

A Zonal Mean Model of Stratospheric Tracer Transport in Isentropic Coordinates: Numerical Simulations for Nitrous Oxide and Nitric Acid

MALCOLM K. W. KO

Atmospheric and Environmental Research, Inc., Cambridge, Massachusetts

KA KIT TUNG

Department of Mathematics, Massachusetts Institute of Technology, Cambridge

DEBRA K. WEISENSTEIN AND NIEN DAK SZE

Atmospheric and Environmental Research, Inc., Cambridge, Massachusetts

The use of the linear wave treatment to describe the eddy motions in a zonal mean model leads to the identification of the irreversible transient waves as the process responsible for the diffusive transport of tracers in the stratosphere. Recent estimates based on an idealized general circulation model (GCM) calculation and the observed transient wave statistics between 20°N and 80°N suggest that, in the stratosphere, the horizontal diffusion coefficient should be $\lesssim 3 \times 10^9 \text{ cm}^2 \text{ s}^{-1}$, a value about 1 order of magnitude smaller than values previously used in most two-dimensional models. A simple model of zonally averaged transport of stratospheric trace gases in isentropic coordinates is developed to test this hypothesis of small eddy diffusion. In this model, the eddy transport arising from the irreversible transient waves is assumed to act along the isentropic surfaces and is represented by a single horizontal diffusion coefficient, K_{yy} . The advective transport in the model is effected by the zonal mean diabatic circulation calculated diagnostically from a given zonal mean diabatic heating rate, of which no post hoc adjustment has been attempted. We show that the observed stratospheric distribution of N_2O and HNO_3 may be adequately simulated in the model with small values of K_{yy} that are consistent with physical estimates mentioned above. The result that the observed concentrations of the stratospheric trace gases can be simulated using small values of eddy diffusion coefficients is expected to be equally applicable for models in pressure coordinates using the residual mean circulations.

1. INTRODUCTION

Recent theoretical developments over the past decade have gradually established the physical basis for the so-called two-dimensional zonal mean models in which the tracer transport equation can be derived from the three-dimensional continuity equation by averaging along the zonal (i.e., longitudinal) direction. This should be contrasted with one-dimensional models in which the transport equation cannot be constructed by averaging the three-dimensional equation over the two horizontal dimensions.

A major problem in the formulation of a two-dimensional model lies in the treatment and the quantification of the so-called eddy flux terms, which must be modelled by additional physical considerations concerning the nature of the motions in the three-dimensional atmosphere. To define the terms that will be used in this discussion, let us first write down the three-dimensional form of the equation governing the transport of a species whose volume mixing ratio is f . This is

$$\frac{D}{Dt} f = P \quad (1)$$

where P is the local chemical production/loss rate of the species in mixing ratio units and D/Dt is the substantial derivative given by

$$\frac{D}{Dt} \equiv \frac{\partial}{\partial t} + \mathbf{u} \cdot \nabla \quad (2)$$

Equation (1) expresses the local time rate of change of $f(\partial f/\partial t)$ in terms of the local rate of photochemical production (P) and the advection of $f(\mathbf{u} \cdot \nabla f)$ by the three-dimensional motion field of the air (\mathbf{u}).

When (1) is averaged zonally, with the averaging operator denoted by angle brackets, it becomes

$$\frac{\partial}{\partial t} \langle f \rangle + \langle \mathbf{u} \rangle \cdot \nabla \langle f \rangle + \langle \mathbf{u}' \cdot \nabla f' \rangle = \langle P \rangle \quad (3)$$

The primed quantities are the deviations from the averages, i.e., $(\)' \equiv (\) - \langle (\) \rangle$, and are referred to as eddies or waves. Expressing ∇ and \mathbf{u} in component form,

$$\nabla = \left(\frac{\partial}{\partial x}, \frac{\partial}{\partial y}, \frac{\partial}{\partial z} \right)$$

and

$$\mathbf{u} = (u, v, w)$$

where z is the vertical coordinate, x and y are the horizontal coordinates in the zonal and meridional directions, and (u, v, w) are the velocities in the corresponding (x, y, z) directions, (3) can be rewritten as

$$\begin{aligned} \frac{\partial}{\partial t} \langle f \rangle + \langle v \rangle \frac{\partial}{\partial y} \langle f \rangle + \langle w \rangle \frac{\partial}{\partial z} \langle f \rangle + \frac{\partial}{\partial y} \langle v f' \rangle \\ + \frac{\partial}{\partial z} \langle w f' \rangle = \langle P \rangle \end{aligned} \quad (4)$$

(In arriving at (4), we have assumed for the purpose of the discussion that the velocity field in the chosen coordinates

system is nondivergent, i.e., $\nabla \cdot \mathbf{u} = 0$.) Equation (4) partitions the transport process into two parts which are referred to as mean and eddy transports. The former refers to the advective transport of $\langle f \rangle$ by the zonally averaged air velocities ($\langle v \rangle$, $\langle w \rangle$) in the latitude-height plane, whereas the latter arises from the eddy fluxes of $\langle f \rangle$ in the y and the z directions, ($\langle v'f' \rangle$, $\langle w'f' \rangle$).

As was first demonstrated by *Mahlman* [1966], the partitioning of the transport process into the eddy and mean components is not unique, since it depends on how the average is taken. For instance, an average taken along the jet stream maximum is different from the zonal average along a coordinate axis. Similarly, the zonal average of a three-dimensional field taken on isentropic surfaces could be very different from the zonal average of the same quantity performed on isobaric surfaces. This is the case for some of the dynamical terms entering into the transport equation (3). The nonuniqueness in the separation of the transport processes opens the possibility that the role played by the eddies in the two-dimensional transport of tracers may perhaps be minimized by a judicious choice of the averaging procedure. In the case of the generalized Lagrangian mean formulation [*Andrews and McIntyre*, 1978], in which the zonal averaging is performed along the eddy-displaced path, the eddy flux transport is found to be identically zero. However, due to the fact that this averaging procedure cannot be performed without a knowledge of the eddy field, which is not calculated within a two-dimensional theory, it is by now recognized that the application of the theory to the tracer transport problem would face insurmountable difficulties [*McIntyre*, 1980; *Sze et al.*, 1981].

Since the eddies which make up the eddy flux transport are defined by the deviations of the three-dimensional fields from the corresponding two-dimensional mean, they cannot be calculated from within the two-dimensional model. One current approach to resolving this difficulty is to calculate the eddy fluxes using linear perturbation wave theory [*Clark and Rogers*, 1978; *Plumb*, 1979; *Matsumo*, 1980; *Pyle and Rogers*, 1980a; *Holton*, 1980, 1981; *Tung*, 1982]. By subtracting (4) from (1), an equation for f' can be obtained if one ignores, first, the quadratic terms in the perturbation (primed) quantities and, second, the meridional and vertical mean advection of f' , which is much smaller than the zonal mean advection. The equation for f' is given by

$$\left(\frac{\partial}{\partial t} + \langle u \rangle \frac{\partial}{\partial x} \right) f' + v' \frac{\partial}{\partial y} \langle f \rangle + w' \frac{\partial}{\partial z} \langle f \rangle = P' \quad (5)$$

With the assumption that $\partial \langle f \rangle / \partial t \lesssim \partial f' / \partial t$ (i.e., eddy fluctuation time scale much shorter than the time scale of the variation for the mean quantities), (5) can be solved (see above references) to give

$$f' = -\eta' \frac{\partial}{\partial y} \langle f \rangle - \zeta' \frac{\partial}{\partial z} \langle f \rangle + \sigma' \quad (6)$$

In (6), η' and ζ' are the eddy displacements of the air motions in the y and z directions satisfying

$$\begin{aligned} \left(\frac{\partial}{\partial t} + \langle u \rangle \frac{\partial}{\partial x} \right) \eta' &= v' \\ \left(\frac{\partial}{\partial t} + \langle u \rangle \frac{\partial}{\partial x} \right) \zeta' &= w' \end{aligned} \quad (7)$$

The quantity σ' arises from the eddy chemical term P' , with

$$\left(\frac{\partial}{\partial t} + \langle u \rangle \frac{\partial}{\partial x} \right) \sigma' = P' \quad (8)$$

Using (6), the eddy fluxes take the form

$$\begin{bmatrix} \langle v'f' \rangle \\ \langle w'f' \rangle \end{bmatrix} = - \begin{bmatrix} K_{yy} & K_{yz} \\ K_{zy} & K_{zz} \end{bmatrix} \begin{bmatrix} \frac{\partial \langle f \rangle}{\partial y} \\ \frac{\partial \langle f \rangle}{\partial z} \end{bmatrix} + \begin{bmatrix} \langle v'\sigma' \rangle \\ \langle w'\sigma' \rangle \end{bmatrix} \quad (9)$$

where the elements of the eddy transport tensor may be expressed in terms of the product of the eddy displacements and eddy velocities:

$$\begin{aligned} K_{yy} &= \langle \eta'v' \rangle & K_{yz} &= \langle \zeta'v' \rangle \\ K_{zz} &= \langle \zeta'w' \rangle & K_{zy} &= \langle \eta'w' \rangle \end{aligned} \quad (10)$$

Note that there is no explicit dependence on $\langle f \rangle$ or f' in the definition of the elements of the eddy tensor; thus the same eddy tensor is applicable to all trace gases. The role of the chemical eddy terms ($\langle v'\sigma' \rangle$ and $\langle w'\sigma' \rangle$) for specific chemical species was first examined in detail by *Pyle and Rogers* [1980b]. In its linearized form, these terms will depend on the zonal mean concentrations of the trace gases (but not f') in addition to eddy displacements and eddy velocities. It may provide strong couplings between different chemical species in certain regions of the atmosphere [*Pyle and Rogers*, 1980b; *Garcia and Solomon*, 1983; *Rogers and Pyle*, 1984]. In this work, we will concentrate on the discussion of the eddy transport tensor arising from transient waves. However, the role of the chemical eddy may be important in the calculation of certain species.

Equations (5) through (10) provide a procedure for eliminating the unknown f' from the tracer transport equation. However, the eddy air displacement terms still remain (in the definition of the K 's and the chemical eddy terms) and need to be determined somehow. Different two-dimensional models can be classified according to their treatment of the eddy transport tensor and the manner in which the two-dimensional advective field ($\langle v \rangle$, $\langle w \rangle$) is obtained. The question of whether the formulation of a particular two-dimensional model has a physical basis can be addressed by considering if the K 's can be derived from a physically justifiable procedure. Furthermore, the model can be regarded as diagnostically useful if the magnitudes of the transport parameters can be constrained by rigorous dynamical principles.

In early two-dimensional models using pressure coordinates, the eddy transport tensor is parameterized by treating the deviations (v' , w' , and η' , ζ') as due to turbulent motions [*Reed and German*, 1965]. Instead of calculating the K 's from derived values for v' , w' , η' , and ζ' , the magnitudes of the K 's are inferred from observations of temperature and wind field statistics with the assumption that the heat fluxes ($\langle v'T' \rangle$ and $\langle w'T' \rangle$) are given by the product of the K tensor and the zonal mean temperature gradients [cf. *Reed and German*, 1965]. Models using these values of the K 's have shown that eddy transport is at least as important as advection in defining the transport of the stratospheric trace gases. In contrast, recent work attempts to derive expressions for v' , w' , η' , and ζ' by considering the eddy motions as arising from planetary and transient wave motions [*Clark and Rogers*, 1978; *Plumb*, 1979; *Matsumo*, 1980; *Holton*, 1981; *Tung*, 1982]. The linear wave treatment of the eddy displacements provides a physical basis

for separating the eddy tensor into its symmetric and antisymmetric parts. The antisymmetric part of the eddy tensor introduces the eddy-induced mean flow which can be combined with the mean circulation to give the diabatic circulation. The symmetric part of the eddy tensor gives the diffusive transport arising from transient wave motions. Although the diabatic circulation formulation has been adopted by several models [Pyle and Rogers, 1980b; Miller et al., 1981; Holton, 1981; Garcia and Solomon, 1983], the role of the diffusive transport from the transient waves in determining the distribution of stratospheric tracers remains unclear.

Preliminary estimates based on results from an idealized general circulation model calculation [Kida, 1983] and observed transient wave statistics [Tung, 1984] indicate that the horizontal diffusion coefficient has a value of $\lesssim 3 \times 10^9 \text{ cm}^2 \text{ s}^{-1}$, compared with values of $\sim 3 \times 10^{10} \text{ cm}^2 \text{ s}^{-1}$ derived for the high latitudes in the winter hemisphere by Reed and German [1965] and Luther [1973]. The recent work of Tung [1982, 1984] showed that the horizontal diffusion should dominate over vertical diffusion in isentropic coordinates. The purpose of this paper is to demonstrate that the stratospheric distributions of nitrous oxide (N_2O) and nitric acid (HNO_3) may be simulated by a model using the diabatic circulation with diffusion coefficients subject to the above constraints. Our results indicate that, in a properly formulated zonal mean model, the transport of tracer is primarily affected by advection driven by the diabatic circulation with diffusion playing the role of smoothing out the large gradients created by the diabatic circulation.

