the same sign (positive) while the sign of K_{yz} is determined by that of the angle α.

Introducing now the slope of the mixing ratio surface

$$\bar{\beta} \cong \tan \bar{\beta} = - \frac{\partial f/\partial y}{\partial f/\partial z} \qquad (19)$$

the following expressions are obtained

$$\phi_y = - n(M)\, K_{yy} \left(1 - \frac{\bar{\alpha}}{\bar{\beta}}\right) \frac{\partial f}{\partial y} \qquad (20a)$$

$$\phi_z = - n(M)\, K_{zz} \left(1 - \frac{\bar{\alpha}\bar{\beta}}{\bar{\alpha}^2 + \overline{\alpha'^2}}\right) \frac{\partial f}{\partial z} \qquad (20b)$$

These equations show that the meridional flux of trace species becomes countergradient if

$$\bar{\alpha} \geqslant \bar{\beta} \qquad (21)$$

that is when the slope of the prefered mixing surface becomes larger than the slope of the mixing ratio surface. This condition applies in the lower stratosphere but not in the extratropical troposphere where, according to Eady (1949), $\alpha \cong \beta/2$.

The same type of argument can be presented for heat transport. In this case, the heat flux components are written in the form

$$F_y = - n(M) \left[K_{yy} \frac{\partial \bar{\theta}}{\partial y} + K_{yz} \frac{\partial \bar{\theta}}{\partial z}\right] \qquad (22a)$$

$$F_z = - n(M) \left[K_{zy} \frac{\partial \bar{\theta}}{\partial y} + K_{zz} \frac{\partial \bar{\theta}}{\partial z}\right] \qquad (22b)$$

with $K_{yz} = K_{zy}$. Countergradient transport appears when the slope $\bar{\alpha}$ becomes larger than that of the isentropic surfaces.

Adopting expressions (14a and b) and (9), the continuity/ transport equation becomes

$$n(M) \frac{\partial f}{\partial t} - \frac{\partial}{\partial y}\left(K^*_{yy} \frac{\partial f}{\partial y}\right) - \frac{\partial}{\partial y}\left(K^*_{yz} \frac{\partial f}{\partial z}\right) - \frac{\partial}{\partial z}\left(K^*_{zy} \frac{\partial f}{\partial y}\right)$$

$$- \frac{\partial}{\partial z}\left(K^*_{zz} \frac{\partial f}{\partial z}\right) + \left(v^* + \frac{K^*_{yy}\ \mathrm{tg}\ \varphi}{a}\right) \frac{\partial f}{\partial y} + \left(w^* + \frac{K^*_{yz}\ \mathrm{tg}\ \varphi}{a}\right) \frac{\partial f}{\partial z}$$

$$= P - L \qquad (23)$$

where $K^*_{ij} = n(M).K_{ij}$, $\bar{v}^* = n(M).\bar{v}$, $\bar{w}^* = n(M).\bar{w}$, and $\bar{v}$ and $\bar{w}$ are the mean wind components. The numerical solution of this equation will provide the distribution of the mixing ratio (or concentration) of the trace species under consideration if all the parameters are known and if suitable boundary conditions are specified. In particular, the values of the exchange coefficients have to be established in the whole physical domain. The ellipticity condition associated with equation (23) implies that

$$K_{yz}^2 \leqslant K_{yy} K_{zz} \qquad (24)$$

which is always verified as shown when expressions (17) and (18) are introduced in (24).

Since the diffusion tensor (or matrix) is symmetrical, it is possible to rotate (by an angle γ) the (y,z) axis such that the new axes (Y,Z) become principal axes in which the off-diagonal elements $K_{yz} = K_{zy}$ are eliminated. Reed and German show that the matrix in the principal axis system is given by

$$\begin{bmatrix} K_Y & 0 \\ 0 & K_Z \end{bmatrix} = \begin{bmatrix} K_{yy} \cos^2\gamma + K_{yz} \sin 2\gamma + K_{zz} \sin^2\gamma & \frac{K_{zz} - K_{yy}}{2} \sin 2\gamma + K_{yz} \cos 2\gamma \\ \frac{K_{zz} - K_{yy}}{2} \sin 2\gamma + K_{yz} \cos 2\gamma & K_{yy} \sin^2\gamma - K_{yz} \sin 2\gamma + K_{zz} \cos^2 \gamma \end{bmatrix}$$

The angle γ corresponding to a principal axis system is thus given by

$$\frac{K_{zz} - K_{yy}}{2} \sin 2\gamma + K_{yz} \cos 2\gamma = 0 \qquad (25)$$

or, since $\overline{\alpha}$ is small,

$$\gamma = \overline{\alpha} \qquad (26)$$

In other words, the inclination of the principal axis and the slope of the preferred mixing surface are identical.

Since the values of K_{ij} depend on the adopted axes and their inclination upon the direction of preferred mixing, it is sometimes convenient to use the following expressions which relate K_{ij} and the principal eddy diffusion components :

$$K_{yy} = K_Y \cos^2 \alpha + K_Z \sin^2 \alpha, \qquad (27)$$

$$K_{yz} = K_{zy} = (K_Y - K_Z) \sin \alpha \cos \alpha, \qquad (28)$$

$$K_{zz} = K_Y \sin^2 \alpha + K_Z \cos^2 \alpha.$$

A geometrical representation is given by the diffusion ellipse (figure 4)

$$K_Y Y^2 + K_Z Z^2 = 1 \qquad (30)$$

whose principal axes have a lengths respectively, of $1/\sqrt{K_Y}$ and $1/\sqrt{K_Z}$. The magnitude of an eddy diffusion in a direction characterized by an angle γ can be derived from such a geometry (see fig. 4).

5. EVALUATION OF THE 2-D EXCHANGE COEFFICIENTS VALUES: The magnitude of the eddy diffusion coefficients vary with the scales of space-time averaging from a lower limit of molecular diffusion to an upper limit of global atmospheric mixing. This dependence of the K's versus space and time scales can be derived from a dispersion distance

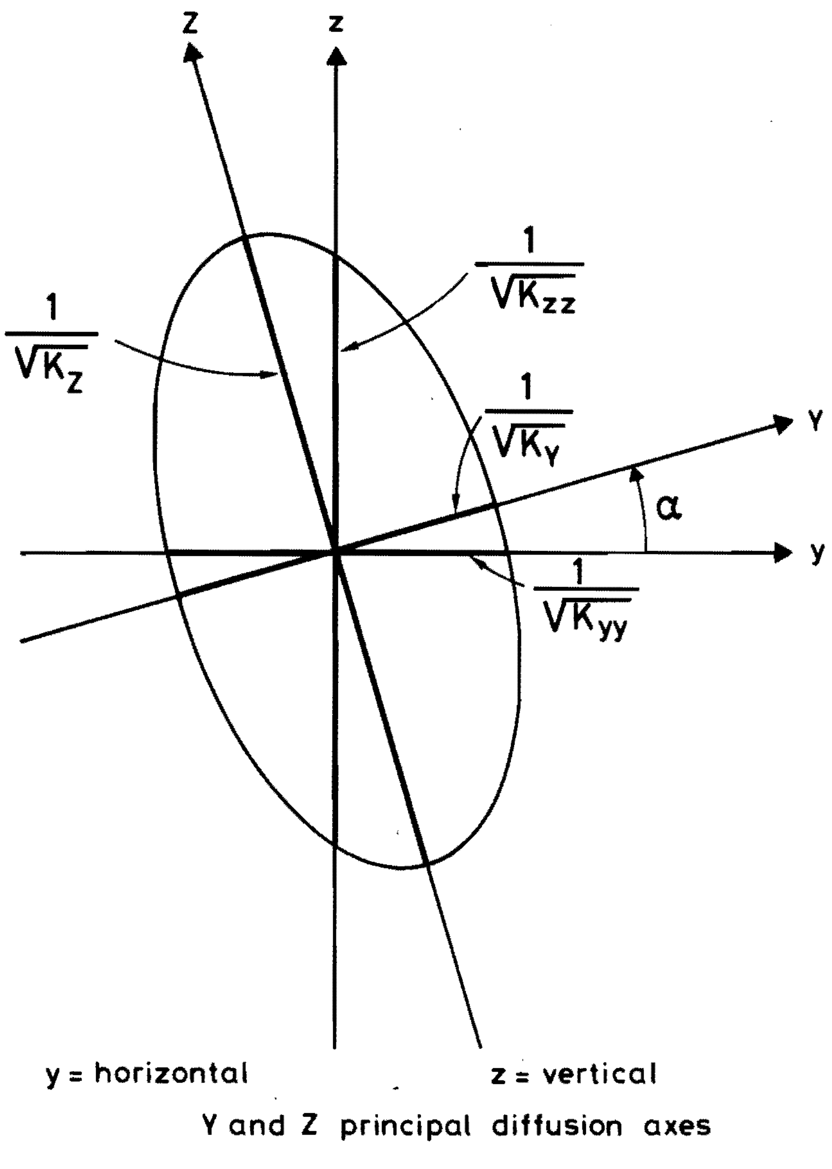

Fig. 4.- Diffusion ellipse.

(expressed by mean cloud width) as illustrated in figure 5. The lower limit on K_{yy} and K_{zz} (molecular diffusivity) decreases with height. The graph refers to a pressure of 100 mb. At these small scales, the turbulence is approximatively isotropic and homogeneous. The global scale is characterized by anisotropy and by the presence of off-diagonal terms. In the intermediate range, the turbulence is intermittent and localized. The curve refers to average values which can be several orders of magnitude smaller than the values observed locally. In the following paragraphs, we will confine our attention on large scale eddy diffusion only.

A procedure for evaluating the K coefficients has been given by Reed and German (1965). The authors have derived the K_{yy} component in the baroclinically active troposphere from the heat flux data (F_y) and the temperature distribution (Peixoto, 1960). In other atmospheric regions, they have computed K_{yy} by assuming that it is proportionnal to the variance of the meridional wind component as given by Buch (1954), Murakami (1962) and Peng (1963). The angle $\overline{\alpha}$ has then been obtained from expression (20.a) introducing the values of the heat flux and the temperature compiled by Oort (1963). K_{yz} has then been computed with equation (17). Since, for symmetry reasons, $\overline{\alpha} = 0$ at the equator, relation (18) provides $\overline{\alpha'^2} = K_{zz}/K_{yy}$ in these regions. Adopting $K_{zz} = 10^3$ cm^2 s^{-1} in the equatorial zone, as suggested by the study of the vertical spread of tungsten 185, $\overline{\alpha'^2}$ has been calculated and assumed to remain constant at all other latitudes. Equation (18) was then employed to estimate K_{zz} in the whole domain.

Davidson, Friend and Seitz (1966) have developed a numerical model of diffusion and rain out of stratospheric radioactive material using a fairly simple distribution of K's. K_{yy} varies smoothly from 10^8 cm^2 s^{-1} at the pole to 10^{10} cm^2 s^{-1} at the equator while K_{zz} is equal to 10^3 cm^2 s^{-1} in the stratosphere and about 4×10^4 cm^2 s^{-1} in the troposphere with a transition region near the tropopause.

Gudiksen, Fairhall and Reed (1968) have considered simultaneously, mean motions and large scale eddy diffusion to model the dispersion of tungsten 185 released in the atmosphere during nuclear weapons tests. They extended the work of Reed and German to derive seasonal values of the K's up to 27 km. The exchange coefficients obtained by Reed and German were reduced by a factor of 7-10 for K_{yy} and a factor of 2 for equatorial K_{zz}. The discrepency between the two sets of data was, mainly, attributed to the fact that the coefficients derived from heat flux data by Reed and German may not be quantitatively applicable to the transport of particulate debris. In fact, the potential temperature may not behave as conservatively as tungsten 185 in the lower stratosphere while the transport of the gaseous species may physically differ from the transport of solid particulates.

Seitz, Davidson, Friend and Feely (1968) also extended their previous work by introducting the complementary effects of mean and turbulent motions. These authors were able to simulate relatively well the evolution of several different tracers with the same transport coefficients, showing that large scale diffusion could be described with K's which are almost independent of the tracers.

Luther (1973) in a new investigation of the problem computed the values of K_{yy}, K_{yz} and K_{zz} between 0 and 50 km using the method of Reed and German but adopting the heat flux associated with standing and transient eddies and the temperature and the wind variance as compiled by Oort and Rassmusson (1971) for the 1958-1963 period. Values in regions where observational data were not available were derived by Luther (1973) by extrapolation using the results of Wofsy and McElroy (1973) and Newell et al. (1966).