In section 2, we will review the different treatments of the eddy flux term. The methods for assessing the importance of eddy diffusion for the transport of stratospheric trace gases will be presented in section 3. The model developed for this study is described in sections 4 through 6. Results from numerical simulations of the zonal mean distribution of nitrous oxide and nitric acid are presented in sections 7 and 8.

2. REVIEW OF TREATMENTS OF THE EDDY FLUX

The two-dimensional eddy tensor (K) can be separated into its symmetric and antisymmetric part:

$$\begin{bmatrix} K_{yy} & K_{yz} \\ K_{zy} & K_{zz} \end{bmatrix} = \begin{bmatrix} K_{yy} & K_{yz}^{(s)} \\ K_{zy}^{(s)} & K_{zz} \end{bmatrix} + \begin{bmatrix} 0 & \Psi \\ -\Psi & 0 \end{bmatrix} \quad (11)$$

where $K_{yz}^{(s)} = K_{zy}^{(s)} \equiv (K_{yz} + K_{zy})/2$ and $\Psi \equiv (K_{yz} - K_{zy})/2$. Because a symmetric tensor can be diagonalized via a coordinate transformation and a diagonalized eddy transport tensor gives rise to Fickian diffusion, the symmetric part of the eddy transport process represents diffusion (or mixing) along sloped paths. The term arising from the antisymmetric part Ψ can be written as

$$\begin{aligned} \frac{\partial}{\partial y} \left(\Psi \frac{\partial \langle f \rangle}{\partial z} \right) + \frac{\partial}{\partial z} \left(-\Psi \frac{\partial \langle f \rangle}{\partial y} \right) \\ = \frac{\partial \Psi}{\partial y} \frac{\partial \langle f \rangle}{\partial z} - \frac{\partial \Psi}{\partial z} \frac{\partial \langle f \rangle}{\partial y} \end{aligned} \quad (12)$$

where Ψ can be identified as a stream function for the advective velocity ($-\partial\Psi/\partial z, \partial\Psi/\partial y$) giving rise to the eddy-induced advective transport of $\langle f \rangle$. We will now review the different treatments of the eddy flux term where specific physical processes are invoked to provide a physical interpretation for the symmetric and antisymmetric part of the eddy tensor.

It has long been recognized that the observed transport of

tracers cannot be explained solely in terms of advection by the observed zonal mean circulation defined on isobaric surfaces. The so-called Brewer-Dobson circulation [Brewer, 1949; Dobson, 1956], which is deduced from the observed directions of tracer transport in the latitude-height plane, has a two-cell global structure in the lower stratosphere (below $\sim 25 \text{ km}$) with rising motion in the tropics and sinking motion near both poles. In the upper stratosphere and mesosphere (between 25 km and 50 km), the Brewer-Dobson circulation has only one global cell, rising from the summer pole and subsiding in the winter pole. The observed zonal mean circulation when viewed in pressure coordinates ($\langle v \rangle, \langle w \rangle_p$) is quite different, being opposite to the direction of tracer transport in some regions of the lower stratosphere during winter [Reed et al., 1963; Newell, 1963]. Thus eddy flux transports were thought to play a significant role in determining the direction of tracer transport in these regions if $(\langle v \rangle, \langle w \rangle_p)$ is used to provide the advective portion of the transport.

The inability of $(\langle v \rangle, \langle w \rangle_p)$ to simulate tracer transport is recognized by Reed and German [1965] in their classical formulation of the two-dimensional tracer transport models. The effect of eddy transport from large-scale eddies in the atmosphere is included to complement the advective transport from $(\langle v \rangle, \langle w \rangle_p)$. The eddies, including those arising from planetary waves, are however assumed to be turbulent in nature. Instead of using (7), the eddy flux is derived using the mixing length hypothesis where the eddy displacements η' and ζ' entering into the K 's are defined as

$$\frac{\eta'}{\Delta t} = v' \quad \frac{\zeta'}{\Delta t} = w' \quad (13)$$

As a consequence of (13), the K tensor becomes symmetric, since

$$K_{yz} \equiv \langle \zeta' v' \rangle = \frac{1}{\Delta t} \langle \zeta' \eta' \rangle = \langle \eta' w' \rangle = K_{zy} = K_{yz}^{(s)} \quad (14)$$

The magnitudes of the elements of the symmetric K tensor are inferred [Reed and German, 1965; Luther, 1973] from temperature data and wind field statistics with the assumption that the eddy heat fluxes ($\langle v'T' \rangle, \langle w'T' \rangle$) are also entirely diffusive and are given by the product of the symmetric K tensor and the gradients of the zonal mean temperature. In the original study of Reed and German [1963], the two-dimensional model was formulated to study the dispersion of nuclear debris from bomb tests. Since the gradient of the debris is quite large, diffusion may play a more important role than advection in transporting the debris. In such cases, the transport of the tracer is given by diffusion along the mixing paths determined from the relative magnitudes of the elements of the K tensor. These paths are found to slope poleward and downward in the lower stratosphere in a manner similar to the observed surfaces of constant mixing ratio for several trace gases.

In later modeling studies, values of the eddy diffusion coefficients derived in similar ways are used to provide diffusive transport to complement advective transport in simulating the distributions of stratospheric trace gases. With the assigned values of the diffusion coefficients, the calculated mixing ratio surfaces of the trace gases are found to follow the mixing paths. Partly because the mixing paths are, to some extent, correctly fixed by the parameterization of the K 's, transport models adopting the values from Luther [1973] and Reed and German [1965] appear to give reasonable results for tracer concentration when compared to available observational data

[Harwood and Pyle, 1977; Vupputuri, 1978; Pyle, 1980; Ko et al., 1984]. Despite these reasonable agreements, the physical basis of such models is still lacking [Harwood, 1980]. In particular, the adoption of the mixing length hypothesis (equation (13)) which results in the vanishing of the antisymmetric part of the eddy tensor appears to be in conflict with the recent development [Clark and Rogers, 1978] which emphasizes the importance of planetary wave motions in tracer transport.

As indicated in (11), the eddy transport tensor need not be symmetric. If one follows the linear perturbation procedure consistently, the displacement field should instead be defined via (7), which is different from the definition (13) adopted in the mixing length theory. If (7) is used, one can now show that there is an antisymmetric component due to stationary planetary waves. In fact, for a steady wave field, ($\partial\eta'/\partial t = \partial\zeta'/\partial t = 0$), an integration by parts would show

$$K_{yz} = \left\langle \zeta' \langle u \rangle \frac{\partial}{\partial x} \eta' \right\rangle = - \left\langle \eta' \langle u \rangle \frac{\partial}{\partial x} \zeta' \right\rangle = -K_{zy} = \Psi \quad (15)$$

The net advective transport in two-dimensional models should then be obtained by combining the advection due to the observed mean circulation ($\langle v \rangle$, $\langle w \rangle$) and the advective transport from the eddy-induced circulation ($-\partial\Psi/\partial z$, $\partial\Psi/\partial y$). The function Ψ is related to the heat flux and can be expressed in terms of the potential temperature θ as $\langle \theta'v' \rangle / (\partial\langle \theta \rangle / \partial z)$ in pressure coordinates [Holton, 1981].

It has been found that in the lower stratosphere the two types of advection are often opposite in direction and the net advection is a small residue from the cancellation [Hunt and Manabe, 1968; Mahlman and Moxim, 1978; Dunkerton et al., 1981]. A physical explanation for this near cancellation can be found in the so-called "noninteraction theorems" [see Andrews and McIntyre, 1976, 1978; Boyd, 1976; Holton, 1980]. The noninteraction theorems state that if the atmosphere is adiabatic (no diabatic heating), and the eddies are conservative, steady, and of small amplitude, then the mean circulation ($\langle v \rangle$, $\langle w \rangle$) exactly cancels the induced advection from the eddy fluxes. When there is diabatic heating, exact cancellation no longer occurs, and there is now a small residual transport driven directly by the mean diabatic heating. The diabatic heating is essentially given by the radiative heating in the stratosphere although the latent heat release in cumulus clouds should be included for the troposphere. The portion of the circulation driven by diabatic heating should have mean upward motion in the region of heating and mean subsidence in the region of cooling. It is this residual (diabatic) circulation that Dunkerton [1978] found to have qualitative features similar to the Brewer-Dobson circulation deduced from tracer distributions.

The new generation of two-dimensional models formulated in Eulerian coordinates attempts to take into account a priori the near cancellation of the two types of advective transports to arrive at the diabatic circulation directly. There are two ways of accomplishing this objective. In the "transformed Eulerian mean" models [Andrews and McIntyre, 1976; Boyd, 1976; Dunkerton, 1978; Matsuno, 1980; Pyle and Rogers, 1980a; Holton, 1981], the observed mean circulation in pressure coordinates ($\langle v \rangle$, $\langle w \rangle_p$) is combined with the eddy-induced mean flow ($-\partial\Psi/\partial z$, $\partial\Psi/\partial y$) to obtain a new "residual mean circulation," which reduces to the diabatic circulation responsible for tracer transport provided that the eddy field is steady, adiabatic, and of small amplitude. A second approach is to

formulate the two-dimensional model in isentropic coordinates [Tung, 1982; Mahlman et al., 1984]. In isentropic coordinates, the vertical "velocity," i.e., the time rate of change of the vertical coordinate (in this case the potential temperature) is directly related to the diabatic heating through the thermodynamic equation. Because of this, the observed mean circulation is the diabatic circulation if the zonal average is taken on isentropic (instead of isobaric) surfaces. No "residual circulation" needs to be defined. Similar to the situation for the transformed Eulerian mean models, the diabatic circulation is found to be the circulation for the net advective transport of tracers, provided the eddies are adiabatic and of small amplitude.

The second-generation models take care of the bulk of the antisymmetric part of the eddy transport tensor by absorbing it into the mean advective part. When the eddy field is not steady, the elements of the symmetric part of the eddy transport tensor become nonzero. One can easily demonstrate from (10) that the symmetric K tensor can be expressed in terms of time derivatives of the eddy displacement fields [see Holton, 1981; Tung, 1982]. For example,

$$\begin{aligned} K_{yy} &= \left\langle \eta' \left(\frac{\partial}{\partial t} + \langle u \rangle \frac{\partial}{\partial x} \right) \eta' \right\rangle = \left\langle \eta' \frac{\partial}{\partial t} \eta' \right\rangle \\ &= \frac{\partial}{\partial t} \left\langle \frac{1}{2} \eta'^2 \right\rangle \\ K_{zz} &= \frac{\partial}{\partial t} \left\langle \frac{1}{2} \zeta'^2 \right\rangle \end{aligned} \quad (16)$$

$$\begin{aligned} \frac{1}{2}(K_{yz} + K_{zy}) &= \frac{1}{2} \left(\left\langle \zeta' \frac{\partial}{\partial t} \eta' \right\rangle + \left\langle \eta' \frac{\partial}{\partial t} \zeta' \right\rangle \right) \\ &= \frac{\partial}{\partial t} \left\langle \frac{1}{2} \eta' \zeta' \right\rangle = K_{yz}^{(s)} \end{aligned}$$

All these components vanish when the wave field is steady or periodic. It is the irreversible transient processes that give rise to the symmetric K 's, which lead to diffusive transport.

3. THE ROLE OF EDDY DIFFUSION IN ZONAL MEAN MODELS OF THE STRATOSPHERE

The values of the diffusion coefficients used in classical two-dimensional models [Reed and German, 1965; Luther, 1973] are quite large in magnitude. The adopted values of these coefficients were inferred from temperature data based on the assumption that the eddy transport of heat is related to the same tensor without the antisymmetric components, which we now find to be important. The validity of the previous procedure of deducing the diffusion coefficients appears questionable. Consequently, there is considerable uncertainty concerning the magnitude of the diffusion coefficients that one should adopt in the new generation of two-dimensional models. It is obviously desirable to constrain the choice of eddy diffusion coefficients by some objective means.

There have been a number of numerical implementations of the second-generation formulation of two-dimensional models in which a diabatic circulation is adopted [Pyle and Rogers, 1980a; Holton, 1981; Miller et al., 1981; Garcia and Solomon, 1983; Rogers and Pyle, 1984; Guthrie et al., 1984]. These models, however, differ from each other in their choice of eddy diffusion coefficients. The work of Pyle and Rogers [1980a, b] represents the first attempt to assess the importance of the eddy transport in their model study of ozone. In the work of Pyle and Rogers [1980a], a diabatic circulation is first ob-

tained by excluding the eddy terms in calculating the wind fields. The results showed that, in the absence of explicit eddy terms in the trace gas equation, their diabatic circulation failed to reproduce the observed latitudinal and seasonal distribution of the column density of O₃. In contrast, a much better result is obtained if the diabatic circulation is augmented by the eddy fluxes parameterized by Luther's [1973] values for the eddy diffusion coefficients. They concluded that a large eddy transport is needed to augment the poleward transport of O₃ and that the eddy terms should be interpreted as an approximation for the chemical eddies ($\langle v'\sigma' \rangle$, $\langle w'\sigma' \rangle$) in their calculation. However, the result is somewhat inconclusive, since the numerical experiment for the case without eddy diffusion is not run to steady state because of the occurrence of negative mixing ratio. In a more recent paper [Rogers and Pyle, 1984], the chemical eddy portion of the eddy flux for O₃ is calculated to have equivalent K_{yy} values ranging from 10⁹ to 10¹⁰ cm² s⁻¹. With this smaller value of the eddy diffusion coefficient, a satisfactory result for O₃ is obtained if a subgrid diffusion term is included in the trace gas equation. We shall show later that, for the case of N₂O and HNO₃, satisfactory results with a small eddy diffusion can be obtained without introduction of a subgrid mixing when numerical instability is suppressed by a new differencing scheme.