Different attempts to establish more accurate distributions of the K's have been carried out in the past years especially because of the demand by chemical modelers studying the stability of ozone in the stratosphere. Values have been proposed by Louis (1974), Kao, Obrasinski and Lordi (1978) and others. Moreover, Nastrom and Brown (1978) have recently derived exchange coefficients from 30 to 60 km altitude where the meridional component K_{yy} has been obtained using G.I. Taylor's theorem

$$K_{yy} = \int_0^{\infty} \overline{v'(t)\ v'(t+\tau)}\, d\tau = \overline{v'^2} \int_0^{\infty} R_{vv}(\tau)\ d\tau \qquad (31)$$

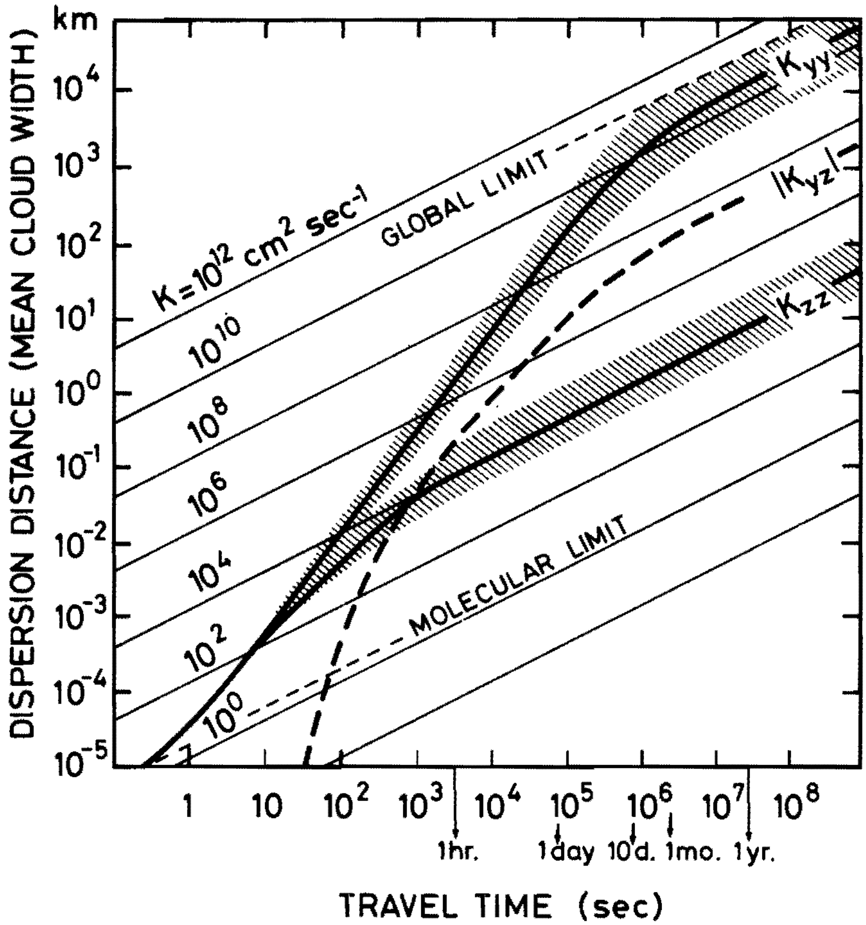

Fig. 5.- Stratospheric exchange coefficients as a function of dispersion distance and travel time. The range in values for a given travel time is given by the toned area. The dashed line represents the upper bound for $|K_{yz}|$. After Reiter et al. (1975).

where $v'(t)$ is the meridional wind fluctuation, $\overline{v'^2}$ its variance and

$$R_{vv}(\tau) = \frac{\overline{v'(t)\ v'(t+\tau)}}{\overline{v'^2}} \tag{32}$$

the autocorrelation coefficient of the meridional wind. This approach has been previously used by Murgatroyd (1969) who adopted for the autocorrelation coefficient a damped cosine function

$$R_{vv}(\tau) = e^{-p\tau} \cos q\ \tau \tag{33}$$

with p and q being obtained from wind trajectory data. The technique used by Nastrom and Brown to derive K_{yz} is based on that of Reed and German while the determination of the K_{zz} value follows a method suggested by Hines (1970). This author has assumed that the normal growth of gravity wave amplitude with hight arising from decreasing density will be offset by energy lost to turbulence so that the wave amplitude is constant with altitude. Zimmerman (1974) has argued that no amplitude growth is a pour approximation and balancing the vertical gradient of the specific wave energy with an effective turbulent viscosity he derived the following expression

$$K_{zz} = \left(\frac{\lambda_z}{4\pi^2 T} \right) \left\{ \frac{1}{H} - \frac{1}{z}\ \ell n \frac{V^2}{V_o^2} \right\} \tag{34}$$

where λ_z is the vertical wavelength of the upward propagating gravity wave responsible for turbulence, T is its period, V and V_o the perturbation velocity, respectively, at level z and at a reference level.

Figure 6a, b and c represents the exchange coefficients K_{yy}, K_{yz} and K_{zz} adopted by Reed and German (1965), Gudiksen et al. (1968) and Luther (1974) versus latitude at two different levels, namely 100 mb (14 km) and 50 mb (20 km), and for two seasons (winter and summer). The shape of the latitudinal variation is generally the same but the magnitude of the data sometimes varies considerably. All of the three authors agree on the fact that K_{yy} increases from the equator to the pole during the winter period while it varies only slightly and remains small during the summer. The off-diagonal term K_{yz} which is negative in the Northern hemisphere (in standard spherical coordinates) is also larger during the winter than during the summer. Its value is almost zero at the equator and at the poles (for symmetry reasons) and peaks in the mid-latitude regions. The vertical exchange coefficient K_{zz} seems also to reach its maximum value between 30 and 50 degrees latitude with the most pronounced values during the winter. Similar data have been adopted in two-dimensional models of stratospheric minor constituents (Brasseur, 1978; Crutzen, 1975; Prinn, 1973; Pyle, 1978; Rao-Vupputuri, 1973; Widhopf, 1975; etc...) but they have been adjusted by a "trial and error" method to give the best agreement between observed and calculated distributions of trace species such as ozone or water vapor. Figure 7 shows and compares the values of K_{yy} at 20 km adopted by various authors. It should be noted, however, that these values have been adjusted for different distributions of the mean wind components (see e.g. Cunnold et al., 1974; Louis 1974).

The meridional distribution of eddy diffusion coefficients determined by Luther between the ground and the stratopause is illustrated in figures 8, 9 and 10 while the same coefficients provided by Nastrom and Brown between 30 and 60 km are reproduced in tables 1, 2 and 3. In both cases, K_{yy} appears to increase with latitude in the winter period and also with height above 30 km. The values derived during the winter are about a factor of ten larger than the data obtained during the summer. The chart representing K_{yz} shows that the sign of this coefficient changes from one hemisphere to the other and also when crossing the tropopause. The values are the highest in the winter mid-latitude region. Hence, the countergradient flux becomes greatest mostly during the winter season. The K_{zz} coefficient has a high value in the troposphere but the its magnitude

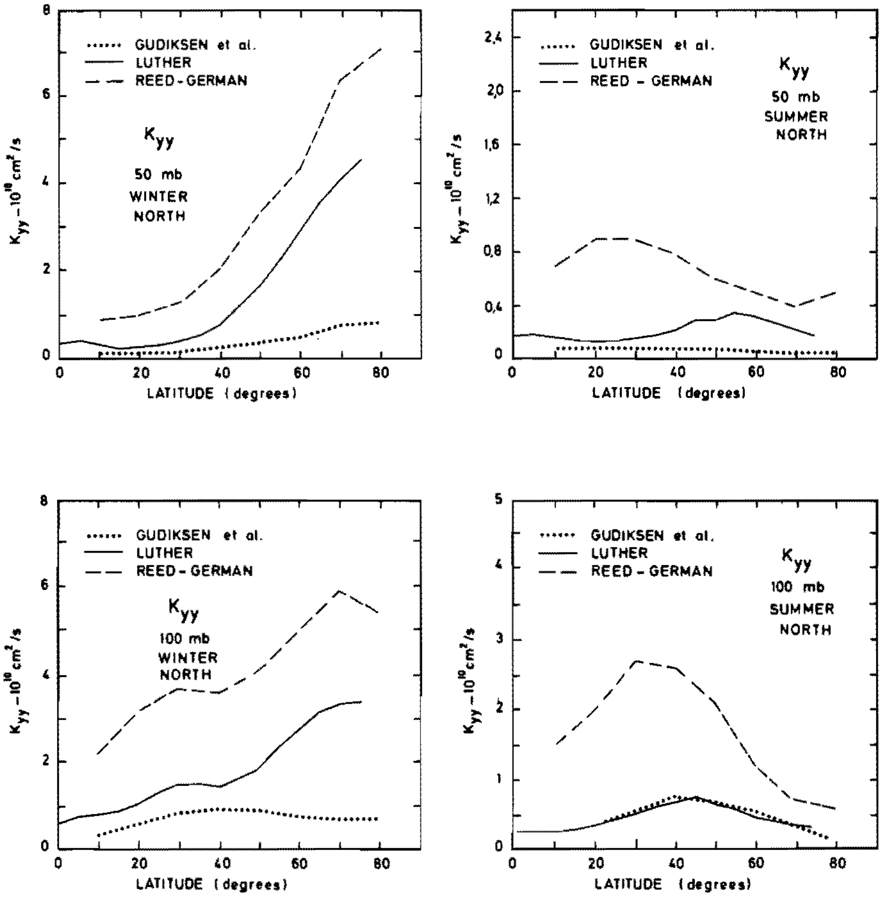

Fig. 6a.- Latitudinal distribution of the exchange coefficient K_{yy} according to different authors. The values are given for winter and summer conditions and for 50 and 100 mb levels.

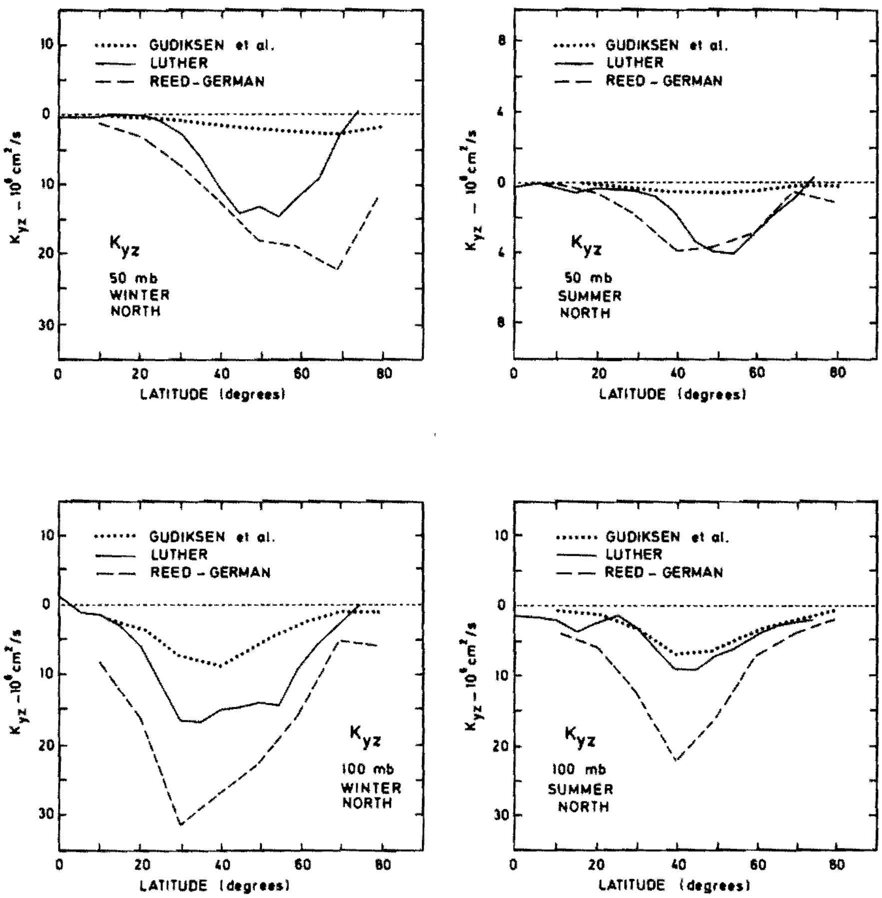

Fig. 6b.- Latitudinal distribution of the exchange coefficient K_{yz} according to different authors. The values are given for winter and summer conditions and for 50 and 100 mb levels.

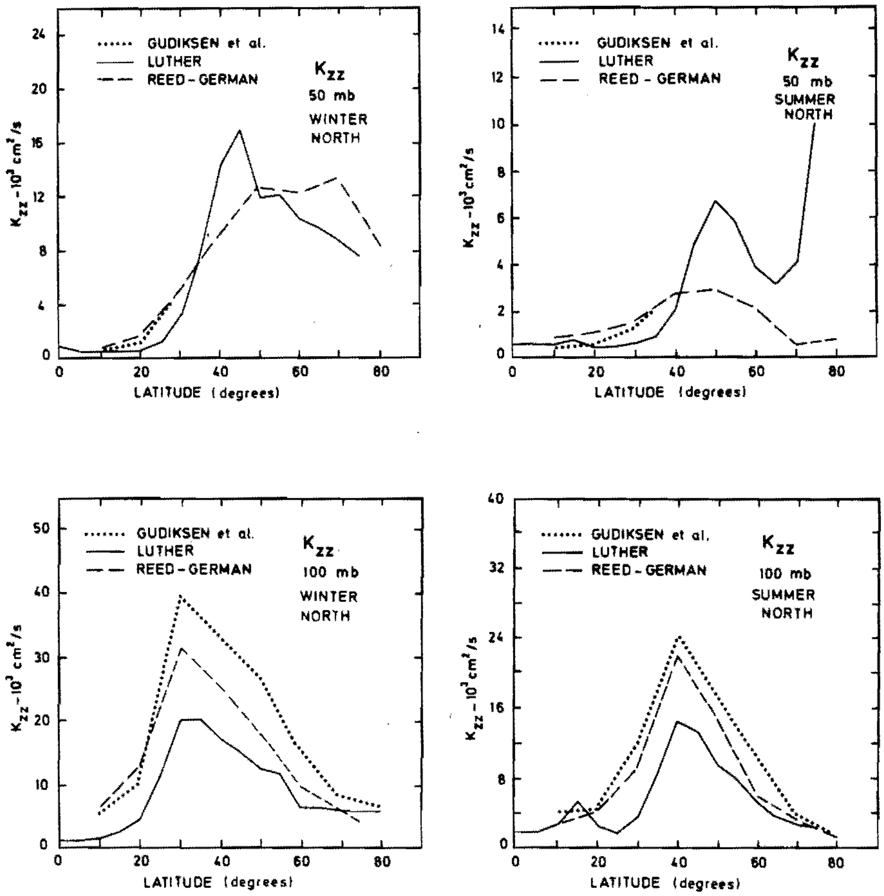

Fig. 6c.- Latitudinal distribution of the exchange coefficient K_{zz} according to different authors. The values are given for winter and summer conditions and for 50 and 100 mb levels.

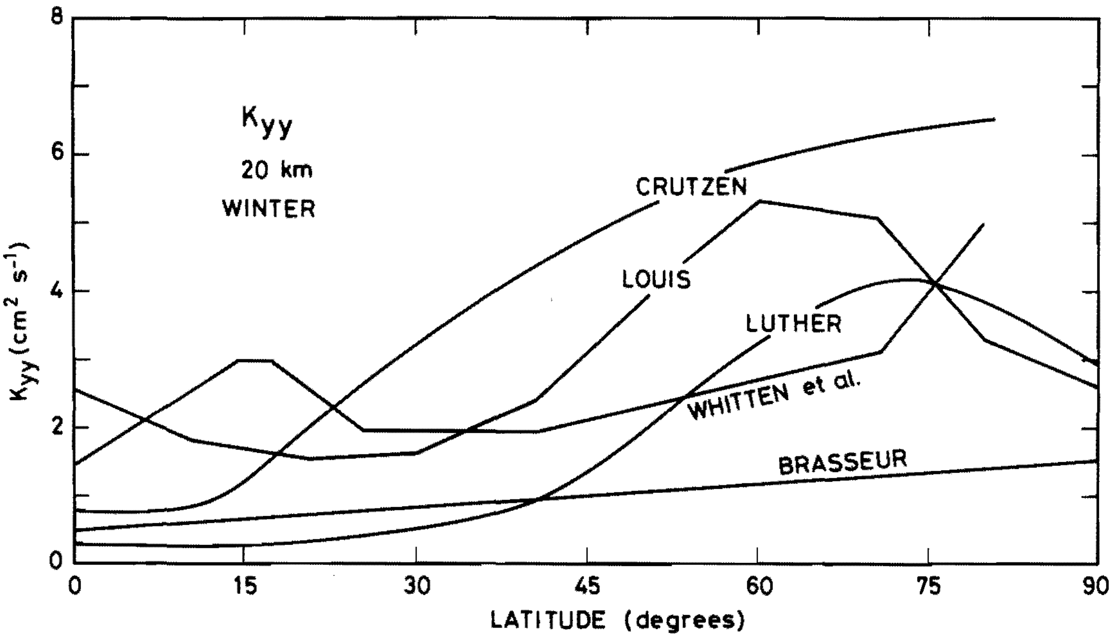

Fig. 7.- Latitudinal distribution of K_{yy} at 20 km adopted during the winter season in various stratospheric models.

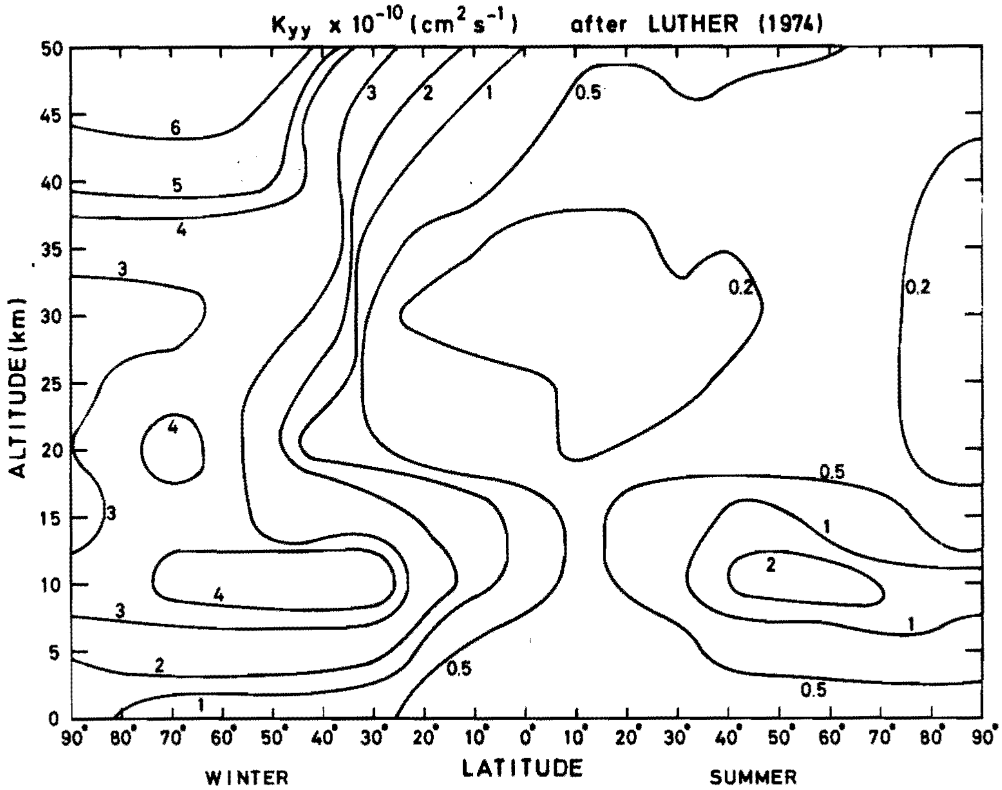

Fig. 8.- Meridional distribution of K_{yy} determined by Luther (1974).

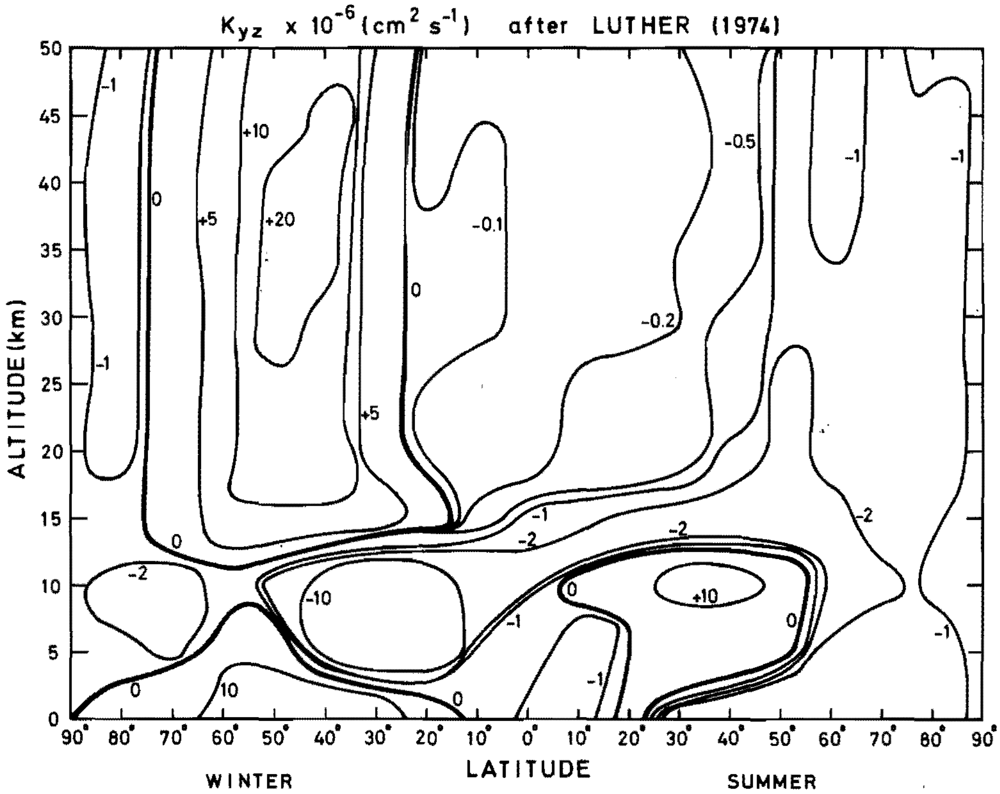

Fig. 9.- Meridional distribution of K_{yz} determined by Luther (1974). The sign of K_{yz} has been chosen so that the corresponding eddy flux is positive when it is directed from the North (winter) pole to the South (summer) pole.

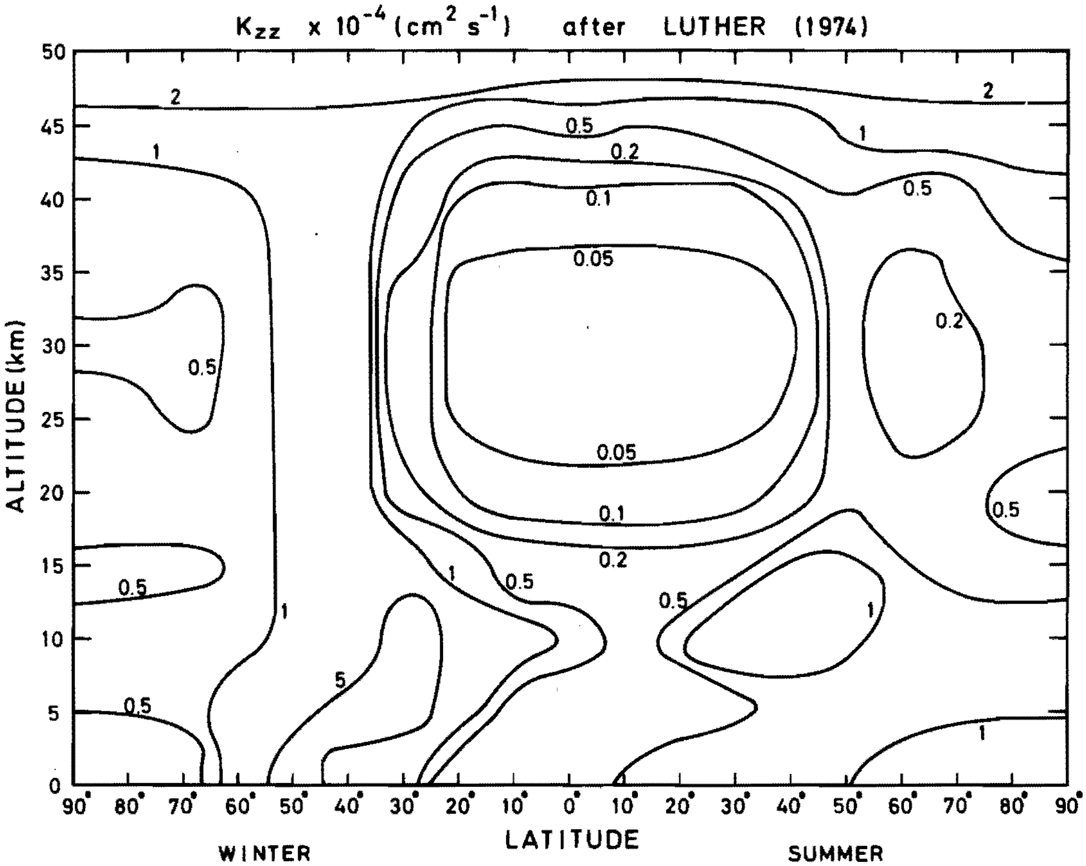

Fig. 10.- Meridional distribution of K_{zz} determined by Luther (1974).

TABLE 1.- Seasonal values of K_{yy} ($10^4\ m^2\ sec^{-1}$) after Nastrom and Brown (1978)