There is as yet no comprehensive study to determine the magnitude and distribution of the diffusion coefficients using transient eddy statistics and (16). Nonetheless, some order of magnitude estimates are available. In a tracer experiment using a simplified three-dimensional general circulation model, Kida [1983] deduced that the horizontal component of the diffusion coefficient, K_{yy} , should have a maximum value of about 3 × 10⁹ cm² s⁻¹ in the stratosphere, while the vertical component, K_{zz} , is found to be at most 1 × 10³ cm² s⁻¹, in pressure coordinates. Based on the observed magnitude of transient large-scale eddies in the atmosphere between 20°N and 80°N [Lau and Oort, 1982], the magnitude of K_{yy} in the stratosphere has been estimated by Tung [1984] to be less than 4 × 10⁹ cm² s⁻¹, in either pressure or isentropic coordinates. These estimates for K_{yy} are about an order of magnitude smaller than values deduced for the high latitudes in the winter hemisphere by Reed and German [1965] and Luther [1973]. Tung [1984] also gave estimates for the ratio of the various diffusion fluxes in isentropic coordinates. From those estimates, one can deduce values of $K_{yz}^{(s)}$ and K_{zz} in isentropic coordinates to be ≤ 10⁶ cm² s⁻¹ and ≤ 10² cm² s⁻¹, respectively.

The magnitude of the eddy flux term for a trace gas depends on the magnitudes of the K 's as well as the gradient of $\langle f \rangle$ for the trace gas. The relative magnitudes of the contributions from advection and diffusion in either the horizontal or vertical direction for the transport of a particular trace gas with volume mixing ratio $\langle f_i \rangle$ can be roughly estimated by examining the following ratios:

$$R_1^i \equiv \frac{(\partial/\partial y)(\langle v \rangle \langle f_i \rangle)}{(\partial/\partial y)[K_{yy} (\partial \langle f_i \rangle / \partial y)]} \sim \frac{\langle v \rangle L_i}{K_{yy}} \quad (17a)$$

$$R_2^i \equiv \frac{(\partial/\partial y)(\langle v \rangle \langle f_i \rangle)}{(\partial/\partial y)[K_{yz} (\partial \langle f_i \rangle / \partial z)]} \sim \frac{\langle v \rangle H_i}{K_{yz}} \quad (17b)$$

$$R_3^i \equiv \frac{(\partial/\partial z)(\langle w \rangle \langle f_i \rangle)}{(\partial/\partial z)[K_{zz} (\partial \langle f_i \rangle / \partial z)]} \sim \frac{\langle w \rangle H_i}{K_{zz}} \quad (17c)$$

$$R_4^i \equiv \frac{(\partial/\partial z)(\langle w \rangle \langle f_i \rangle)}{(\partial/\partial z)[K_{yz} (\partial \langle f_i \rangle / \partial y)]} \sim \frac{\langle w \rangle L_i}{K_{yz}} \quad (17d)$$

where L_i and H_i are the horizontal and vertical length scales of $\langle f_i \rangle$ (the distance over which $\Delta \langle f_i \rangle$ is comparable to $\langle f_i \rangle$) and are species dependent. The ratios R_1^i and R_2^i (R_3^i and R_4^i) may be taken as crude measures of the dominance of advection in the horizontal (vertical) direction. Diffusion is important if the ratios are comparable to or smaller than unity. Using the estimates of Tung [1984] for the K 's and values of 0.25 m/s for $\langle v \rangle$ and 0.5 mm/s for $\langle w \rangle$, we obtain the following estimates of the R 's for the model in the isentropic coordinate system

$$R_1^i \approx \frac{L_i}{2000 \text{ km}}$$

$$R_2^i \approx \frac{H_i}{0.5 \text{ km}}$$

$$R_3^i \approx \frac{H_i}{2 \times 10^{-2} \text{ km}}$$

$$R_4^i \approx \frac{L_i}{200 \text{ km}}$$

The above expressions suggest that in isentropic coordinates the horizontal diffusion would be comparable to the horizontal advection if $L_i \approx 2 \times 10^3$ km or $H_i \approx 0.5$ km. The condition for the dominance of the vertical diffusion is more stringent requiring that $L_i \approx 200$ km or $H_i \approx 2 \times 10^{-2}$ km.

In classical two-dimensional models where the magnitudes for K_{yy} , K_{yz} , and K_{zz} are as large as 4 × 10¹⁰ cm² s⁻¹, 10⁷ cm² s⁻¹, and 10⁴ cm² s⁻¹, respectively, the corresponding values for the ratios are

$$R_1^i \approx \frac{L_i}{2 \times 10^4 \text{ km}}$$

$$R_2^i \approx \frac{H_i}{5 \text{ km}}$$

$$R_3^i \approx \frac{H_i}{1 \text{ km}}$$

$$R_4^i \approx \frac{L_i}{2 \times 10^3 \text{ km}}$$

Thus the effect of diffusion is comparable to or larger than that of advection for a much larger range of values for L_i and H_i length scales. This finding is consistent with the fact that the calculated mixing ratio surfaces of the trace gases in model simulations using the large values of eddy diffusion coefficients [cf. Harwood and Pyle, 1977; Vupputuri, 1978; Pyle, 1980; Ko et al., 1984] follow the mixing surfaces defined by the eddy diffusion coefficients.

In view of the inherent uncertainty in the statistics of both the observed and general circulation model generated data for the transient waves, it is perhaps instructive to examine the stratospheric tracer distributions calculated with different values of diffusion coefficients. Hopefully, by comparing the model results with observational data, additional insights may be gained on the magnitude of the diffusion coefficients. There are in general three diffusion coefficients, K_{yy} , K_{zz} , and $K_{yz}^{(s)}$, that should be considered. In pressure coordinates, they are of comparable importance. If our proposed study of model sensitivity to diffusion were performed in pressure coordinates, a combination of the three would have to be varied, a very time-consuming task. Fortunately, it is found [Tung, 1984] that the isentropic coordinate system behaves like a principal

axis system for the eddy diffusion tensor. Not only is such a tensor approximately "diagonalized" in isentropic coordinates, the horizontal diffusion term K_{yy} is the only element of significance in the stratosphere. Therefore if the isentropic coordinate system is adopted, eddy diffusion is represented mainly by mixing along isentropic surfaces, and only a single parameter, K_{yy} , need be varied in a series of model calculations to determine the proper choice of eddy diffusion coefficients. It is for this reason that the isentropic coordinate formulation is adopted here. Our conclusion concerning the role of eddy diffusion in determining the transport of stratospheric trace gases should be applicable to the transformed Eulerian mean circulation formulation in pressure coordinates as well.

In the troposphere, eddy mixing is expected to play a more important role. Vertical mixing due to convective processes is known to be important in the troposphere but has not been incorporated in the above estimates, which are intended to be for the stratosphere only. Thus a model omitting the mixing process may not be extended to the troposphere in a straightforward manner. However, such a model can still be applied to species such as nitrous oxide (N_2O) and total odd nitrogen (NOY) which are not sensitive to the detailed specification of the eddy diffusion coefficients in the troposphere.

4. MODEL IN ISENTROPIC COORDINATES

The detailed description of the formulation of a two-dimensional model in isentropic coordinates can be found in the work of *Tung* [1982]. The key features of the formulation are summarized in the appendix, along with an approximate procedure for converting data in pressure coordinates into isentropic coordinates. Here we will recast that formulation into a pressurelike coordinate for easier interpretation.

The potential temperature is defined by

$$\theta \equiv T \left(\frac{p_{00}}{p} \right)^\kappa \quad (18)$$

where T is the temperature, p is the pressure, p_{00} is 1000 mbar, and κ is R/c_p with $R = c_p/c_v$, where c_p and c_v are the specific heat at constant pressure and volume, respectively. We define a new pressurelike function in the isentropic coordinate system by

$$p_e(\theta) \equiv p_{00} \left(\frac{\tilde{T}_e}{\theta} \right)^{1/\kappa} \quad (19)$$

where p_e is the equilibrium pressure [Tung, 1982]. It is a function of θ only and can thus be used instead of θ as the vertical coordinate. In (19), \tilde{T}_e is the latitudinally averaged radiative equilibrium temperature. As far as the definition (19) is concerned, \tilde{T}_e can be any function of θ with the dimension of temperature. However, our choice of \tilde{T}_e enables p_e to be within 20% of the actual pressure p .

The hydrostatic equation in isentropic coordinates is

$$\frac{\partial p}{\partial \theta} = -g\rho_\theta \quad (20)$$

where g is acceleration due to gravity, $\rho_\theta = \rho(\partial z/\partial \theta)$ is mass per unit coordinate volume, or "density," in isentropic coordinates. Evaluating (20) under radiative equilibrium condition and averaging latitudinally (or globally averaging (20)), one obtains

$$\frac{dp_e}{d\theta} = -g\rho_\theta^{(0)} \quad (21)$$

where $\rho_\theta^{(0)} \equiv (\tilde{\rho}_\theta)_e$ is the basic state density profile and is a function of θ only. Equation (21) provides the Jacobian for the transformation from θ to p_e coordinates.

We define the vertical velocity, ω_e , in the p_e coordinate as

$$\omega_e \equiv \frac{d}{dt} p_e = -g\rho_\theta^{(0)} \dot{\theta} \quad (22)$$

Thus

$$\langle \omega_e \rangle = -g\rho_\theta^{(0)} \langle \dot{\theta} \rangle \quad (23)$$

where the angle brackets denote zonal averaging keeping the latitude and θ fixed.

In isentropic coordinates, the energy equation can be written in the form

$$\dot{\theta} = \theta Q/T \quad (24)$$

where Q is the diabatic heating rate per unit mass divided by c_p . The zonal mean of (24) is, approximately

$$\langle \dot{\theta} \rangle \simeq \theta \frac{\langle Q \rangle}{\langle T \rangle} \simeq \theta \langle Q \rangle / \tilde{T}_e \quad (25)$$

assuming that $|(T'/\langle T \rangle) \cdot (Q'/\langle Q \rangle)|$ and $(\langle T \rangle - \tilde{T}_e)/\tilde{T}_e$ are small.

Combining (23), (25), and the definition of the static stability parameter

$$\Gamma^{(0)} \equiv \frac{\tilde{T}_e}{\theta_e} \left(\frac{\partial \theta}{\partial z} \right)_e$$

we obtain

$$\langle \omega_e \rangle \simeq -g\rho_\theta^{(0)} \langle Q \rangle / \Gamma^{(0)} \quad (26)$$

The form (26) is preferred here for practical purposes to the formulae for the mass flow rate $\langle W \rangle$ used by *Tung* [1982]. The calculation for the latter requires the knowledge of $\langle q \rangle$, the diabatic heating rate per unit volume, which is not readily available in the literature. In the present diagnostic study, $\Gamma^{(0)}$ is replaced by $\langle \Gamma \rangle = (\langle T \rangle / \theta) (\partial \langle \theta \rangle / \partial z)$ and $\rho_\theta^{(0)}$ by $\langle \rho \rangle$.

To calculate the zonal mean horizontal velocity $\langle v \rangle$, we use the following approximate form of the continuity equation:

$$\frac{\partial}{\partial y} (\langle v \rangle \cos \phi) + \frac{\partial}{\partial p_e} \langle \omega_e \rangle \simeq 0 \quad (27)$$

where $y \equiv a \sin \phi$ with a being the radius of the earth and ϕ the latitude.

(In the work of *Tung* [1982], a slightly different form of the continuity equation

$$\frac{\partial}{\partial y} \langle \rho_\theta v \rangle + \frac{\partial}{\partial \theta} \langle \rho_\theta \dot{\theta} \rangle \simeq 0 \quad (28)$$

was shown to be a valid approximation. To go from (28) to (27) involves the additional approximation of replacing ρ_θ in (28) by the basic state density profile $\rho_\theta^{(0)}$. The rationale for such a further approximation is discussed in the appendix.)

The species transport equation used in these studies is

$$\begin{aligned} \frac{\partial \langle f \rangle}{\partial t} = & -\langle v \rangle \cos \phi \frac{\partial \langle f \rangle}{\partial y} - \langle \omega_e \rangle \frac{\partial \langle f \rangle}{\partial p_e} \\ & + \frac{\partial}{\partial y} \left(K_{yy} \cos^2 \phi \frac{\partial \langle f \rangle}{\partial y} \right) + \langle P \rangle \end{aligned} \quad (29)$$

Equation (29) describes the transport of the zonal mean

volume mixing ratio $\langle f \rangle$ by, first, the advection of the nondivergent mean diabatic circulation ($\langle v \rangle$, $\langle \omega_e \rangle$) and, second, the horizontal diffusion by transient horizontal eddy displacements, $K_{yy} = (\partial/\partial t) \frac{1}{2} \langle \eta' \eta' \rangle$, where η' is the horizontal eddy displacement to be determined from the small-amplitude perturbation equation $[(\partial/\partial t) + \langle u \rangle (\partial/\partial x)] \eta' = v'$.

The diffusion terms K_{yz} and K_{zz} which arise from the vertical transient eddy displacements are neglected in (29). Tung [1984] has presented some observational evidence for the stratosphere showing that the vertical eddy displacement terms in the isentropic coordinate system are small. Consequently, the contributions from the K_{yz} and K_{zz} terms are 1 to 2 orders of magnitude smaller than the horizontal diffusion term retained in (29). On the other hand, the neglect of eddy advective transport in (29) is less well justified. The eddy diabatic advective transport arises when the perturbation diabatic heating rate (Q') is nonzero [cf. Tung, 1982]. It can be neglected in comparison with the mean advection ($\langle v \rangle$, $\langle \omega_e \rangle$) if Q' is smaller than the mean heating $\langle Q \rangle$. However, there is some evidence that Q' is about 1 to 2°C/day in the stratosphere, about 2 to 4 times larger than $\langle Q \rangle$ in the same region. If this is true, the eddy diabatic advective transport should be comparable in magnitude to the mean diabatic transport. Rood and Schoeberl [1983] estimated a value for the eddy advective transport of about 30% of that due to the mean diabatic circulation. Nevertheless, it seems that there is no need at present to explicitly incorporate the eddy advection terms because, first, they can be implicitly included in ($\langle v \rangle$, $\langle \omega_e \rangle$) by redefining the diabatic heating rate $\langle Q \rangle$ to be $\langle Q \rangle^* \equiv \langle Q \rangle + (\partial/\partial y) \langle \eta' Q' \rangle$ [see Tung, 1984] and, second, with the large uncertainty in $\langle Q \rangle$ in the present diagnostic study we cannot distinguish $\langle Q \rangle$ from $\langle Q \rangle^*$.

Finally, in the present calculation, only the mean photochemical source term is included in the term $\langle P \rangle$. The eddy source terms include terms arising from the covariance of the deviations of the species' concentrations from their zonal mean values [Tuck, 1979] and the chemical eddy terms $\langle v' \sigma' \rangle$ and $\langle w' \sigma' \rangle$ [Pyle and Rogers, 1980b; Tung, 1982; Garcia and Solomon, 1983; Rogers and Pyle, 1984]. In the present study, both forms of eddy source terms are excluded from the transport equation.

5. NUMERICAL METHOD

The present model uses the latitude (ϕ) and the logarithm of the equilibrium pressure ($\zeta_e = \log(p_0/p_e)$, $p_0 = 1000$ mbar) as coordinates. The two-dimensional space (latitude/altitude) is covered by a 19×17 grid. There are 19 latitudinal belts covering the globe from pole to pole, each $\sim 10^\circ$ (10^3 km) wide. The vertical levels are constant in ζ_e with $\Delta\zeta_e = 0.5$ (~ 3.5 km). The lower boundary, defined by $p_e = 1000$ mbar, corresponds to a geometrical altitude of 0–2 km depending on the atmospheric temperature. The upper boundary is at about 55 km.

One of the purposes of this study is to assess the role of the (horizontal) diffusion due to transient (horizontal) displacements in determining trace gas distributions. Our experience showed that if K_{yy} is set equal to zero in (29), the solution of the equation depends rather critically on the numerical method used. Since (29) is a partial differential equation with nonconstant coefficients, aliasing instability can arise in the solution [cf. Roache, 1982]. This problem is very severe if centered differencing is used for the equation without any explicit damping term in the form of eddy diffusion coefficients

or higher-order subgrid scale mixing. The upwind differencing scheme alleviates this problem at the expense of introducing a large implicit numerical diffusion [cf. Molenkamp, 1968; Roache, 1982; Chock and Dunker, 1983]. Thus the adoption of either method makes it difficult to ascertain the role of K_{yy} in the solution.

Our model uses the iterative upstream finite differencing scheme proposed by Smolarkiewicz [1983]. This scheme is positive definite, because it uses only upstream space differencing, and conservative, since fluxes are computed at grid box boundaries. We found it preferable to centered space differencing schemes because it also possesses the transportive property; i.e., perturbations in a fluid property can be advected only in the direction of fluid flow, a desirable feature in an advective model. We also find that this scheme does not generate negative species concentrations provided that the time step is not too large, and so no "borrow and fill" type remedy is needed.

The Smolarkiewicz scheme removes much of the implicit diffusion of an upwind scheme by adding a corrective step to each time step. The corrective step again uses the upstream differencing scheme but substitutes an "antidiffusion velocity" for the model wind velocities. The antidiffusion velocity is defined as

$$v_{i+1/2,j}^* = \frac{(|v_{i+1/2,j}| \Delta\phi - \Delta t v_{i+1/2,j}^2)(f_{i+1,j}^* - f_{i,j}^*)}{(f_{i,j}^* + f_{i+1,j}^* + \epsilon) \Delta\phi} \quad (30)$$

$$\omega_{i,j+1/2}^* = \frac{(|\omega_{i,j+1/2}| \Delta\zeta - \Delta t \omega_{i,j+1/2}^2)(f_{i,j+1}^* - f_{i,j}^*)}{(f_{i,j}^* + f_{i,j+1}^* + \epsilon) \Delta\zeta}$$

where f^* is the result of the uncorrected upstream step and ϵ is a small value (10^{-15}) used to avoid division by zero. The original formulation of Smolarkiewicz suggested the use of an optimization factor which produces better results in his sensitivity studies. This factor is not used in the present study as it does not significantly improve our result in the limited number of cases we tried. We have performed sensitivity analysis using advection by a circular wind field similar to the work of Molenkamp [1968] and Chock and Dunker [1983] and estimated that the effective numerical diffusion coefficient corresponds to $K_{yy} \sim 1 \times 10^8$ cm² s⁻¹. The species transport equation (29) is solved using forward time differencing with a time increment of 4 hours.

6. DIAGNOSTIC CALCULATION OF THE CIRCULATION

Heating rates for January were taken from Murgatroyd and Singleton [1961] above 25 km and from Dopplack [1979] between 25 km and the tropopause. Although Dopplack's [1979] result included values for the troposphere, his values are not applicable in that region since latent heat release was not included as evident from the fact that his result shows radiative cooling at all latitudes below 15 km. We have introduced a positive $\langle Q \rangle$ region in the tropical troposphere in accordance with the recent data of Wei et al. [1983]. There is some indication that the heating rates for the 1961 study of Murgatroyd and Singleton are too high by perhaps at least a factor of 2 around the 20- to 35-km region as compared to the more recent result of Dopplack for the same region. To avoid the mismatch of the two data sets near the 25-km level, we choose to globally (instead of point by point) fit the three data sets with a sum of hyperbolic functions, with bias toward

TABLE 1. Parameters Used in Fitting the Heating Rate Data

A_i deg/d	h_i	ϕ_i	z_i km	v_i
5.5000	2.8000	-85.000	62.000	2.2000
1.4000	9.0000	0.000	53.000	2.3000
1.1000	3.0000	-35.000	35.000	5.0000
-13.800	9.0000	85.000	70.000	3.3000
-3.5000	5.5000	64.000	60.000	2.5000
-2.3000	4.2000	40.000	50.000	3.5000
-0.30000	4.0000	40.000	2.0000	3.5000
0.30000	1.9000	0.0000	10.000	3.2000
-1.6000	2.8000	-70.000	25.000	2.6000
0.20000	8.0000	30.000	18.900	4.0000
0.40000	2.0000	20.000	70.000	20.000
-1.5000	2.0000	-10.000	80.000	40.000

Dopplck's values in the lower stratosphere:

$$\langle Q \rangle(\phi, z) = \sum_{i=1}^{12} A_i \operatorname{sech} [h_i(\phi - \phi_i)/90^\circ] \times \operatorname{sech} [v_i(z - z_i)/40 \text{ km}] \quad (31)$$

where z is the geometrical altitude, A_i , h_i , ϕ_i , v_i , and z_i are parameters listed in Table 1.

The vertical velocity in the equilibrium-pressure grid is given by

$$\langle \omega_e \rangle = - \langle \rho \rangle g \langle Q \rangle / \langle \Gamma \rangle \quad (32)$$

where $\langle \Gamma \rangle$ is the static stability (g/c_p) + $(\partial \langle T \rangle / \partial z)$. The value of $\partial \langle T \rangle / \partial z$ is usually small in comparison with g/c_p for the present study. The model calculated temperature of *Harwood and Pyle* [1977], which is close to the observed, is used in the calculation of $\langle \Gamma \rangle$. In evaluating (32), the function $\langle Q \rangle$ is reexpressed approximately in the equilibrium pressure coordinates (see the appendix). The quantities on the right-hand side of (32) are evaluated as functions of ϕ and p_e .

The zonal mean continuity equation (27) implies the exist-

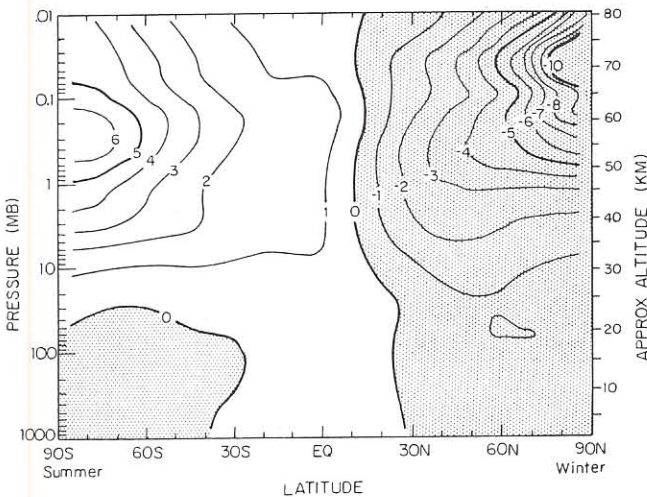


Fig. 1. The latitude-altitude distribution of the zonal mean diabatic heating rate (degrees/day) used for calculating the meridional circulation. The heating rate is obtained by fitting the results of *Murgatroyd and Singleton* [1961], *Dopplck* [1979], and *Wei et al.* [1983] to a sum of hyperbolic functions and making adjustment to ensure mass conservation.

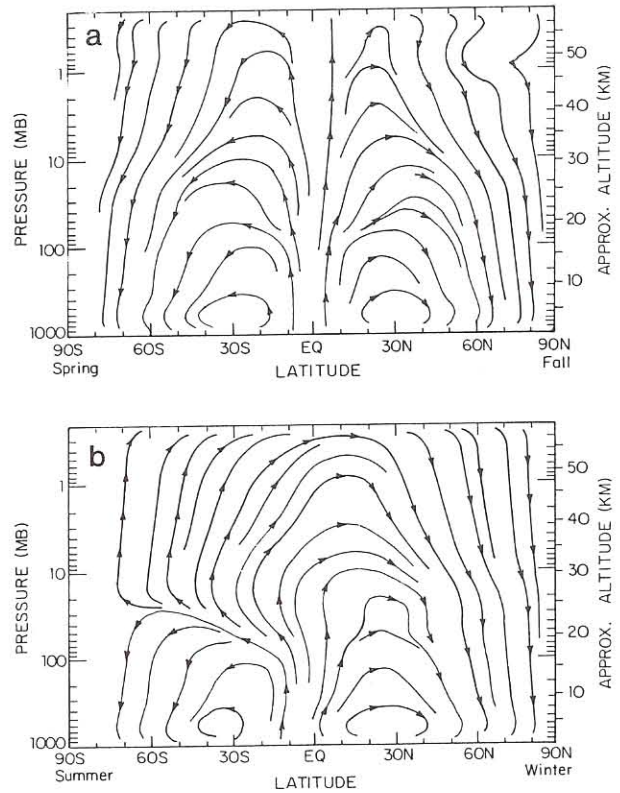


Fig. 2. The streamlines of the calculated circulation for (a) September 1 and (b) January 1. The circulation is computed using the heating rate given in Figure 1 with the top boundary condition imposed at 0.2 mbar.

tence of a stream function ψ with

$$\langle \omega_e \rangle = \frac{1}{a} \frac{1}{\cos \phi} \frac{\partial \psi}{\partial \phi} \quad \langle v \rangle = - \frac{1}{\cos \phi} \frac{\partial \psi}{\partial p_e} \quad (33)$$

The boundary condition imposed on the circulation is that ψ should be zero at the boundaries. The $\langle Q \rangle$ obtained from the data set does not necessarily lead to a ψ that vanishes exactly at the two poles. To ensure strict mass conservation in the computational domain, we employed the technique used by *Murgatroyd and Singleton* [1961] in which a small correction Δ is added to $\partial \langle \omega_e \rangle / \partial p_e$ at each pressure level such that

$$\int_{-\pi/2}^{\pi/2} \left(\frac{\partial \langle \omega_e \rangle}{\partial p_e} + \Delta \right) \cos \phi \, d\phi = 0 \quad (34)$$

The corrected $\langle v \rangle$ and $\langle \omega_e \rangle$ fields were used to calculate the stream function ψ and also to recalculate the heating rate $\langle Q \rangle$. This new $\langle Q \rangle$, which is only slightly changed from the original, is shown in Figure 1. The calculated $\langle Q \rangle$ is also in overall agreement with the heating rate of *Wehrbein and Leovy* [1982] in the upper stratosphere.