LATITUDE	75	70	65	60	55	50	45	40	35	30	25	20	15	10	5	0	-5
Winter																	
60.0 KM	884	741	566	445	322	220	194	173	223	165	132	131	123	119	100	105	115
57.5	759	681	562	487	395	302	251	219	236	170	137	135	123	126	116	117	112
55.0	634	622	558	529	468	385	307	264	250	174	143	139	123	132	131	129	110
52.5	500	515	513	513	476	405	325	268	233	151	114	113	112	121	101	98	89
50.0	365	409	469	498	484	425	344	272	215	128	86	88	101	110	70	68	68
47.5	312	360	453	446	409	347	279	229	196	127	82	72	79	80	56	54	54
45.0	258	310	438	395	334	269	215	187	177	125	78	57	57	50	41	40	41
42.5	360	399	469	408	330	254	191	153	139	106	76	57	48	40	36	35	33
40.0	462	487	500	420	327	238	165	118	100	88	74	56	39	30	30	29	25
37.5	415	475	469	403	313	219	140	92	78	67	52	40	35	28	26	23	20
35.0	367	462	438	386	298	201	115	65	55	46	31	24	31	26	22	17	15
32.5	258	346	339	299	231	156	91	50	41	38	29	22	24	20	13	10	10
30.0	149	230	239	211	163	111	66	36	26	30	26	20	18	13	4	2	5
Spring																	
60.0 KM	270	223	209	159	116	89	85	105	128	90	79	90	100	127	174	167	140
57.5	296	231	179	141	109	89	81	90	101	73	62	71	84	93	115	106	91
55.0	322	238	149	123	103	88	78	75	74	56	45	53	69	58	56	46	42
52.5	235	199	144	120	97	77	64	60	64	53	48	51	56	52	52	46	41
50.0	148	160	139	118	91	66	50	47	53	50	50	50	42	45	48	47	41
47.5	131	135	132	108	82	61	49	47	54	50	50	48	38	38	41	40	36
45.0	113	110	124	98	74	57	48	49	54	50	49	45	33	30	33	33	31
42.5	109	103	106	85	64	49	40	39	44	44	46	42	29	25	26	26	25
40.0	104	96	88	72	55	41	32	30	33	37	42	39	25	19	19	20	20
37.5	100	95	79	67	53	40	31	27	28	29	29	27	21	18	22	21	19
35.0	96	93	71	62	51	39	29	23	22	22	17	15	17	18	25	23	19
32.5	72	81	70	64	53	38	26	19	18	16	15	13	13	14	20	19	16
30.0	48	70	69	66	54	38	23	14	14	11	12	12	9	10	15	15	12
Summer																	
60.0 KM	193	137	79	61	59	70	87	99	100	96	96	95	102	143	246	232	185
57.5	118	93	65	59	59	63	72	83	90	83	78	74	74	101	165	160	133
55.0	43	49	51	58	58	56	56	66	80	69	59	53	47	60	85	88	81
52.5	35	40	41	46	47	45	45	50	59	54	47	43	43	53	76	77	70
50.0	26	30	31	34	35	34	33	35	39	39	35	34	39	46	68	66	59
47.5	22	24	24	26	26	25	25	27	32	36	35	33	37	42	53	54	52
45.0	18	19	18	18	17	16	16	19	26	34	35	33	33	37	39	42	44
42.5	15	16	14	14	13	12	13	16	22	26	27	26	24	27	33	37	39
40.0	12	13	10	11	10	8	9	13	19	19	20	20	16	17	28	32	34
37.5	12	13	9	10	10	8	7	10	15	15	15	15	13	16	26	28	28
35.0	12	13	9	10	9	7	6	7	11	10	10	10	10	14	23	24	21
32.5	11	10	7	7	6	5	4	6	9	8	9	9	8	12	22	22	20
30.0	9	8	6	4	3	2	2	4	7	7	8	8	6	9	20	21	18
Autumn																	
60.0 KM	792	628	501	334	202	124	99	119	154	135	121	103	87	159	359	332	229
57.5	635	528	442	315	208	141	114	124	147	124	110	101	88	117	222	202	144
55.0	479	428	384	295	214	158	129	129	140	113	100	98	89	76	86	71	58
52.5	393	374	355	276	194	132	101	110	140	106	84	79	68	52	54	46	44
50.0	307	320	325	257	175	106	72	92	141	97	68	60	47	29	21	21	30
47.5	300	293	297	236	170	117	87	89	111	89	67	55	43	29	25	25	29
45.0	292	265	270	216	166	128	102	86	81	80	66	49	40	29	29	28	28
42.5	258	252	244	199	151	110	81	67	67	66	55	41	32	25	25	25	25
40.0	224	238	218	181	136	93	61	47	52	52	44	34	25	20	21	22	22
37.5	196	217	205	168	122	79	48	37	43	39	33	26	19	16	19	19	19
35.0	168	195	191	155	109	66	36	26	33	25	21	19	13	12	17	16	15
32.5	140	162	161	131	92	57	31	22	25	21	17	15	11	10	13	14	14
30.0	111	128	130	105	76	48	27	17	18	17	13	9	9	9	10	11	12

TABLE 2.- Seasonal values of K_{yz} ($10^1\ m^2\ sec^{-1}$) after Nastrom and Brown 1978

LATITUDE	75	70	65	60	55	50	45	40	35	30	25	20	15	10	5	0	-5
Winter																	
60.0 KM	-188	-165	-58	-27	188	340	317	474	381	-88	-238	345	182	-71	-31	3	-32
57.5	-225	-188	-55	-24	184	397	351	622	386	-112	-214	213	117	-62	-24	4	-28
55.0	-261	-211	-52	-21	180	454	675	770	391	-135	-189	80	52	-52	-17	4	-23
52.5	-218	-181	-31	-12	112	314	497	578	326	-78	-128	54	31	-48	-10	3	-17
50.0	-174	-152	-11	-3	44	174	319	386	261	-20	-66	27	9	-43	-4	2	-12
47.5	-168	-145	0	2	-11	24	103	206	154	-22	-26	26	14	-21	-3	0	-6
45.0	-162	-139	12	8	-67	-125	-111	25	48	-25	12	24	18	0	-2	0	0
42.5	-239	-193	1	7	-76	-144	-124	4	33	-27	-2	22	14	0	-1	0	0
40.0	-317	-246	-9	7	-86	-164	-137	-16	18	-28	-17	20	9	0	0	0	0
37.5	-259	-215	-21	4	-76	-143	-109	-11	8	-22	-9	12	4	-1	0	0	0
35.0	-201	-184	-32	2	-66	-122	-81	-7	-1	-16	0	4	0	-3	0	0	0
32.5	-129	-125	-27	0	-44	-81	-47	7	-7	-23	-2	1	1	0	0	0	0
30.0	-56	-66	-22	0	-22	-40	-13	22	-13	-30	-3	-1	2	2	0	0	0
Spring																	
60.0 KM	67	-1	5	-172	-205	-143	-185	-240	-275	-61	60	-660	-536	604	350	-103	-230
57.5	-97	-136	-64	-163	-180	-143	-183	-215	-212	-4	64	-422	-382	375	196	-70	-144
55.0	-262	-272	-135	-155	-156	-144	-181	-191	-148	52	68	-184	-228	146	43	-36	-57
52.5	-139	-153	-87	-127	-133	-112	-131	-138	-122	33	77	-134	-133	103	30	-30	-46
50.0	-16	-35	-39	-99	-110	-81	-81	-85	-95	14	85	-84	-39	60	18	-23	-34
47.5	8	0	13	-33	-60	-51	-57	-66	-91	-4	78	-56	-15	56	18	-15	-25
45.0	32	36	66	32	-9	-21	-34	-48	-87	-23	70	-28	9	53	18	-7	-16
42.5	-12	-10	50	44	17	0	-14	-34	-66	-15	59	-14	19	45	15	-4	-11
40.0	-57	-57	34	56	43	19	4	-21	-45	-6	48	0	29	38	12	-1	-6
37.5	-98	-105	-8	31	43	26	15	-7	-26	4	35	-8	7	29	11	-2	-6
35.0	-139	-154	-50	6	42	33	25	7	-7	16	21	-15	-13	19	11	-3	-6
32.5	-114	-155	-72	-18	29	27	20	7	1	17	18	-12	-12	7	5	0	-1
30.0	-89	-155	-94	-43	16	21	15	7	10	17	15	-9	-11	-5	0	3	3
Summer																	
60.0 KM	0	0	-3	-10	-18	-24	-9	41	43	15	11	-28	114	156	27	17	137
57.5	0	0	-3	-8	-14	-18	-9	23	25	18	29	-17	64	96	17	10	88
55.0	0	0	-3	-7	-10	-11	-8	5	6	22	46	-6	14	36	8	4	39
52.5	0	0	-2	-4	-6	-6	-5	2	0	15	36	-9	11	35	8	2	29
50.0	0	0	-1	-2	-2	-2	-1	-1	-7	8	25	-11	8	35	8	1	19
47.5	0	0	0	-1	-1	-1	0	0	-7	5	25	-9	6	27	6	1	15
45.0	0	0	0	0	0	0	0	0	-6	2	25	-6	5	20	4	0	11
42.5	0	0	0	0	0	0	0	0	-4	1	14	-3	4	12	2	0	6
40.0	0	0	0	0	0	0	0	0	-1	0	4	0	3	4	1	0	2
37.5	0	0	0	0	0	0	0	0	-1	0	2	0	1	2	0	0	0
35.0	0	0	0	0	0	0	0	0	0	1	1	0	0	0	0	0	-1
32.5	0	0	0	0	0	0	0	0	0	1	2	0	0	0	0	0	0
30.0	0	0	0	0	0	0	0	0	0	0	3	-1	0	0	0	0	0
Autumn																	
60.0 KM	10	296	200	210	182	162	112	99	67	0	36	-90	-135	278	186	-351	-641
57.5	8	245	191	207	172	154	106	88	51	0	26	-69	-97	161	100	-207	-377
55.0	5	194	182	204	161	146	101	76	35	0	17	-48	-60	45	13	-64	-114
52.5	4	140	133	147	109	95	65	52	25	0	9	-29	-33	27	8	-36	-71
50.0	2	86	85	91	57	44	28	27	16	0	2	-11	-6	9	4	-8	-29
47.5	1	61	52	46	18	11	12	17	8	0	3	-7	-1	14	9	-10	-32
45.0	1	36	20	1	-20	-21	-3	8	1	0	5	-2	3	19	13	-12	-35
42.5	0	30	18	-2	-28	-28	-7	4	0	0	4	1	6	16	9	-6	-20
40.0	0	23	17	-7	-36	-35	-11	0	0	0	3	5	9	12	5	0	-4
37.5	0	19	18	-2	-29	-30	-10	-1	-1	0	3	3	6	10	5	0	-6
35.0	0	16	19	1	-23	-25	-8	-2	-1	0	2	0	3	8	5	-2	-8
32.5	0	10	14	2	-15	-18	-7	-2	-1	0	1	0	2	5	3	-1	-6
30.0	0	4	9	3	-7	-11	-6	-3	-1	0	1	0	1	2	1	0	-3

TABLE 3.- Seasonal values of K_{zz} (10^3 cm^2 sec^{-1}) after Nastrom and Brown 1978

LATITUDE	75	70	65	60	55	50	45	40	35	30	25	20	15	10	5	0	-5
Winter																	
60.0	1050	1000	1419	1575	1600	1550	1475	1375	1150	1100	1050	1125	1375	1681	2100	2300	2450
57.5	800	760	1161	1313	1413	1333	1280	1200	1050	975	413	963	1125	1329	1575	1750	1875
55.0	550	520	903	1050	1225	1115	1085	1025	950	850	775	800	875	976	1050	1200	1300
52.5	450	415	650	763	875	845	825	773	708	643	605	615	663	724	755	850	950
50.0	350	310	396	475	525	575	565	520	465	435	435	430	450	470	460	500	600
47.5	310	280	321	378	410	448	423	383	325	310	315	328	345	367	375	390	425
45.0	270	250	245	280	295	320	280	245	185	185	195	225	240	263	290	280	250
42.5	210	190	173	205	225	225	180	150	113	115	135	163	175	183	195	185	165
40.0	150	130	100	130	155	130	80	55	40	45	75	100	110	103	100	90	80
37.5	110	96	69	83	104	84	52	37	27	31	51	67	81	76	65	60	57
35.0	70	62	39	36	51	37	24	18	13	17	26	33	51	49	30	30	33
32.5	51	45	31	28	34	26	19	17	15	18	21	24	36	34	20	22	26
30.0	32	28	23	20	17	15	13	15	17	19	16	14	20	19	10	14	18
Spring																	
60.0	650	600	456	395	390	410	495	580	700	815	1000	975	775	669	700	900	1050
57.5	550	485	366	318	330	363	443	528	638	758	893	825	650	564	585	750	950
55.0	450	370	275	240	270	315	390	475	575	700	785	675	525	459	470	600	850
52.5	380	325	256	233	255	298	355	418	503	595	633	543	438	415	450	540	685
50.0	310	280	236	225	240	280	320	360	430	490	480	410	350	370	430	480	520
47.5	285	255	210	195	205	233	270	308	358	393	385	333	290	311	365	385	410
45.0	260	230	184	165	170	185	220	255	285	295	290	255	230	253	300	290	300
42.5	200	180	154	140	140	148	165	185	203	205	213	195	178	166	165	185	205
40.0	140	130	124	115	110	110	110	115	120	115	135	135	125	79	30	80	110
37.5	91	87	88	77	71	75	76	75	76	76	93	95	83	56	25	51	70
35.0	41	43	50	38	32	40	41	34	31	36	51	55	40	33	20	22	30
32.5	31	30	35	25	21	25	26	24	24	28	38	40	31	27	20	21	28
30.0	20	17	20	12	10	10	10	13	17	20	24	23	22	20	19	20	25
Summer																	
60.0	550	480	431	425	475	515	480	455	685	1120	1330	1525	1500	1338	1300	2000	2150
57.5	485	440	385	395	428	430	390	365	488	755	1010	1170	1163	1047	975	1550	1975
55.0	420	400	339	365	380	345	300	275	290	390	690	815	825	756	650	1100	1800
52.5	385	365	298	310	330	303	255	225	258	328	518	625	620	559	475	800	1350
50.0	350	330	256	255	280	260	210	175	225	265	345	435	415	361	300	500	900
47.5	285	295	228	215	238	205	153	128	183	235	290	343	300	265	200	335	625
45.0	220	260	199	175	195	150	95	80	140	205	235	250	185	169	100	170	350
42.5	165	190	150	135	145	113	70	78	128	163	170	168	133	124	75	125	225
40.0	110	120	101	95	95	75	45	75	115	120	105	85	80	78	50	80	100
37.5	71	78	73	73	75	61	44	61	78	77	67	57	57	57	37	54	67
35.0	31	35	43	50	53	46	43	47	41	34	29	28	34	35	24	28	33
32.5	25	28	33	37	38	31	27	30	31	27	25	25	29	30	23	23	23
30.0	19	20	22	23	23	16	11	13	20	20	20	23	24	25	21	17	13
Autumn																	
60.0	570	590	960	1110	1040	915	835	825	925	1075	1375	1525	1300	1500	2100	1800	1450
57.5	535	540	848	945	880	800	723	658	695	825	1138	1300	1193	1377	1900	1650	1250
55.0	500	490	735	780	720	685	610	490	465	575	900	1075	1085	1254	1700	1500	1050
52.5	410	410	594	628	588	570	510	438	423	488	690	875	910	1024	1350	1175	825
50.0	320	330	451	475	455	455	410	385	380	400	480	675	735	794	1000	850	600
47.5	250	265	318	328	320	330	330	325	315	333	385	500	570	612	750	645	465
45.0	180	200	185	180	185	205	250	265	250	265	290	325	405	429	500	440	330
42.5	150	160	140	128	128	145	190	200	185	190	210	243	295	314	350	300	230
40.0	120	120	94	75	70	85	130	135	120	115	130	160	185	199	200	160	130
37.5	105	95	75	58	56	66	88	92	86	80	90	110	126	133	132	107	88
35.0	90	70	56	40	42	46	46	48	52	44	49	60	65	66	64	53	45
32.5	58	47	40	30	30	31	32	34	37	34	36	42	45	46	46	38	31
30.0	25	24	25	19	18	16	17	20	23	23	22	23	25	27	28	22	17

increases with height above 30 km. One also notes a latitudinal variation below 45 km but, as shown also in the Nastrom and Brown data, the patterns of the K's tend to be more or less horizontal in the upper stratosphere and lower mesosphere. The cross sections represented here refer to zonal mean values. However, as shown by Nastrom and Brown and illustrated in figure 11, the values of the exchange coefficients may be quite different at two separate longitudes.