The stream function for January (ψ_w) is calculated using the $\langle Q \rangle$ in Figure 1. Annual variation is obtained by recalculating the stream function at 10-day intervals, assuming a sinusoidal seasonal variation with

$$\psi(t) = \frac{\psi_w + \psi_s}{2} + \frac{\psi_w - \psi_s}{2} \cos \frac{2\pi t}{365} \quad (35)$$

where t is the day of the year (January 1 = 1), and ψ_s is the July stream function obtained from ψ_w by switching the two hemispheres. Streamlines for January 1 and September 1 are

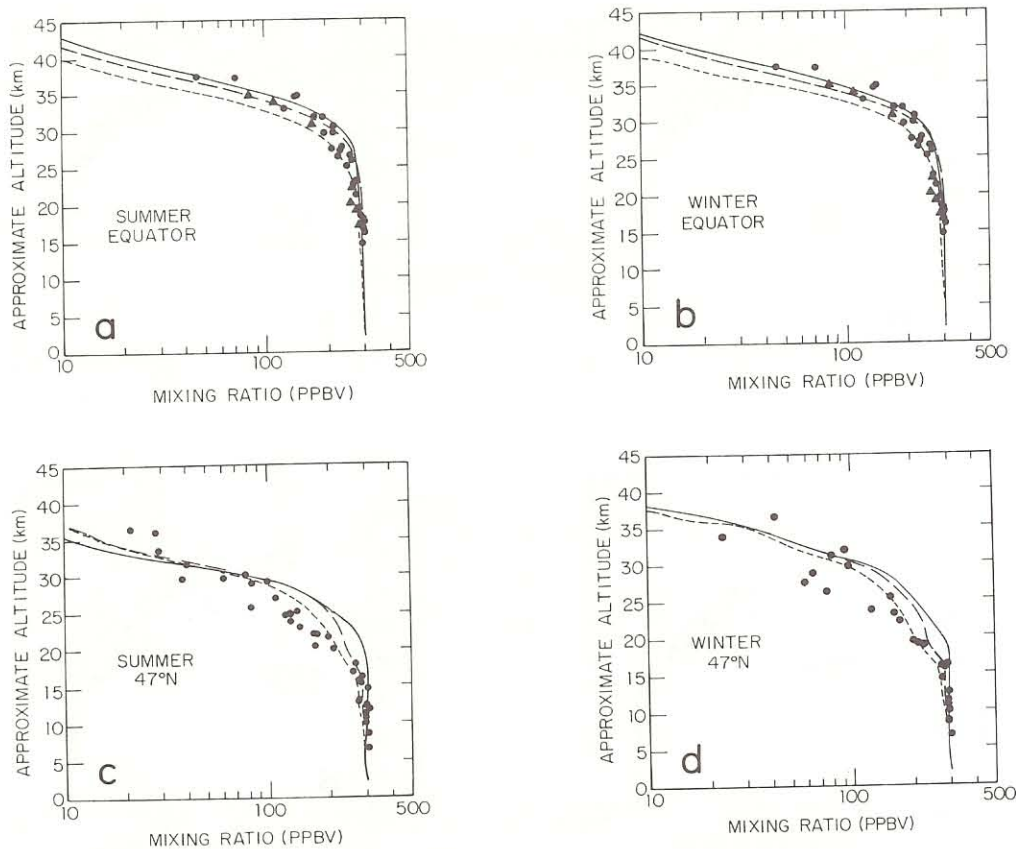


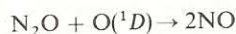
Fig. 3. Calculated altitude profiles of N_2O for different values of K_{yy} together with observations. Results are presented for the equatorial and mid-latitude region for summer and winter. The observations are from the National Center for Atmospheric Research and National Oceanic and Atmospheric Administration group as compiled in *WMO/NASA* [1981]. These curves are for $K_{yy} = 0$ (solid curves), $K_{yy} = 1 \times 10^9 \text{ cm}^2 \text{ s}^{-1}$ (dashed curves), and $K_{yy} = 3 \times 10^9 \text{ cm}^2 \text{ s}^{-1}$ (dotted curves).

shown in Figure 2 where they are expressed in terms of pressure coordinate converted approximately from θ using $\langle T \rangle$ in place of T (see the appendix).

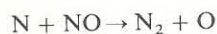
7. SIMULATION OF TRACE GAS DISTRIBUTIONS

The availability of recent field data for many stratospheric species makes it possible to diagnose the appropriateness of various transport mechanisms by comparing the model calculations with observations. The upward diffusing species including N_2O , CH_4 , and CFC's are particularly useful for this purpose, since they are characterized by relatively simple chemistry and their photochemical time constants in the stratosphere are comparable to those of transport.

Nitrous oxide (N_2O) is released at ground level and is transported into the stratosphere where it is removed mainly by photolysis. The reaction of $O(^1D)$ with N_2O



is a relatively minor sink for N_2O , but it provides the dominant source of odd nitrogen in the stratosphere. The sum of various odd nitrogen species $[N] + [NO] + [NO_2] + [HNO_4] + [HNO_3] + [ClNO_3] + 2[N_2O_5]$ is often referred to as total NOY. Removal of NOY in the stratosphere is mainly due to



In addition, the NOY species as a group is transported into

the troposphere where it is removed by heterogeneous processes such as rainout, washout, and cloud scavenging. In its role as a downward diffusing species, NOY, which has a relatively simple chemistry, can be used as a role model for the behavior of O_3 . The concentration of HNO_3 may be readily derived from the NOY distribution assuming the partition between various nitrogen species is determined by equilibrium chemistry. A full diurnal photochemical model [Ko *et al.*, 1984] is used in the calculation of the NOY species including HNO_3 . In this calculation, the distribution of ozone is taken from Ko *et al.* [1984].

Our numerical results include simulations for N_2O , NOY, and HNO_3 . Each of these species may be used for testing the various aspects of the transport scheme incorporated in our model. For instance, the relatively steep vertical gradient of N_2O as revealed by observations should be useful for testing the appropriate choice of K_{zz} in the model. However, the horizontal gradient of N_2O is relatively small; thus we expect N_2O to be less sensitive to different choices of K_{yy} . Nitric acid (HNO_3), because of its shorter photochemical lifetime of about several weeks, is expected to be much more sensitive to K_{yy} . Observations [Girard *et al.*, 1983; Gille *et al.*, 1984] indicate that the column density of HNO_3 varies by more than a factor of 10 between the equator and 60° latitude, implying a horizontal length scale as small as 10^3 km. A rough estimate from (17) would suggest that the spatial distribution of HNO_3 could be sensitive to K_{yy} even if its magnitude is less than

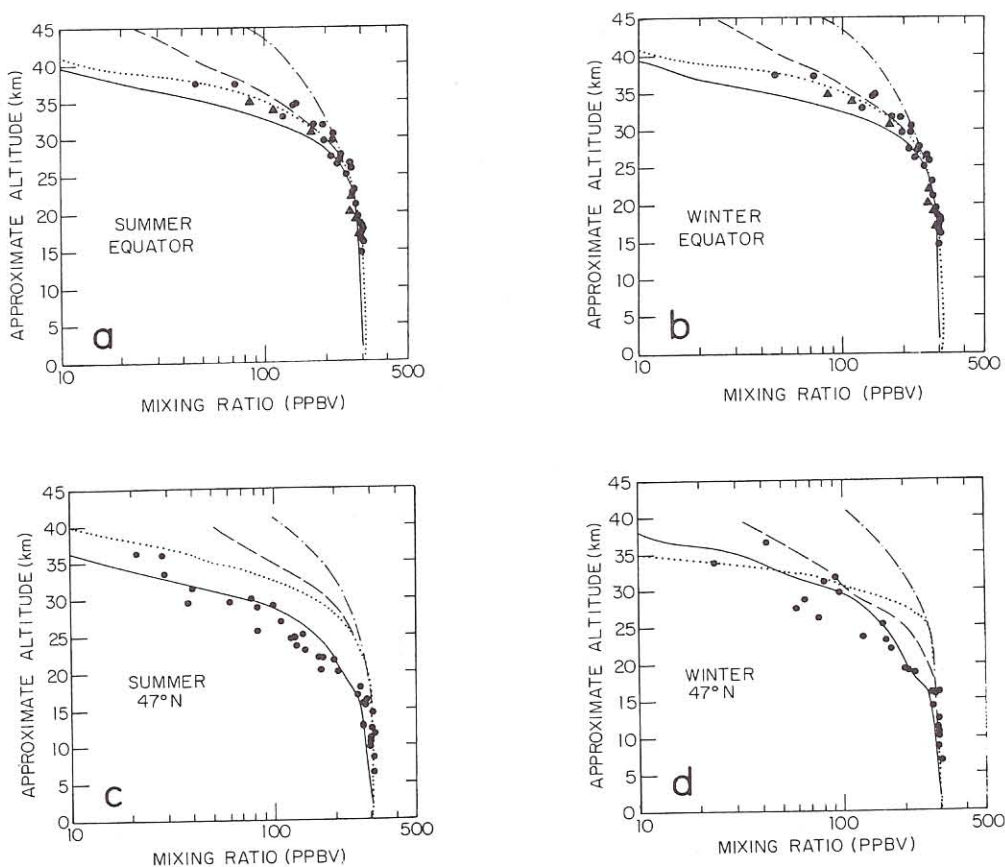


Fig. 4. Comparison of the calculated altitude profiles of N_2O with results from other models. The calculated profiles (solid curves) are calculated with $K_{yy} = 0$ for latitude of 0° (Figures 4a, 4b) and $47^\circ N$ (Figures 4c, 4d). The results from other models are $11^\circ N$ and $30^\circ N$ (dash-dot curves) from Garcia and Solomon [1983], 0° and 40° – $45^\circ N$ (dotted curves) from Guthrie et al. [1984], and $5^\circ N$ and $45^\circ N$ (dashed curves) from Miller et al. [1981]. For the results of Garcia and Solomon [1983] and Miller et al. [1981], the same profiles are used for either season.

$3 \times 10^9 \text{ cm}^3 \text{ s}^{-1}$. The present work is designed as a first step to estimate the order of magnitude for the eddy diffusion coefficient. A constant value of K_{yy} will be used in each of the calculations. It is likely, however, that K_{yy} could be a function of latitude, altitude, and seasons.

The calculated stratospheric distributions of N_2O and NO_y are not sensitive to the precise formulation of mixing processes in the troposphere in our model. Nitrous oxide has a long tropospheric lifetime and is observed to be well mixed in the troposphere. A fixed mixing ratio lower boundary condition is used for N_2O in the simulation to maintain the observed surface concentration of 300 ppbv at all latitudes. Because of the lack of gradient in the troposphere, the calculated distribution of N_2O is not sensitive to the representation of a mixing process in the troposphere. The total odd nitrogen is removed rapidly in the troposphere by a heterogeneous process with a lifetime of about 10 days. Thus little tropospheric NO_y will be transported into the stratosphere. This also means that the choice of lower boundary values for NO_y , set at 0.1 ppbv, should not affect the model-simulated stratospheric distribution.

As mentioned in section 4, we have not incorporated the chemical eddy terms into our present model. Consequently, $\langle P \rangle$ is calculated using the zonal mean values for the species concentrations. In order to facilitate comparison with observations, the results calculated in the equilibrium pressure co-

ordinates will be transformed into the usual pressure coordinates according to (18) and (19).

8. RESULTS AND DISCUSSION

N_2O

Calculations were performed using the derived diabatic circulation but with different values for K_{yy} ($K_{yy} = 0, 1 \times 10^9 \text{ cm}^2 \text{ s}^{-1}, 3 \times 10^9 \text{ cm}^2 \text{ s}^{-1},$ and $1 \times 10^{10} \text{ cm}^2 \text{ s}^{-1}$). In each case, the model was run to an annually periodic state. Figure 3 shows the vertical profiles of the volume mixing ratio of N_2O for the equatorial and mid-latitude region for summer and winter with observed data. Note that the present calculations are performed using the values of the O_2 cross section in the Herzberg continuum as recommended by World Meteorological Organization/National Aeronautics and Space Administration [1981] (hereinafter referred to as WMO/NASA [1981]). If the smaller values of the O_2 cross section [Herman and Mentall, 1982] were used, the calculated stratospheric concentrations could be reduced by about 20% above 30 km.

In general, the model calculated results compare quite well with observation in these latitudes. There is no significant display of sensitivity to the value of horizontal diffusion coefficient used for this particular species. However, we will show below that the model results for N_2O are quite sensitive to the choice of vertical diffusion coefficient.

To see how our advective model performs in relation to other two-dimensional models using the residual mean formulation, we present in Figure 4 a comparison with the model results of Miller et al. [1981], Garcia and Solomon [1983], and Guthrie et al. [1984]. The differences between these models should be attributed, in our opinion, mainly to the difference between the various sets of transport parameters and not to the difference between the residual mean and isentropic coordinate formulations, as all models use a similar diabatic circulation approximately computed from the mean diabatic heating rate in essentially the same manner. Both Miller et al. [1981] and Garcia and Solomon [1983] used diffusion coefficients with magnitudes comparable to those used in the conventional models. As discussed in section 3, these values are probably too large by an order of magnitude in models already incorporating a residual diabatic circulation. The large diffusion coefficients, in particular, K_{zz} for the present case, yield a smaller vertical gradient for the N_2O than observations indicate.

The calculated profiles from the model of Guthrie et al. [1984] appear to be closest to ours (and to the observed data), though there are still some significant differences in the mid-latitude region. Guthrie et al. used $K_{yy} = 2 \times 10^9 \text{ cm}^2 \text{ s}^{-1}$ and $K_{zz} = 2 \times 10^3 \text{ cm}^2 \text{ s}^{-1}$, which are within the physical range suggested by Kida [1983] and Tung [1984], and consequently obtained a sharper vertical decay in the vertical profiles compared to the results of Miller et al. [1981] and Garcia and Solomon [1983]. The excess N_2O in mid-latitude regions in Guthrie et al.'s model appears to be attributable to their diabatic heating rate, which has a discontinuity near the 25-km level (a result of their merging the data sets of Murgatroyd and Singleton [1961] and Dopplick [1979]). The sharp vertical

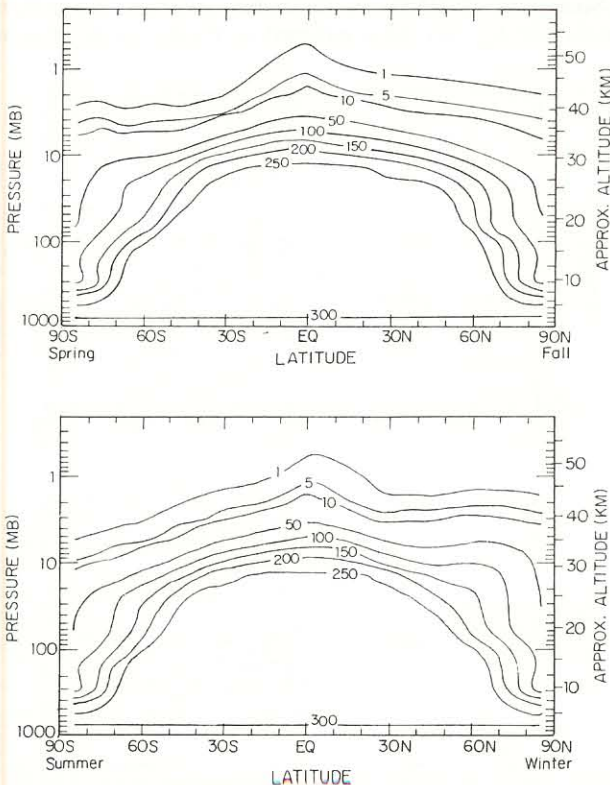


Fig. 5. Calculated altitude-latitude distribution of the volume mixing ratio of N_2O for different seasons with $K_{yy} = 0$. The contours are labeled in units of parts per billion by volume.

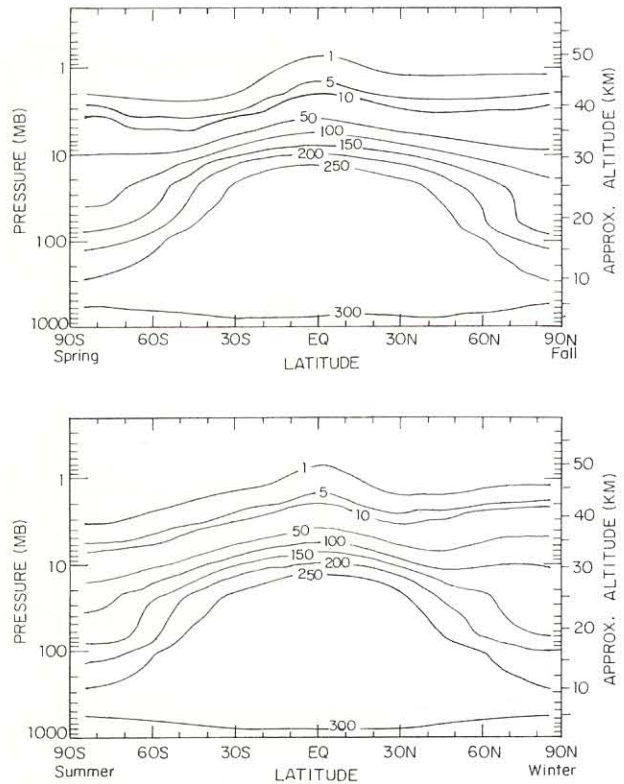


Fig. 6. Same as in Figure 5, but for $K_{yy} = 1 \times 10^9 \text{ cm}^2 \text{ s}^{-1}$.

gradient in the heating rate (and hence in the vertical velocity) tends to give rise to an enhanced meridional advection (from the continuity equation) which transports the richer tropical air to the mid-latitude regions. This minor technical problem is avoided in our model with a global definition of the diabatic heating rate.

Although the calculated vertical profiles of N_2O above the equator and mid-latitudes (Figure 3) do not show large sensitivity to different choices of K_{yy} , examination of the calculated latitude-height contours of N_2O (Figures 5, 6, and 7) reveals the dependence of the calculated N_2O isopleths on different K_{yy} near the mid-latitude region. In the case in which K_{yy} is set equal to zero (Figure 5), the calculated isopleths tend to slope steeply downward near the poles. This feature may be readily explained in terms of the balance between the advection by the diabatic circulation (see Figure 2) and the removal of N_2O by photochemical processes. As air crosses the tropical tropopause, it carries relatively high concentrations of N_2O into the tropical stratosphere. Part of the N_2O is depleted by photolysis and the reaction with $O(^1D)$ in the stratosphere. As a result, the return flow of stratospheric air into the polar troposphere must carry lower concentrations of N_2O . Introducing diffusion or mixing along the isentropic surfaces has the effect of smoothing (see Figures 6 and 7) the very large horizontal gradient (Figure 5) implied by the balance between the photochemical removal and advection by the diabatic circulation. The results in Figures 6 and 7 show that the smoothing of horizontal gradients of N_2O may be effectively achieved with a relatively small K_{yy} , about $1\text{--}3 \times 10^9 \text{ cm}^2 \text{ s}^{-1}$.

The observed N_2O mixing ratio deduced from the NIMBUS 7 SAMS measurements [Jones and Pyle, 1984] is also available for comparison. Our calculated values are somewhat smaller than the measured values, although they

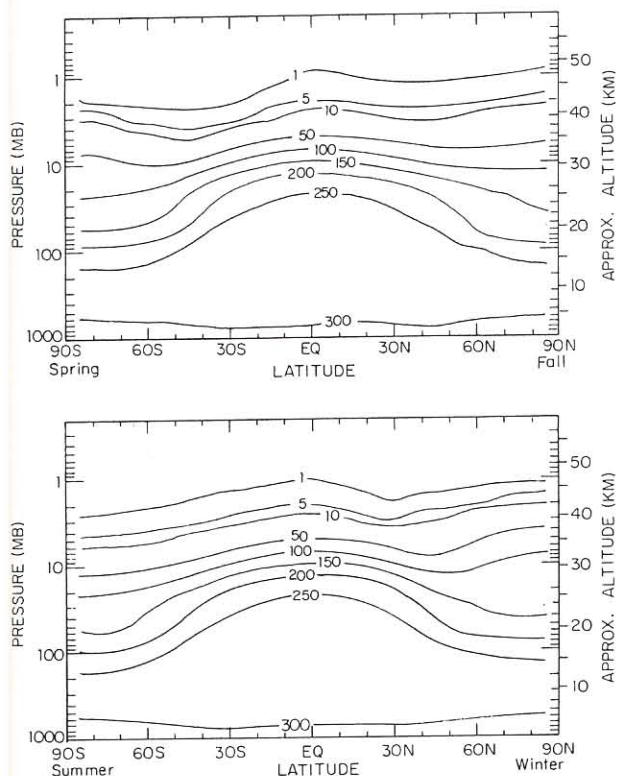


Fig. 7. Same as in Figure 5, but for $K_{yy} = 3 \times 10^9 \text{ cm}^2 \text{ s}^{-1}$.

are within the quoted precision estimates [Jones and Pyle, 1984]. In addition, the satellite data reveal significantly more robust features in the seasonal variations than our model indicates. This discrepancy could be due to the lack of seasonality of the eddy coefficient used in our model.

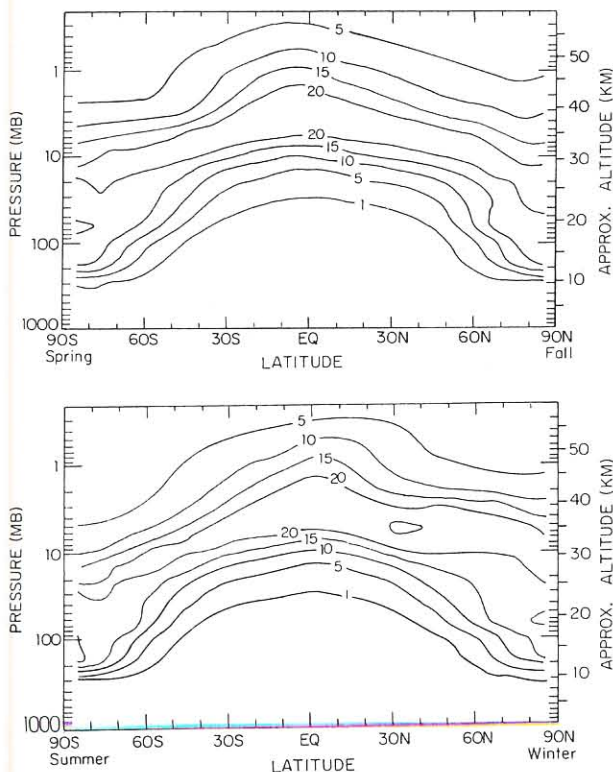


Fig. 8. Calculated altitude-latitude distribution of the volume mixing ratio of NOY for different seasons for $K_{yy} = 0$. The contours are labeled in units of parts per billion by volume.

NOY and HNO_3

The mixing ratios of NOY calculated with $K_{yy} = 0$ and $3 \times 10^9 \text{ cm}^2 \text{ s}^{-1}$ are shown in Figures 8 and 9, respectively. Since N_2O is the precursor for NOY in the stratosphere, the calculated abundance of NOY varies linearly with N_2O concentration. As noted in the preceding section, the stratospheric abundance of N_2O is somewhat sensitive to the choice of O_2 cross section in the Herzberg continuum. The use of the smaller O_2 cross section [Herman and Mentall, 1982] would yield lower NOY concentrations (by about 10%) than those presented in Figures 8 and 9, which are calculated with the larger O_2 cross section as recommended by WMO/NASA [1981].

The shape of the NOY mixing ratio isopleths in Figures 8 and 9 is similar to those of N_2O (Figures 5 and 7) calculated with the corresponding K_{yy} . Air entering the stratosphere from the tropical troposphere contains relatively low mixing ratio of NOY. Since NOY is formed in the middle to upper stratosphere over the tropics, the return flow of stratospheric air into the high-latitude troposphere contains high mixing ratio of NOY. The steep downward slope (from equator to pole) of NOY isopleths calculated with $K_{yy} = 0$ (Figure 8) reflects the balance between the production of NOY in the tropical mid-stratosphere and the advection by diabatic circulation (Figure 2). As eddy mixing along isentropic surfaces is incorporated in the model, the slope of NOY isopleths becomes less steep (Figure 9). That NOY isopleths slope downward from equator to pole implies lower stratospheric column abundance of NOY above the tropics and higher column abundance above the high-latitude region.

In the lower stratosphere, where the distribution of the NOY is most sensitive to dynamic transport, the bulk of the NOY is in the form of HNO_3 . Figure 10 shows the column abundances of HNO_3 in December calculated for different values of K_{yy} . We have included in Figure 10 the observed

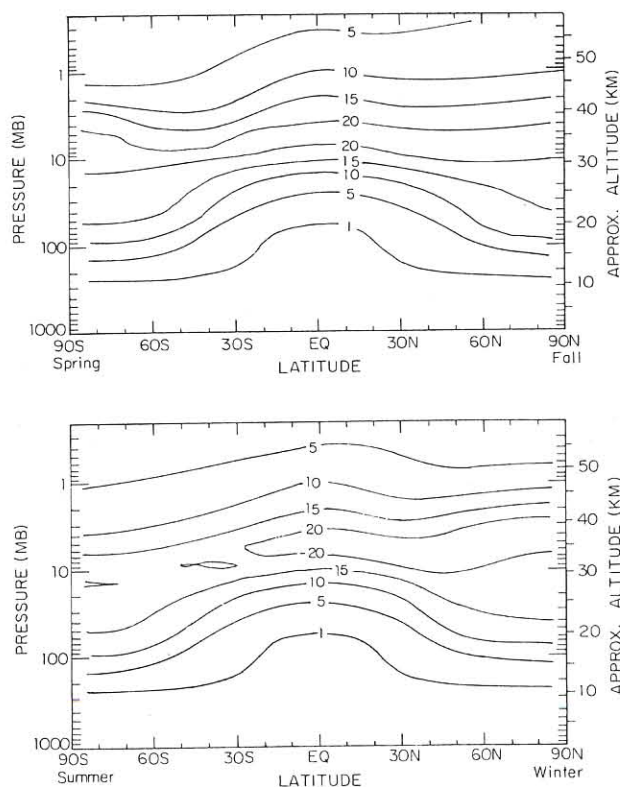


Fig. 9. Same as Figure 8, but for $K_{yy} = 3 \times 10^9 \text{ cm}^2 \text{ s}^{-1}$.