6. EDDY DIFFUSION AND OZONE TRANSPORT: In order to test the effect of each eddy diffusion component on the distribution of an atmospheric trace gase, such as ozone, different computations have been carried out with a two-dimensional numerical model. The full description of this model - including the chemical scheme - with its two versions has been given by Brasseur (1976; 1978). Firstly, one considers a steady state approach with a very simple transport parametrization. The action of the mean circulation is neglected and the dynamics is described only by the three eddy diffusion coefficients. In order to oversimplify the conditions, the following constant and uniform values are adopted : $K_{yy} = 10^{10}$ cm^2 s^{-1} and $K_{zz} = 10^4$ cm^2 s^{-1}. Moreover, K_{yz} is adjusted in the winter and summer hemisphere until the calculated ozone distribution becomes compatible with the observations.

Figure 12 shows the meridional cross section of the ozone concentration when photochemical equilibrium conditions are prescribed (all K's are put equal to zero). In this case, the maximum concentration is located in the equatorial and tropical regions and almost no ozone is present below 10 km or at high latitudes. This is in contradiction with the reality.

When the vertical coefficient $K_{zz} = 10^4$ cm^2 s^{-1} is introduced while the other K's remain equal to zero (figure 13), ozone is present in the lower stratosphere (and troposphere) but its concentration remains insignificant at high latitudes. When the computation is performed with $K_{yy} = 10^{10}$ cm^2 s^{-1} and $K_{zz} = 10^4$ cm^2 s^{-1} (figure 14) a horizontal flux appears and ozone penetrates in ψ the high latitude regions. However, the maximum concentration still occurs in the equatorial zone where O_3 is produced photochemically, which is in contradiction with the observation.

The existence of a countergradient flux becomes possible only with the introduction of the off-diagonal component K_{yz}. Fig. 15 shows the latitudinal variation of total ozone obtained for different values of K_{yz}. It clearly shows that the ozone distribution is very sensitive to K_{yz}, particularly at high latitudes. Therefore, it should be determined with a very high precision. Because of the high sensitivity of the distribution of ozone to K_{yz} and because of the rather large uncertainty on K_{yz}, it is most necessary to "tune" this coefficient with care until the distribution of trace species and/or temperature comes into agreement with the observation. It should be noted, however, that the solution is not unique and the results depend on the other parameters which are adopted, and especially the mean motion and the other K's. Further, it is not proven, but only assumed by most modellers, that the same K's may be used for all the different trace species of the atmosphere. This is only a first order approximation since the theory by Reed and German has its own limitations and assumes that the physical processes governing the transport are the same for all of the different atmospheric species. Adopting the latitudinal distribution of K_{yz} shown in figure 16, the meridional distribution of O_3 as illustrated in figure 17 is obtained.

In order to give a crude estimation of the relative effect of the mean and turbulent transport of ozone, we now consider a second and more elaborate version of the 2-D model. The mean circulation as computed by Cunnold et al. (1974) is now introduced in the model while the eddy diffusion coefficients are adjusted at all latitudes and altitudes. Figure 18 gives some information concerning the distributions of these K's. In order to visualize the action of both types of transport, figure 19, 20 and 21 present, respectively, the mean, turbulent and total transport derived with the model calculation and show that the poleward ozone flux in winter is only possible if the horizontal (countergradient) transport by eddies is taken into account. In fact according to these calculations, horizontal mean motions play a significant role in the equatorial and polar regions while large scale turbulent transport is clearly dominant in the midlatitude zone. Vertical winds in the Hadley cell near the equatorial tropopause prevent ozone from diffusing downward.

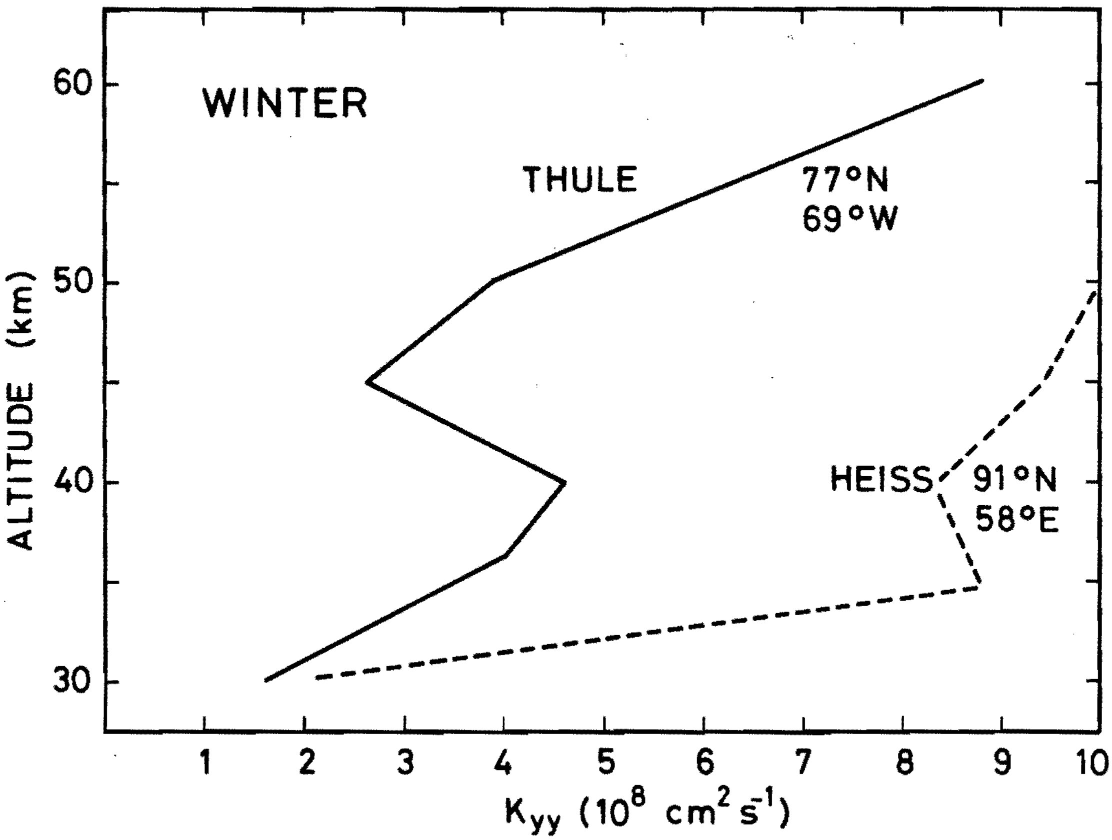

Fig. 11.- Comparison of K_{yy} during winter at Thule and Heiss. From Nastrom and Brown (1978).

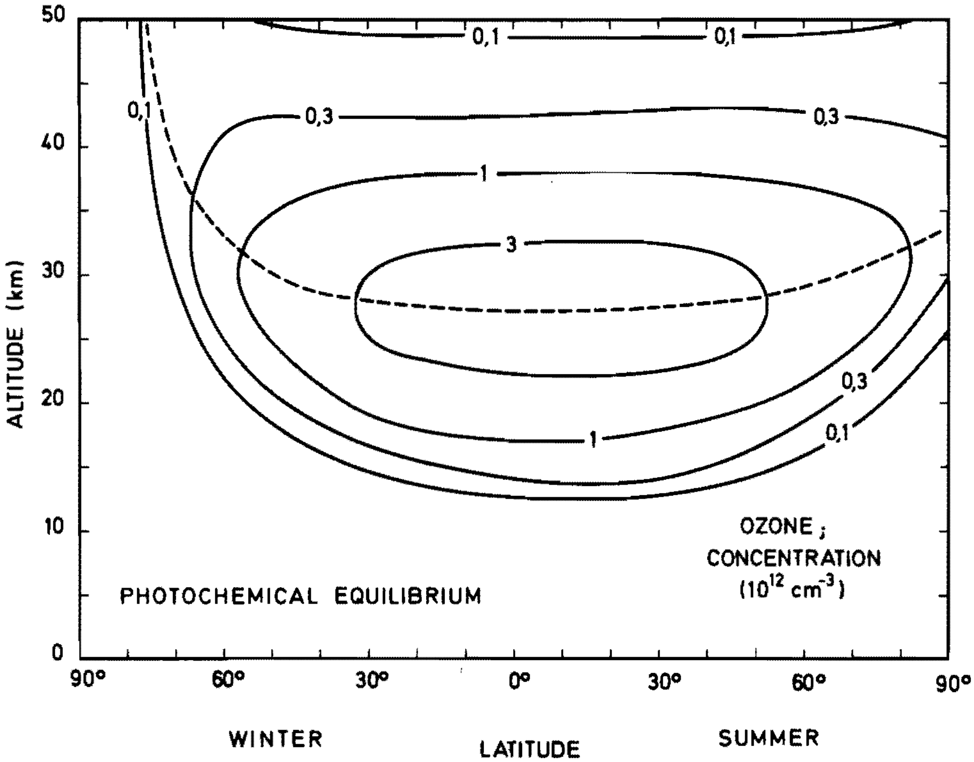

Fig. 12.- Meridional distribution of the ozone concentration assuming photochemical equilibrium conditions.

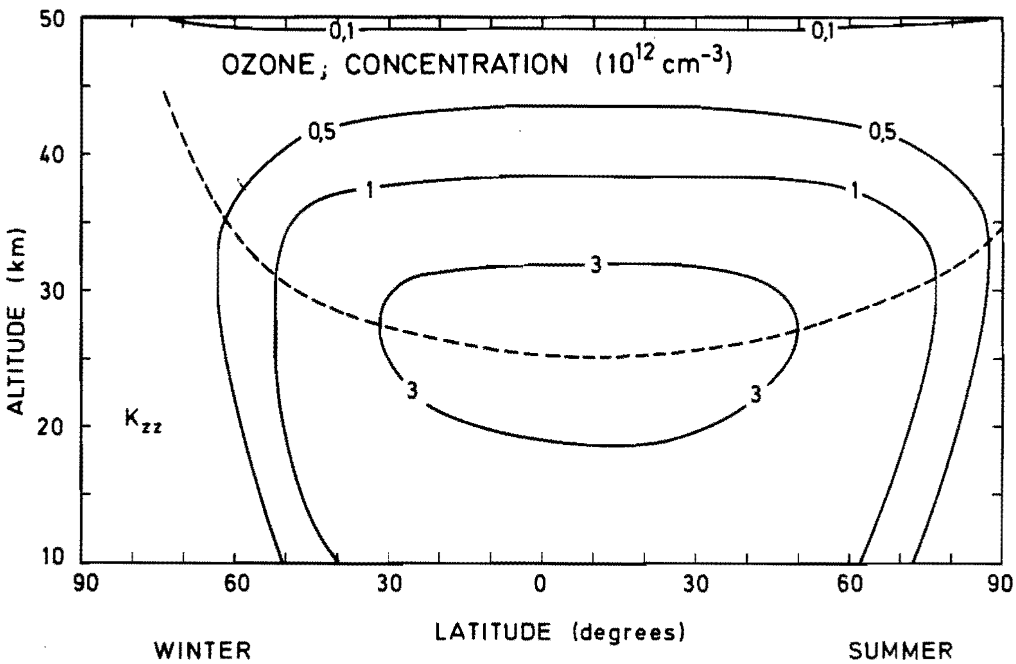

Fig. 13.- Meridional distribution of the ozone concentration when vertical exchange by diffusion is only taken into account.