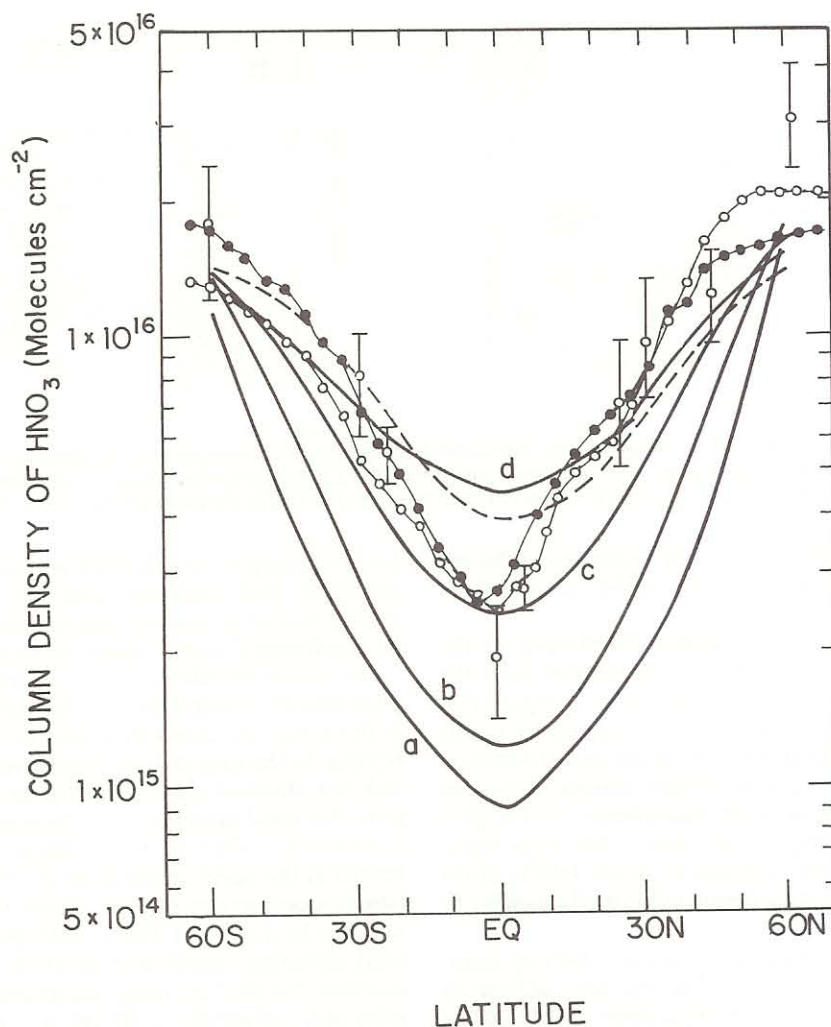


Fig. 10. Calculated latitudinal distribution of the stratospheric column density of HNO_3 . The calculated results are for (curve a) $K_{yy} = 0$, (curve b) $K_{yy} = 1 \times 10^9 \text{ cm}^2 \text{ s}^{-1}$, (curve c) $K_{yy} = 3 \times 10^9 \text{ cm}^2 \text{ s}^{-1}$, and (curve d) $K_{yy} = 1 \times 10^{10} \text{ cm}^2 \text{ s}^{-1}$. The data are April/May 1980 (bars) from Girard *et al.* [1982], May (dash-solid circle curve) and December (dash-open circle curve) from Gille *et al.* [1984]. The calculated column density from Ko *et al.* [1984] (dashed curve) is included for comparison.

abundances reported by Girard *et al.* [1982] and Gille *et al.* [1984]. Earlier observations by Coffey *et al.* [1981] and Murracray *et al.* [1975], which have been shown to be in good agreement with the more recent results [Gille *et al.*, 1984], have been left out for clarity. The observed abundances show an order of magnitude increase from the equator to 60° . The calculated results with $K_{yy} = 1 \times 10^9 \text{ cm}^2 \text{ s}^{-1}$ and $3 \times 10^9 \text{ cm}^2 \text{ s}^{-1}$ (curves b and c) produce a gradient similar to this observed trend. The case with $K_{yy} = 1 \times 10^{10} \text{ cm}^2 \text{ s}^{-1}$ (curve d) exhibits a latitudinal gradient too small in comparison with the observation, with only a factor of 4 change from the equator to the high latitudes. Judging from the observed column density of HNO_3 at the equator and the latitudinal gradient, a value of K_{yy} between $1 \times 10^9 \text{ cm}^2 \text{ s}^{-1}$ and $3 \times 10^9 \text{ cm}^2 \text{ s}^{-1}$ would appear to yield the best results.

The observed column abundance of HNO_3 from the NIMBUS 7 LIMS measurements [Gille *et al.*, 1984] also reveals interesting seasonal behavior of HNO_3 . The data (Figure 10) show that, for the month of December, the abundance at the high latitudes in the winter hemisphere (80°N) is about a factor of 1.5 higher than that in the summer hemisphere (60°S). Given the location of the photochemical source of HNO_3 (in the midstratosphere over the mid-latitudes of the

southern hemisphere and equator), this behavior is consistent with the direction of the circulation shown in Figure 2 where downward motion in the winter hemisphere would enhance the HNO_3 abundance at the high-latitude winter. Our calculation shows a similar trend in the seasonal behavior, although the difference between the hemispheres is only about a factor of 1.2.

Comparison with other model calculations is complicated by the fact that the calculated HNO_3 distributions depend on the different photochemical treatments used in the models as well as the different dynamical treatments. Included in Figure 10 is the result from Ko *et al.* [1984] which uses the same photochemical treatment. The result there is similar to curve d ($K_{yy} = 1 \times 10^{10} \text{ cm}^2 \text{ s}^{-1}$) both in the absolute magnitude and the latitudinal trend. Calculated values for HNO_3 are also reported by Solomon and Garcia [1983] and Miller *et al.* [1981]. The calculated gradient from Solomon and Garcia [1983] is similar to our curve d. In contrast to what we find, they reported no significant change in their result when K_{yy} is changed from $1 \times 10^9 \text{ cm}^2 \text{ s}^{-1}$ to $1 \times 10^{10} \text{ cm}^2 \text{ s}^{-1}$ [Garcia and Solomon, 1983]. The result of Miller *et al.* [1981] is similar to our curve c, although the values for K_{yy} in their model, which are taken from Luther [1973], are about an order of

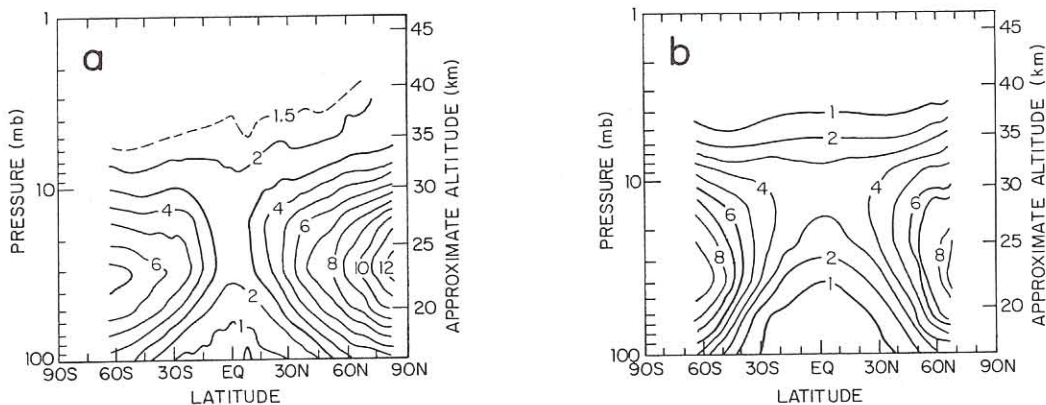


Fig. 11. Comparison of the observed and calculated latitude/altitude distribution of the concentration of HNO_3 (ppbv). The observed concentration (Figure 11a) is from NIMBUS 7 LIMS measurement [Gille *et al.*, 1984] for the month of November. The calculated result (Figure 11b) is obtained using the NOY from the case with $K_{yy} = 3 \times 10^9 \text{ cm}^2 \text{ s}^{-1}$.

magnitude larger. The difference may be attributed to the use of latitudinally dependent values for K_{zz} in their model (which are somewhat arbitrary).

The observed latitudinal and vertical distribution of the zonal mean mixing ratio of HNO_3 for November from the NIMBUS 7 LIMS [Gille *et al.*, 1984] is shown in Figure 11a. The mixing ratio surface can best be described as saddle shaped, with the saddle point occurring in the midstratosphere (~ 20 mbar) above the equator. Higher mixing ratios are found toward the poles in both hemispheres, with higher values of HNO_3 occurring in the winter (northern) hemisphere. Also, note that the altitudes at which HNO_3 peaks tend to follow a slope directing downward from the equator to poles in both hemispheres.

Our model results reproduce all of the above features qualitatively, although some notable quantitative differences remain. Figure 11b shows the latitude/altitude distribution of HNO_3 for December calculated with $K_{yy} = 3 \times 10^9 \text{ cm}^2 \text{ s}^{-1}$. The agreement between Figure 11a (observed) and Figure 11b (calculated) is quite satisfactory, although the model results reveal less seasonal contrast (hemispheric differences) than suggested by observation.

9. CONCLUSION

In this paper, we presented results from the numerical simulation of the distribution of N_2O , NOY, and HNO_3 from a two-dimensional model and found them to be in good overall agreement with observations. We have made no attempt to exploit the latitudinal and seasonal dependence of K_{yy} to obtain better agreement with observations. Our model results appear to support the contention that the large-scale systematic dynamical transport in the stratosphere for globally distributed tracers is mainly advective in nature, with the eddy diffusion playing an important role of smoothing out the large gradient created by the diabatic circulation. This conclusion is expected to be equally valid in models using the residual mean circulation in pressure coordinates.

Compared to the classical models using mean circulation with large diffusion coefficients, our model is conceptually simpler. Transport of tracer, to a large extent, may be described by the advection of the mean meridional circulation, which is in turn related in a simple manner to the distribution of the diabatic heating rate. Additional diffusive transport along isentropic surface is represented by a single horizontal eddy diffusion coefficient. The value of the diffusion coefficient

can, in principle, be calculated explicitly from the eddy displacement field (equation (16)). In contrast, the classical models require the detailed specification of three to four diffusion coefficients, none of which can be rigorously validated.

Our results include numerical calculations for stratospheric gases that are transported from the troposphere (N_2O) as well as those that are produced in situ in the stratosphere (NOY, HNO_3). In the case of N_2O , comparison of the model results with the observed altitude profiles at different latitudes suggests that good agreement may be achieved by setting $K_{zz} = 0$. However, values of K_{zz} of about $10^3 \text{ cm}^2 \text{ s}^{-1}$ may be needed in the upper stratosphere in order to bring the calculated concentrations of N_2O closer to the observed values reported by Jones and Pyle [1984]. By comparing the calculated latitudinal distribution for HNO_3 with observation, we conclude that the best result can be obtained using a constant value of K_{yy} of about $3 \times 10^9 \text{ cm}^2 \text{ s}^{-1}$ which is consistent with the independent estimates of Kida [1983] and Tung [1984]. Larger values of K_{yy} would lead to too small an equator-to-pole gradient for columnar HNO_3 as compared with observation, although our studies do not rule out the possibility of large K_{yy} ($> 10^{10} \text{ cm}^2 \text{ s}^{-1}$) in localized regions of the stratosphere.

Since the response of O_3 to dynamic transport as a downward diffusing species is similar to that of NOY, it is likely that a diabatic circulation model may also be successful in simulating the behavior of ozone. The simple picture of the diabatic circulation having a one-cell structure above 25 km with upward motion in the summer hemisphere and downward motion in the winter hemisphere suggests that there will be enhanced transport of O_3 in winter from the midstratosphere to the lower stratosphere and upper troposphere where it is less prone to photochemical removal. This quasi-horizontal advection of O_3 into the winter hemisphere provides a transport mechanism which appears to work in conjunction with photochemical processes to produce the observed springtime maximum in the column abundance of O_3 [Dütsch, 1978] as well as the observed enhanced tropospheric concentration of O_3 in winter [Wilcox *et al.*, 1977].