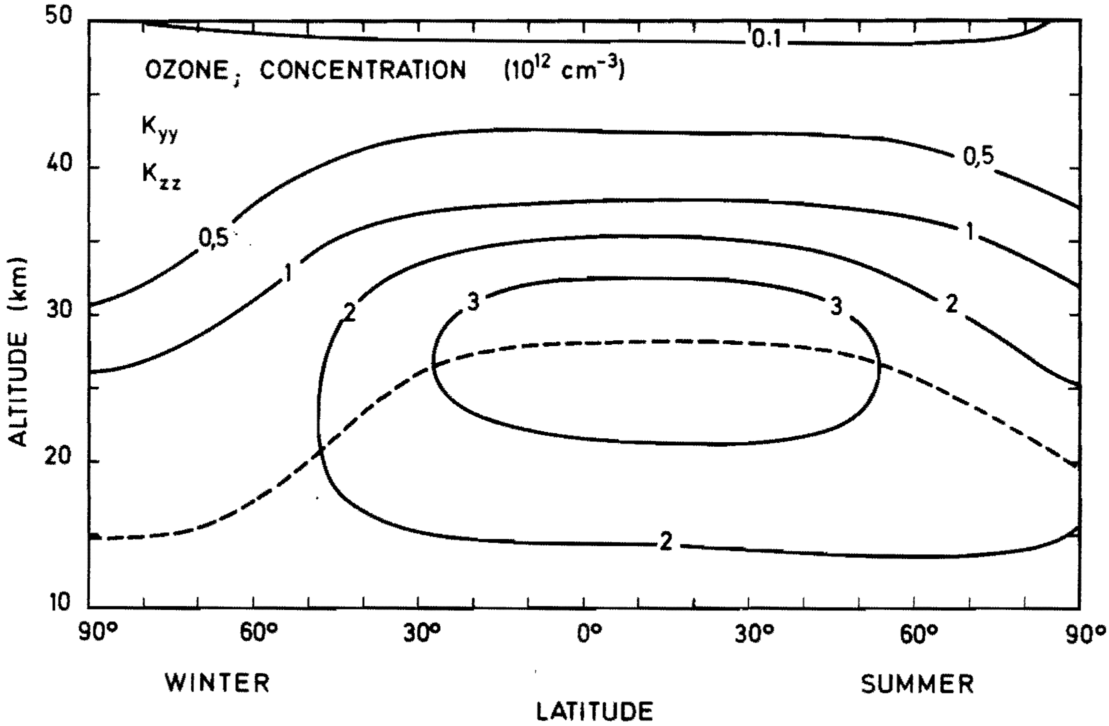

Fig. 14.- Meridional distribution of the ozone concentration when vertical and horizontal transport are taken into account, and are parameterized by K_{yy} and K_{zz} only.

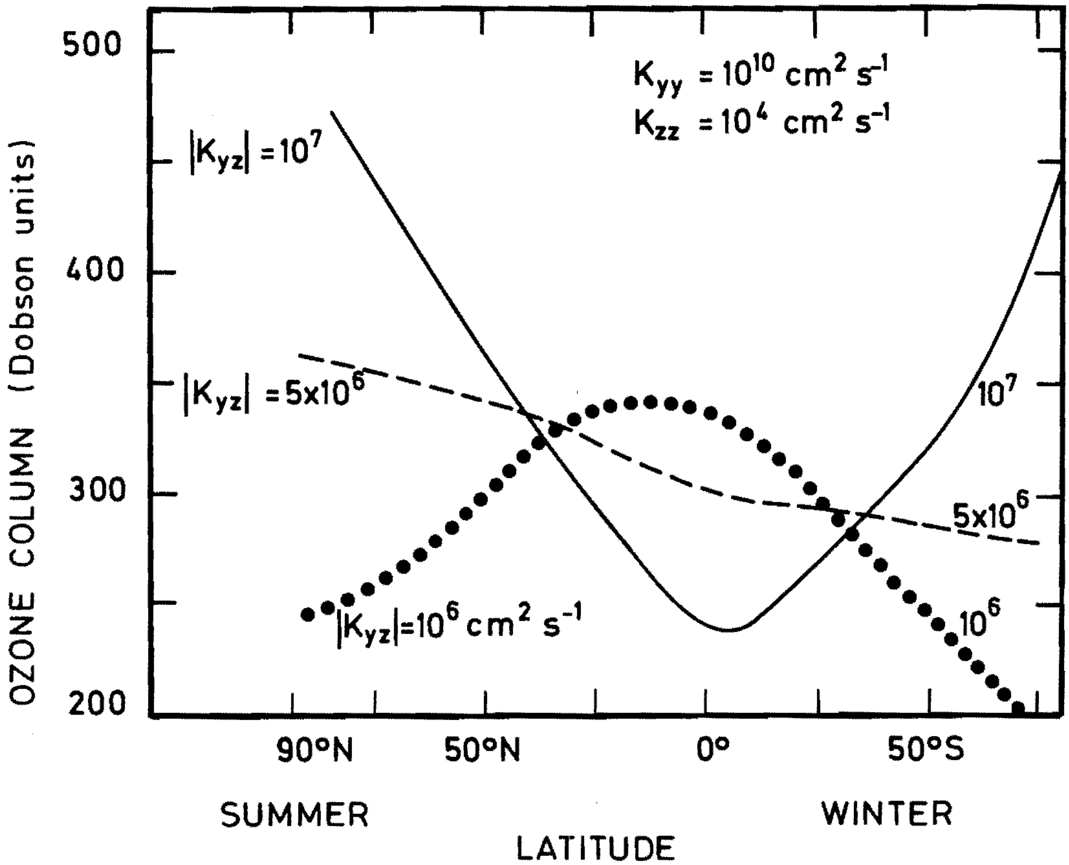

Fig. 15.- Effect of the anisotropic component K_{yz} on the latitudinal distribution of total ozone.

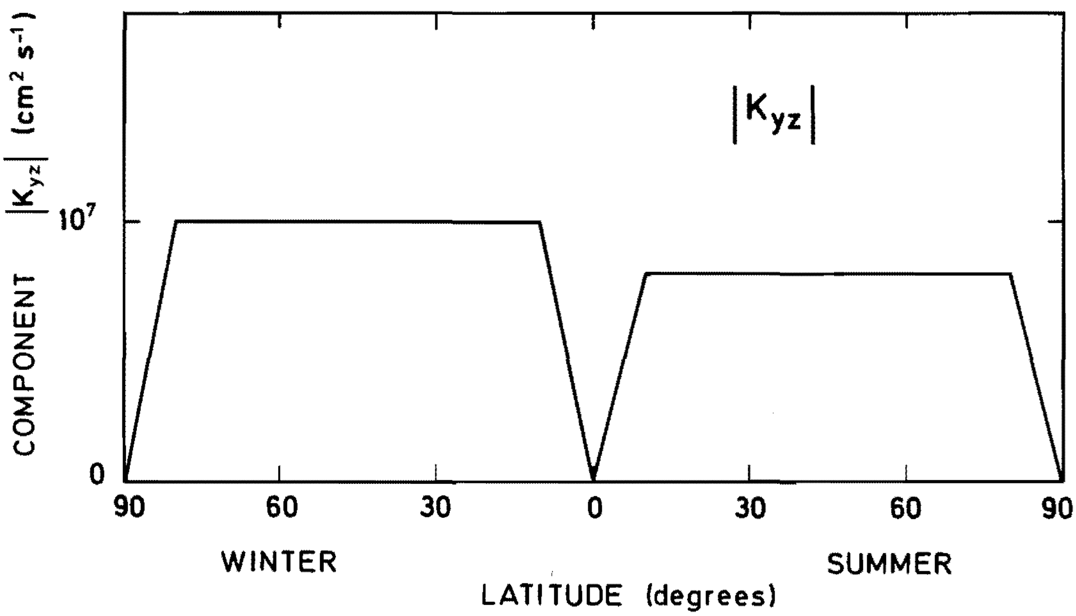

Fig. 16.- Example of a simple latitudinal distribution of $|K_{yz}|$ in the stratosphere.

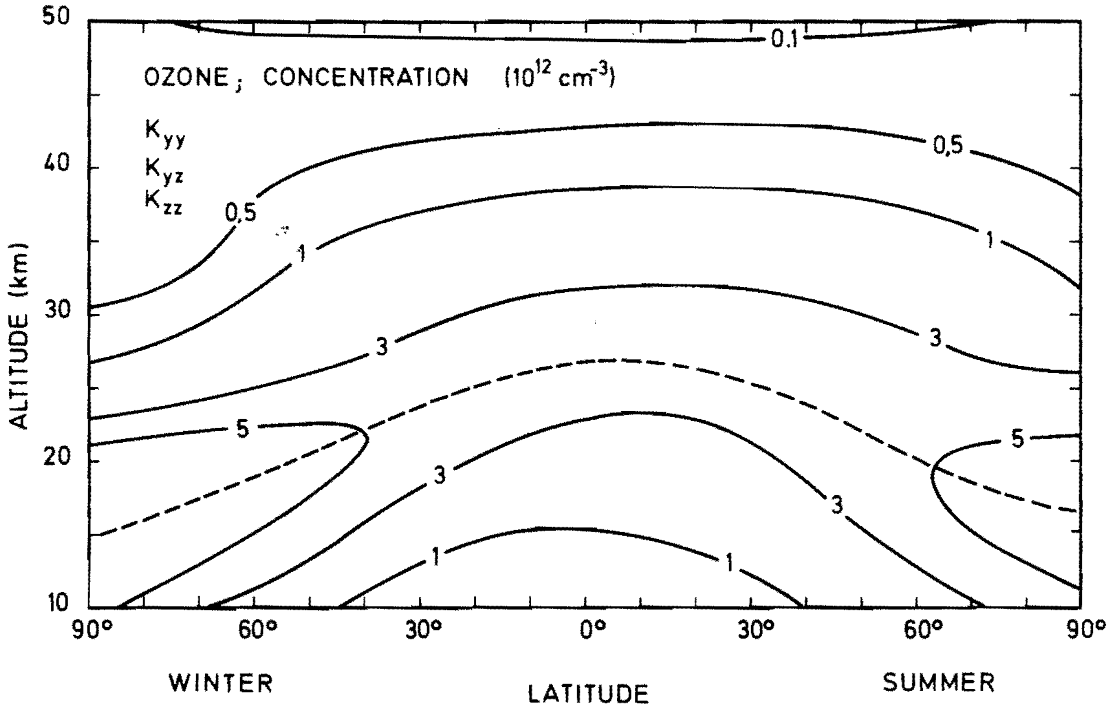

Fig. 17.- Meridional distribution of the ozone concentration when the transport is parameterized by the three components K_{yy}, K_{yz} and K_{zz}.

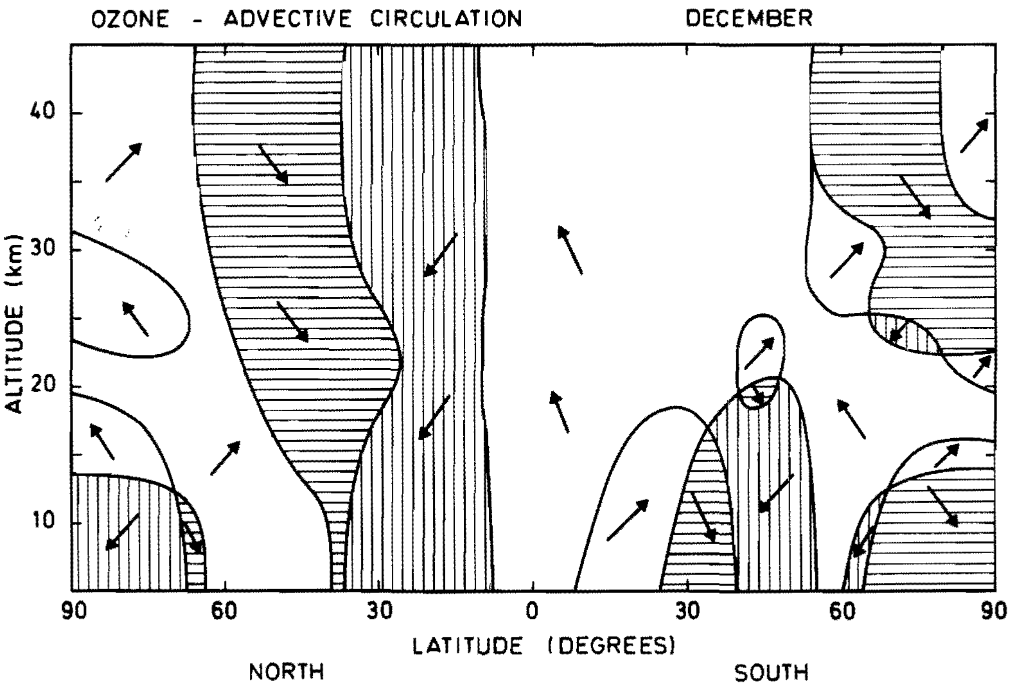

Fig. 19.- Representation of the circulation of ozone by mean motions (v, w).

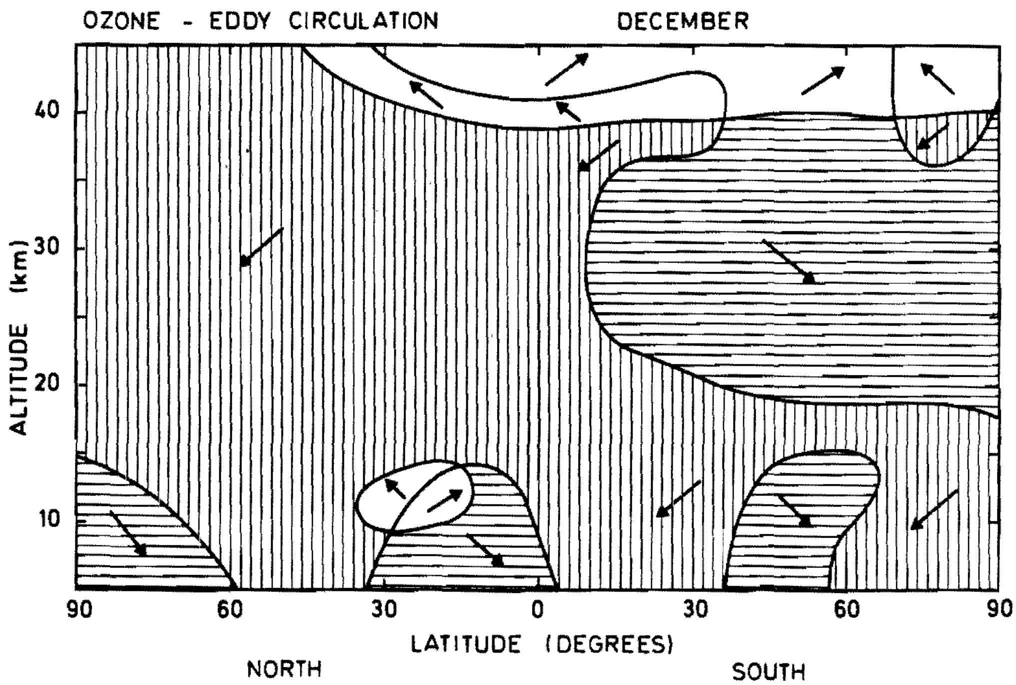

Fig. 20.- Representation of the circulation of ozone by large scale eddy diffusion (K_{yy}, K_{yz}, K_{zz}).

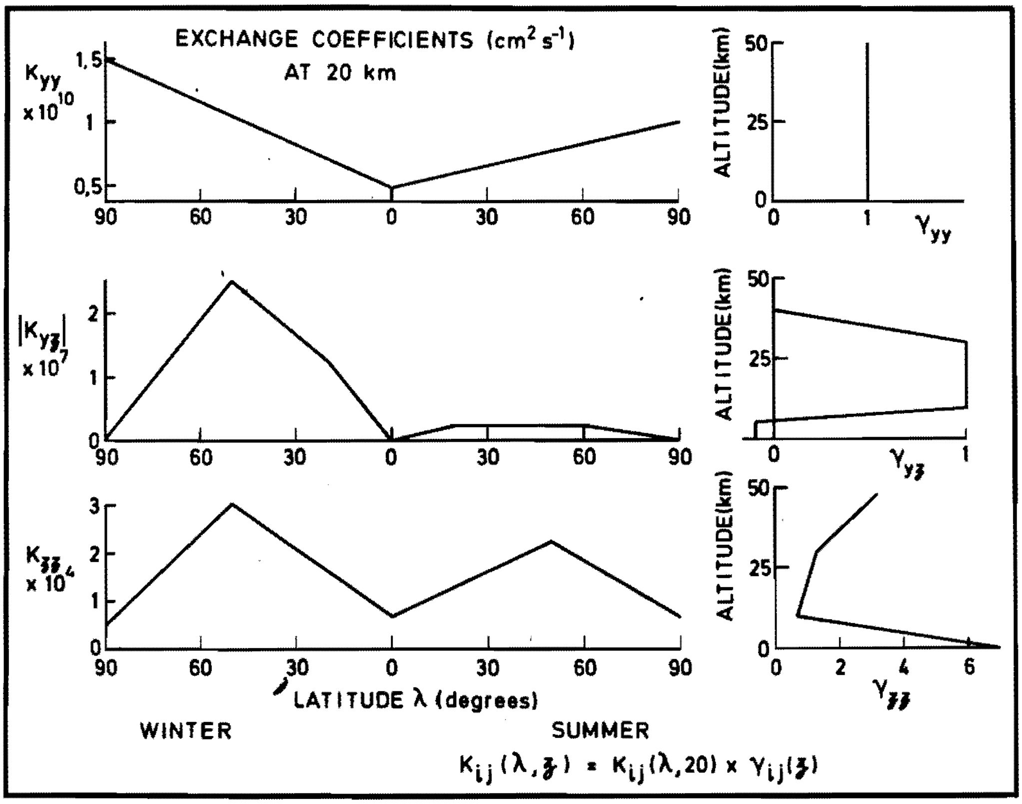

Fig. 18.- Distribution with latitude and altitude of the exchange coefficients adopted in the 2-D model used in this work.

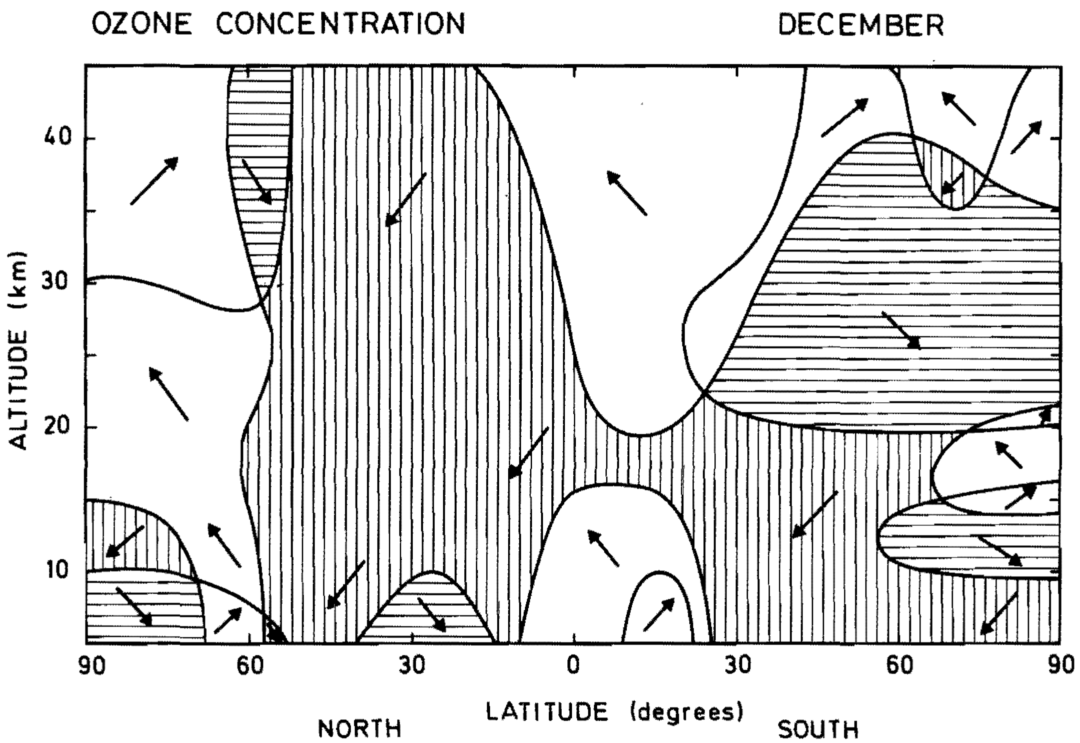

Fig. 21.- Representation of the global circulation of ozone by mean motions and eddy diffusion.

7. VERTICAL 1-D TRANSPORT IN THE ATMOSPHERE: In many aeronomic studies, the problemof the behavior of minor constituents is treated by assuming average conditions over all latitudes and longitudes. In one-dimensional models, which are useful in the estimation of the dominant chemical and photochemical processes as a function of the altitude, the continuity equation becomes

$$\frac{\partial n}{\partial t} + \frac{\partial \phi}{\partial z} = P - L \qquad (35)$$

where it is now assumed that all the quantities are averaged over the entire globe. In this equation, the contribution to the flux is due to large scale eddy mixing; the mean circulation does not appear since, for continuity reasons, the average vertical wind must be equal to zero. Again, the continuity equation (35) requires a closure condition and one assumes that a vertical flux of any minor constituent takes place when the distribution of this species departs from constant mixing ratio. The following equation, indicating that the net vertical flux is proportionnal to the negative gradient of the mixing ratio,

$$\phi = - K\, n(M) \frac{\partial f}{\partial z} \qquad (36)$$

is adopted since it requires that the constituent moves from regions where it has a high mixing ratio to regions where it is low. In this expression, K is a vertical exchange coefficient which refers to global conditions (average over all latitudes and longitudes). This formalism for vertical 1-D transport has been introduced by Lettau (1951) and adopted by Colegrove et al. (1966) to study the transport of oxygen in the lower thermosphere. The vertical flux can also be written in the alternative forms

$$\phi = - K \left[\frac{\partial n}{\partial z} + \frac{n}{H} + \frac{n}{T}\frac{\partial T}{\partial z} \right] \qquad (37)$$

or

$$\phi = - K\, n \left[\frac{1}{H} - \frac{1}{H_1} \right] \qquad (38)$$

where T is the temperature, H the atmospheric scale height and H_1 the scale height of the species being considered.

It should be noted that, while the form of these flux representations can be intuitively understood from the Prandtl's mixing length theory, there is no complete and fundamental theoretical explanation for an expression such as (36). There has been much confusion in the past in the interpretation of the physical sense of the K coefficient when it has been attempted to derive its absolute value from turbulence measurements. In fact, the vertical eddy-mixing coefficient is generally obtained without any explicit reference to the motions and it must be considered as a pure phenomenological parameter. K is simply a proportionality factor relating the flux to the gradient of the mixing ratio.

Studies of the dispersion processes in the mesosphere and the lower thermosphere has been undertaken by different methods, namely using radio meteor trails (e.g. Roper and Elford, 1963; Roper, 1966; Zimmerman, 1973; 1974; Cunnold, 1975) or chemical release observation (e.g. Blamont and de Jager, 1961; Zimmerman and Champion, 1963; Justus, 1969; Zimmerman and Trowbridge, 1973). Values for a diffusion coefficient have been derived in several cases. A profile of the coefficient for the vertical eddy diffusion of heat (which is of the same order of magnitude as the exchange coefficient of trace species) for the region between 50 and 100 km has been deduced by Johnson and Wilkins (1965) based upon the downward flux required to maintain the thermal structure of this atmospheric region. These results were questioned, however, by Hunten (1974) since they did not take into account the heat input associated with the turbulence itself. Estimates of K due to small scale motions and, in particular, to internal gravity waves have been undertaken by Hodges (1969) and Hines (1970) while Justus (1973) has used Hines' theory in conjunction with wind observations to derive the profile of K. Lindzen (1971) has proposed values of K associated with atmospheric tides and Zimmerman (1973; 1974) has analyzed wind observations. Finally, exchange coefficient profiles have been deduced from the vertical distribution of long lived chemical species such as atomic oxygen in the 90-100 km region (Colegrove et al, 1965; da Mata, 1974). Adjustments of the K profiles have been made in most models when studying species such as NO (Strobel, 1971; Brasseur and Nicolet, 1973); CO (Hays and Olivero, 1970). Figure 22 illustrates different distributions of exchange coefficients in the mesosphere and lower thermosphere.

In the stratosphere and the troposphere where the pattern of vertical transport appears essentially to be determined by the meridional motions, the 1-D K profile should be, in principle, derived from elaborate circulation models (see e.g. Mahlman, 1975). However, an order of magnitude profile can be deduced from residence time (τ) considerations since it can be derived from the diffusion equations that

$$K \cong \frac{H^2}{\tau} \tag{39}$$

where H is a typical length, here the atmospheric scale height. Studies concerning the decay of radioactive debris from nuclear explosions have shown that the residence time is of the order of 2 years in the stratosphere while it is of the order of 1 month in the troposphere (see e.g. Reiter et al, 1975). Therefore, typical values for K are 2×10^5 cm^2 s^{-1} below the tropopause and between 10^3 and 10^4 cm^2 s^{-1} above this transition region.

The vertical distribution of the exchange coefficient in the stratosphere can in principle be obtained by inverting the continuity/ transport equation (derived from 35 and 36). If the distribution of the production, the loss rates and the concentration of a tracer are known, it is possible to determine a corresponding K profile. Since the exchange coefficient characterizes a physical state of the atmosphere, it is usually assumed to be independent of particular choices of the species. Also, to make sense the different parameters adopted for the inversion (concentration, etc...) must be globally averaged values. Constituents with horizontal stratification are thus very useful for this type of calculation.

Two types of atmospheric tracers have been used to derive vertical profiles of K : chemically reactive gases such as N_2O or CH_4 or chemically inert radionucleides introduced in the stratosphere by nuclear explosions.

a. CH_4 and N_2O satisfy the conditions for applicability of one-dimensional eddy treatment since they are rather uniformly distributed in the horizontal and since their chemical loss mechanisms are relatively simple. Moreover, these two constituents are only produced at ground level and, therefore, the exchange coefficient profile is given by

$$K(z) = \frac{-\phi}{n(M)\ df/dz} = \frac{-\int_z^\infty L\ dz}{n(M)\ df/dz} \tag{40}$$

where ϕ is the vertical flux, L the atmospheric destruction rate, f the volume mixing ratio and n(M) the total concentration.
Since, in general, large uncertainties remain in the determination of the global mixing ratio and the integrated loss rate, K cannot be derived without significant errors. Moreover, in the lower stratosphere and in the troposphere where n(M) becomes large and df/dz small for constituents such as CH_4 and N_2O, this formula can no longer be applied. Hunten (1975) has used the methane data obtained by Ehhalt et al. (1972) to determine a

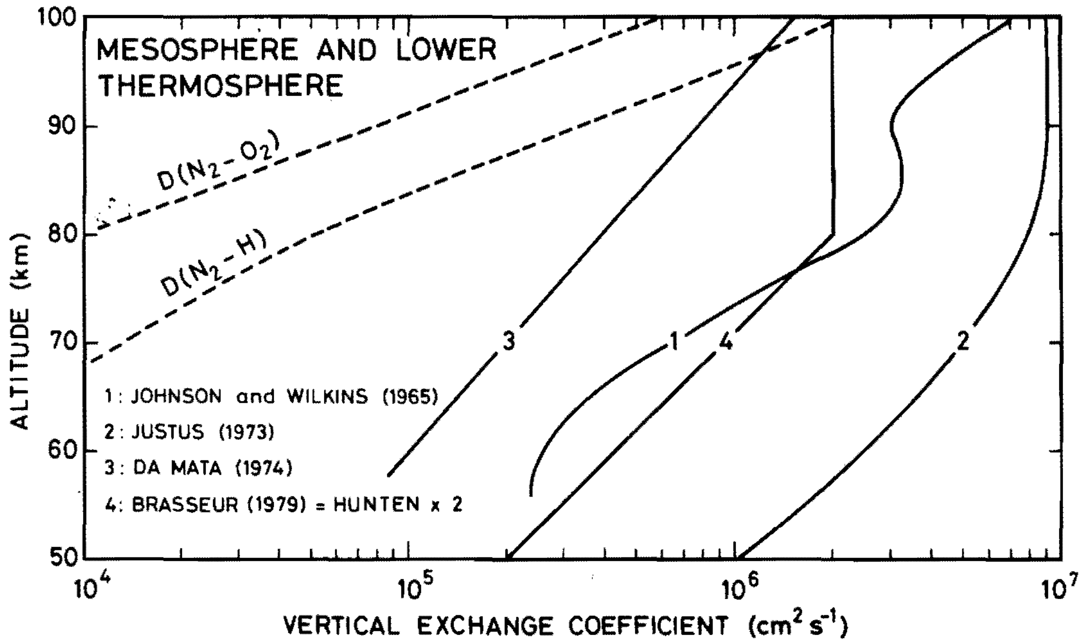

Fig. 22.- Vertical distribution of 1-D exchange coefficients K adopted in different mesospheric models. For comparison purposes, several molecular diffusion coefficients D are also shown.

K profile (figure 23) and has revised an earlier study by Wofsy and McElroy (1973). Dickinson (1976) has carefully analyzed the variability in the K profiles arising from differences in data interpretations. Other profiles have been suggested by various modelers (Liu and Cicerone, 1976; Crutzen and Isaksen, 1978; etc...) but recently, NASA (1977) has suggested consideration of whether to adopt an average of the Dickinson's results or the distribution given by Hunten but multiplied by a factor of 2. This last correction was introduced because the original Hunten's profile did not produce a chemical loss rate of CH_4 which is consistent with that used in its derivation.

b. Tracers injected by nuclear explosions as fine particles (e.g. Sr^{90}, W^{185}, Rh^{102}, Cd^{109} and Zr^{95}) or as a true gas (C^{14}) will provide useful information on stratospheric transport since they are not associated with any chemical source or sink (except the well understood radioactive decay). However such tracers are not uniformly distributed and because of the uncertainties in the meridional distributions (obtained by particle sampling) and due to the difficulties caused by the transient nature of the removal from the stratosphere and by the sendimentation of these particles, this method, which has been analyzed by Chang (1975), raises serious questions and does not provide more feasible results than those associated with chemically active species.

Figure 24 illustrates several profiles of exchange coefficients. Significant differences still occur which limit the validity of 1-D model calculations. To estimate the effect of transport uncertainties on chemical model results, figure 25 (Nicolet and Peetermans, 1972) shows the vertically integrated NO production rate in the stratosphere as a function of the vertically uniform eddy mixing coefficient K used in the calculation. Variations of about a factor of 10 occur. Also, figure 26 (NAS report, 1976) illustrates the different responses in the total ozone concentration to constant release of chlorofluoromethanes in the atmosphere until 1978 when release is suddenly and completely stopped. Again the results calculated with different K profiles differ significantly.

Finally, it should be clear that since the 1-D profile refers to globally average conditions, it cannot satisfactorily represent physical processes related to the details of the atmospheric dynamics, e.g. the formation of tropopause structure or the slope of the mixing surfaces in the lower stratosphere. Also, properties associated with the time variability of the atmospheric conditions are smoothed out by such 1-D approaches. For example, the vertical distribution of water vapor with the discontinuity in its scale height at the tropopause cannot be adequately represented in any 1-D model. Also, as explained by Newell (1977), carbon monoxide distributions can apparently be explained without invoking the 1-D model results that predict large sources from methane. Finally, the ozone distribution and budget can not be adequately described unless one adopts at least a 2-D representation.

8. SUMMARY: The so-called eddy diffusion coefficients are purely phenomenological but useful empirical parameters relating the mean flux to the gradient of the mixing ratio. When treating the transport of minor constituents in chemical models, the K-theory is very convenient but not theoretically verifiable. However, it leads to rather satisfactorily results which should be considered as first approximations. More work is required to improve this parametrization and to introduce a more elaborate - but still handy - treatment of all scales of motions based on dynamical considerations. In the mean time, the K coefficients have to be deduced from the best known distributions of trace species and assumed to be independent of the choice of the minor constituents.

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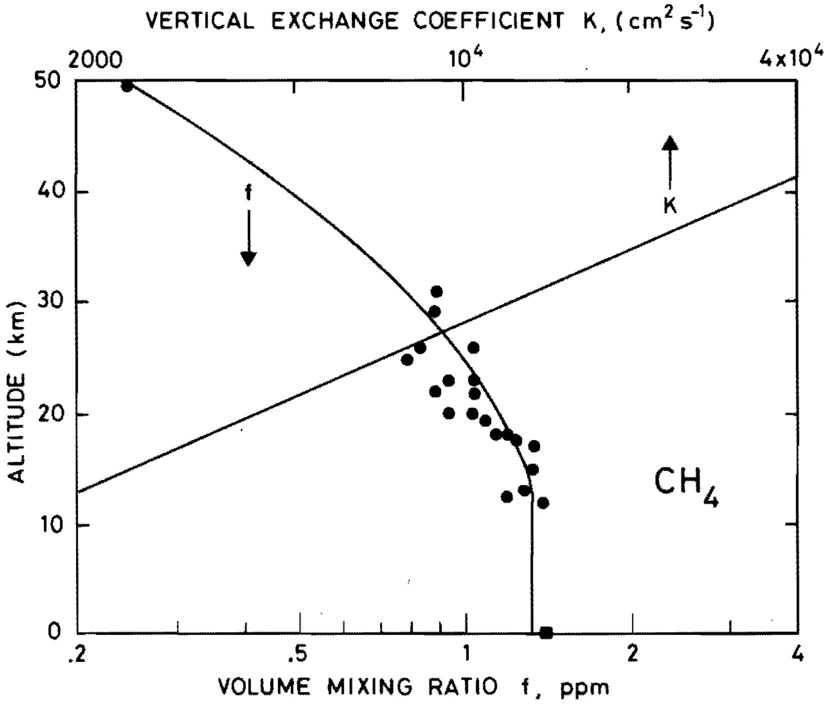

Fig. 23.- Stratospheric exchange coefficient profile derived by Hunten (1975) from methane data.

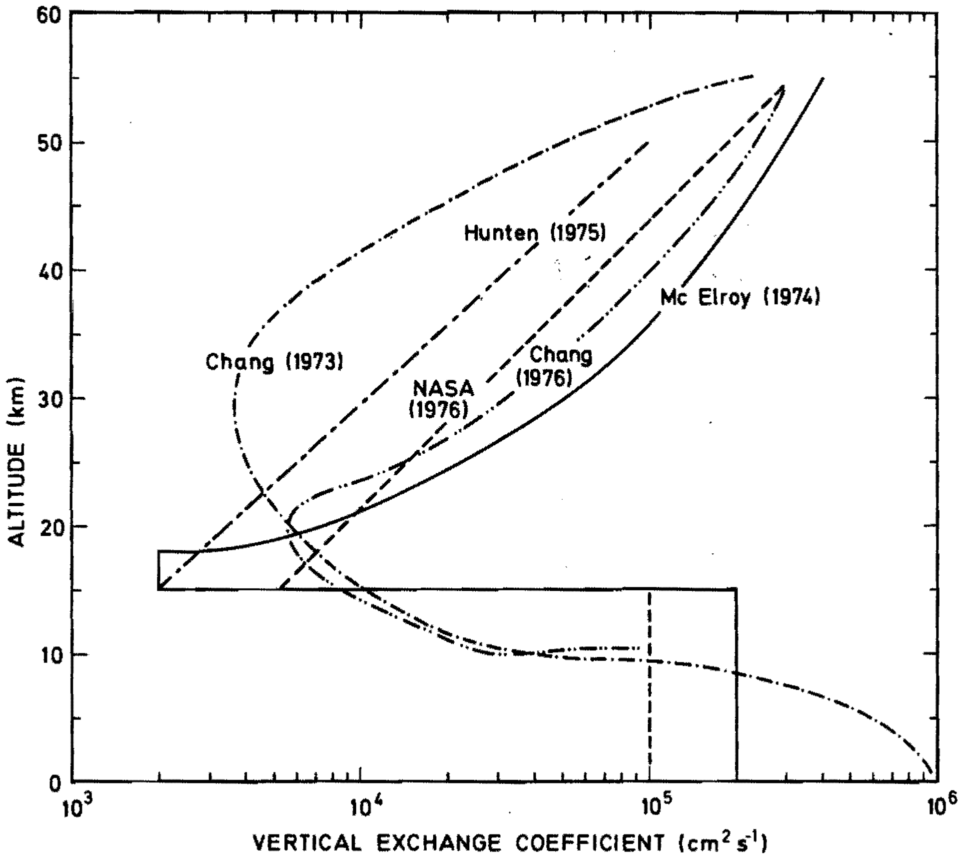

Fig. 24.- Exchange coefficients used in several stratospheric 1-D models.

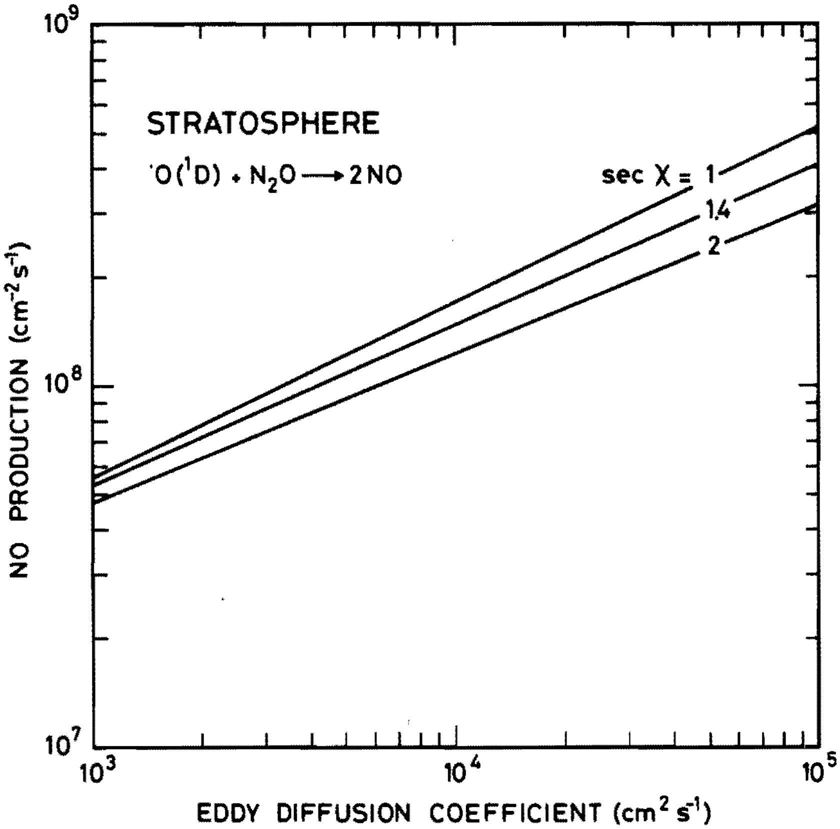

Fig. 25.- Integrated production rate of nitric oxide in the stratosphere as a function of the exchange coefficient K which is chosen constant with altitude. After Nicolet and Peetermans (1972).

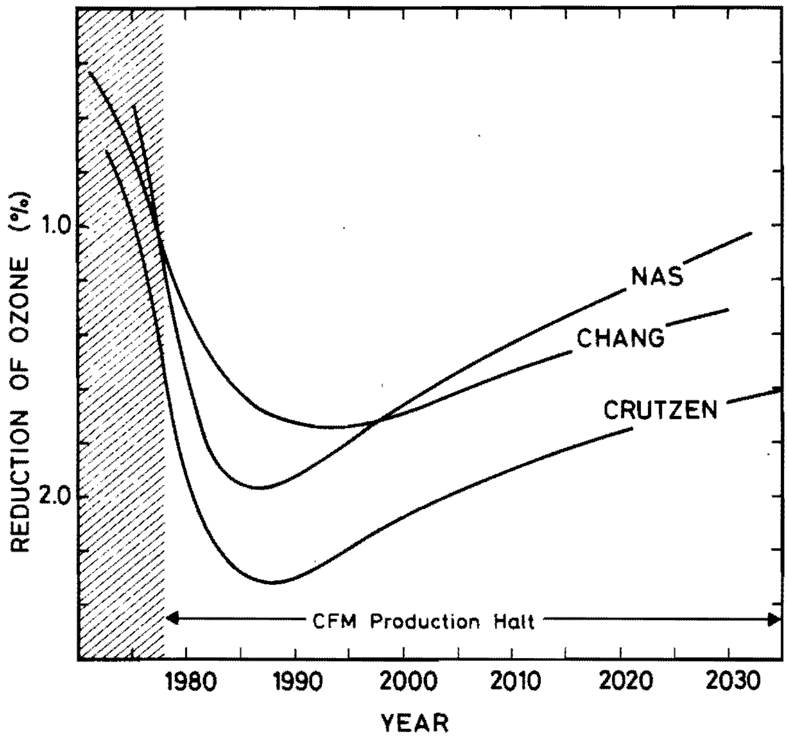

Fig. 26.- Behavior of total ozone when a constant release of chlorofluoromethanes in the atmosphere is completely stopped in 1978. Calculations with different exchange coefficients. After NAS (1976).

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