In this paper, we have studied the effect of eddy motions on tracer transport by using a diabatic circulation diagnostically calculated from a given diabatic heating rate. As a result, the effect of eddy motions on defining the heating rate is not considered. It is sometimes argued that the presence of any diabatic heating is ultimately due to presence of eddies which

drive the atmosphere away from radiative equilibrium. In addition, eddies may also provide some additional advective transports for the cases when $Q' \neq 0$ [Tung, 1982, 1984; Rood and Schoeberl, 1983]. Finally, the effect of eddies can also manifest itself in the form of chemical eddy terms [Matsuno, 1980; Pyle and Rogers, 1980b; Tung, 1982; Garcia and Solomon, 1983; Rogers and Pyle, 1984]. Although all these effects can be incorporated in some form into two-dimensional models in a diagnostic manner, they cannot be included prognostically unless the wave field itself is calculated in a self-consistent manner. The present study, which evaluates the effect of eddy motion that manifests itself as explicit eddy diffusion transport, is intended only as a first step in the direction of exploring the limits of the zonally averaged models. Further studies of the effect of eddy motions on diabatic heating, chemical eddies, and eddy advection as well as the determination of the spatial and temporal dependencies of K_{yy} would help to define the future role of zonal mean models. Because of its better physical basis, we feel that a properly formulated two-dimensional model can best assimilate the dynamic variables from a general circulation model for improved simulation of tracer transport.

APPENDIX: PROCEDURES FOR CALCULATING THE DIABATIC CIRCULATION

In the work of Tung [1982], a slightly different set of zonal mean equations was derived in a more systematic manner than that presented in section 2. However, to use available observational data in pressure coordinates, one needs to make further (sometimes ad hoc) approximations. The resulting set of equations is equivalent to the set presented in section 4 for practical purposes.

The following formulae for zonally averaged mass circulation have been shown in the work of Tung [1982] to be valid approximations:

$$\langle W \rangle \simeq \langle q \rangle / \langle \Gamma \rangle \tag{A1}$$

$$\frac{\partial}{\partial y} \langle V \rangle + \frac{\partial}{\partial \theta} \langle W \rangle \simeq 0 \tag{A2}$$

where $W \equiv \rho_\theta \dot{\theta}$, $V \equiv \rho_\theta v \cos \phi$, $q \equiv \rho Q$. A further approximation of replacing $\langle \Gamma \rangle$ by $\Gamma^{(0)}$, the radiative equilibrium value of Γ , can also be made for prognostic models. For the present diagnostic model, this approximation is not needed.

Once the mass circulation ($\langle V \rangle$, $\langle W \rangle$) is known, the mean diabatic advection of the tracers is given by the mean velocities ($\langle V \rangle / \langle \rho_\theta \rangle$, $\langle W \rangle / \langle \rho_\theta \rangle$), where the mean density, $\langle \rho_\theta \rangle$, is either calculated separately in a two-dimensional model or further approximated by the radiative equilibrium value $\rho_\theta^{(0)}$.

In a prognostic model, $\langle V \rangle$ and $\langle W \rangle$ are calculated at each time step, given $\langle q \rangle$. They are then used in the prognostic momentum equations to predict the new mean zonal flow and mean temperature. The latter is then input into the calculation for $\langle q \rangle$, along with the predicted distribution of the radiatively active species such as ozone and carbon dioxide. The procedure is then repeated with this new value of $\langle q \rangle$ at the next time step.

Our present purpose is more limited, as only a diagnostic study will be performed. That is, we take the mean diabatic heating rate as known from observation. This is used to deduce the advective velocities ($\langle V \rangle / \langle \rho_\theta \rangle$, $\langle W \rangle / \langle \rho_\theta \rangle$), which are then used in a tracer transport model to calculate the distribution of a few species. Favorable comparison of the

calculated species distributions with observations will hopefully give a limited validation of our transport model.

Although Wei et al. [1983] recently calculated the zonal mean diabatic heating rate in the troposphere in isentropic coordinates using the FGGE data, available information on the mean diabatic heating rate deduced from observation is mostly presented in pressure coordinates (e.g., Murgatroyd and Singleton, 1961; Dopplick, 1979; Wehrbein and Leovy, 1982) for the stratosphere. Once the zonal average has been taken in pressure coordinates, the averaged quantity can no longer be transformed to the isentropic coordinates without the original three-dimensional data set. We hope this technical problem can be alleviated in the future when more analyses are presented in isentropic coordinates. In the meantime, the available data in pressure coordinates will be adopted after applying the following transformation:

$$\theta \simeq \langle T \rangle (p_{00}/p)^{\kappa} \tag{A3a}$$

$$\langle Q \rangle (y, p) \simeq \langle Q \rangle [y, P_{00} (\langle T \rangle / \theta)^{1/\kappa}] \tag{A3b}$$

The right-hand side of (A3b) is then taken as a zonal averaged function of y and θ in isentropic coordinates. The procedure is admittedly crude. Nevertheless, given the rather large uncertainty in the "observed" values of $\langle Q \rangle$, as evident in the discrepancies from different data sets (e.g., Murgatroyd and Singleton versus Dopplick in the lower stratosphere), we hope that such a resorting procedure is permissible within the error bounds. Also consistent with this procedure, the mean diabatic heating rate per unit volume, $\langle q \rangle$, is deduced from $\langle \rho \rangle \langle Q \rangle$. At this degree of approximation, it may not be worth the effort in distinguishing $\langle \rho_\theta \dot{\theta} \rangle / \langle \rho_\theta \rangle$ from $\langle \dot{\theta} \rangle$. Consequently, we shall replace the mean advective velocities ($\langle V \rangle / \langle \rho_\theta \rangle$, $\langle W \rangle / \langle \rho_\theta \rangle$) by another set of mean advective velocities ($\langle v \rangle \cos \phi$, $\langle \dot{\theta} \rangle$). This step can be accomplished by setting ρ_θ in the mean advective velocities to its basic state value $\rho_\theta^{(0)}$ (analogous to the Boussinesq approximation), resulting in (26) and (27).

Acknowledgments. The establishment of the advective modeling program by the Fluorocarbon Program Panel of the Chemical Manufacturers Association owes much to the effort of S. R. Orfeo. We would like to thank J. Mahlman for discussions on the numerical schemes associated with the solution to the advection equation and J. Holton and M. B. McElroy for commenting on an earlier draft of the manuscript. D. Hutchison was responsible for much of the computer code development at the early stage of this research. We would like to thank Ann Aubin for preparation of the manuscript. The work is supported by the Fluorocarbon Program Panel of the Chemical Manufacturers Association.

REFERENCES

Andrews, D. G., and M. E. McIntyre, Planetary waves in horizontal and vertical shear: The generalized Eliassen-Palm relation and the mean zonal acceleration, *J. Atmos. Sci.*, **33**, 2031-2048, 1976.
 Andrews, D. G., and M. E. McIntyre, An exact theory of non-linear waves on a Lagrangian-mean flow, *J. Fluid Mech.*, **89**, 609-646, 1978.
 Boyd, J. P., The noninteraction of waves with the zonally-averaged flow on a spherical earth and the interrelationships of eddy fluxes of energy, heat, and momentum, *J. Atmos. Sci.*, **33**, 2285-2291, 1976.
 Brewer, A. W., Evidence for a world circulation provided by measurements of helium and water vapor distribution in the stratosphere, *Q. J. R. Meteorol. Soc.*, **75**, 351-363, 1949.
 Chock, D. P., and A. M. Dunker, A comparison of numerical methods for solving the advection equation, *Atmos. Environ.*, **17**, 11-24, 1983.

- Clark, J. H. E., and T. G. Rogers, The transport of trace gases by planetary waves, *J. Atmos. Sci.*, *35*, 2232–2235, 1978.
- Coffey, M. T., W. G. Mankin, and A. Goldman, Simultaneous spectroscopy determination of the latitudinal, seasonal, and diurnal variability of stratospheric N_2O , NO , NO_2 , and HNO_3 , *J. Geophys. Res.*, *86*, 7331–7341, 1981.
- Dobson, G. M. B., Origin and distribution of polyatomic molecules in the atmosphere, *Proc. R. Soc. London Ser. A*, *235*, 187–192, 1956.
- Dopplack, T. G., Radiative heating of the global atmosphere: Corrigendum, *J. Atmos. Sci.*, *36*, 1812–1817, 1979.
- Dunkerton, T., On the mean meridional mass motions of the stratosphere and mesosphere, *J. Atmos. Sci.*, *35*, 2325–2333, 1978.
- Dunkerton, T., C.-P. Hsu, and M. E. McIntyre, Some Eulerian and Lagrangian diagnostic for a model stratospheric warming, *J. Atmos. Sci.*, *31*, 819–843, 1981.
- Dütsch, H. V., Vertical ozone distribution on a global scale, *Pure Appl. Geophys.*, *116*, 511–529, 1978.
- Garcia, R. R., and S. Solomon, A numerical model of the zonally averaged dynamical and chemical structure of the middle atmosphere, *J. Geophys. Res.*, *88*, 1379–1400, 1983.
- Gille, J. C., J. M. Russell III, P. L. Bailey, E. E. Remsburg, L. L. Gordley, W. F. J. Evans, H. Fischer, B. W. Gandrud, A. Girard, J. E. Harries, and S. A. Beck, Accuracy and precision of the nitric acid concentrations determined by the limb infrared monitor of the stratosphere experiment on NIMBUS 7, *J. Geophys. Res.*, *89*, 5179–5190, 1984.
- Girard, A., L. Gramont, N. Louisnard, S. LeBoiteux, and G. Fergant, Latitudinal variation of HNO_3 , HCl and HF vertical column density above 11.5 km, *Geophys. Res. Lett.*, *9*, 135–136, 1982.
- Guthrie, P. D., G. H. Jackman, J. R. Herman, and C. J. McQuillan, A diabatic circulation experiment in a two-dimensional photochemical model, *J. Geophys. Res.*, *89*, 9589–9602, 1984.
- Harwood, R. S., Dynamical models of the middle atmosphere for tracer studies, *Philos. Trans. R. Soc. London Ser. A*, *296*, 103–127, 1980.
- Harwood, R. S., and J. A. Pyle, Studies of the ozone budget using a zonal mean circulation model and linearized photochemistry, *Q. J. R. Meteorol. Soc.*, *103*, 319–343, 1977.
- Herman, J. R., and J. E. Mentall, O_2 absorption cross section (190–225 nm) from stratospheric solar flux measurement, *J. Geophys. Res.*, *87*, 8967–8975, 1982.
- Holton, J. R., Wave propagation and transport in the middle atmosphere, *Philos. Trans. R. Soc. London Ser. A*, *296*, 73–85, 1980.
- Holton, J. R., An advective model for two-dimensional transport of stratospheric trace species, *J. Geophys. Res.*, *86*, 11,989–11,994, 1981.
- Hunt, G. B., and S. Manabe, Experiments with a stratospheric general circulation model, 2, Large-scale diffusion of tracers in the stratosphere, *Mon. Weather Rev.*, *96*, 503–539, 1968.
- Jones, R. L., and J. A. Pyle, Observations of CH_4 and N_2O by the NIMBUS 7 SAMS: A comparison with in situ data and two-dimensional numerical model calculations, *J. Geophys. Res.*, *89*, 5263–5279, 1984.
- Kida, H., General circulation of air parcels and transport characteristics derived from a hemispheric GCM, 2, Very long-term motions of air parcels in the troposphere and stratosphere, *J. Meteorol. Soc. Jpn.*, *61*, 510–523, 1983.
- Ko, M. K. W., N. D. Sze, M. Livshitz, M. B. McElroy, and J. A. Pyle, Seasonal and latitudinal behaviors of trace gases and O_3 as simulated by a 2-D model of the atmosphere, *J. Atmos. Sci.*, *41*, 2381–2408, 1984.
- Lau, N.-C., and A. H. Oort, A comparative study of observed northern hemisphere circulation statistics based on GFDL and NMC analyses, 2, Transient eddy statistics and energy cycle, *Mon. Weather Rev.*, *10*, 889–906, 1982.
- Luther, F. M., Monthly mean values of eddy diffusion coefficients in the lower stratosphere, *AIAA Pap. 73-498*, Am. Inst. of Aeronaut. and Astronaut., New York, 1973.
- Mahlman, J. D., Atmospheric general circulation and transport of radioactive debris, *Atmos. Sci. Pap.* *103*, 184 pp., Dep. of Atmos. Sci., Colo. State Univ., Fort Collins, 1966.
- Mahlman, J. D., and W. J. Moxim, Tracer simulation using a global general circulation model: Results from a mid-latitude instantaneous source experiment, *J. Atmos. Sci.*, *35*, 1340–1376, 1978.
- Mahlman, J. D., D. G. Andrews, D. C. Hartman, T. Matsuno, and R. J. Murgatroyd, Transport of trace constituents in the stratosphere, in *Dynamics of the Middle Atmosphere*, edited by J. R. Holton and T. Matsuno, pp. 387–416, Terra Scientific Publishing, Tokyo, Japan, 1984.
- Matsuno, T., Lagrangian motion of air parcels in the stratosphere in the presence of planetary waves, *Pure Appl. Geophys.*, *118*, 189–216, 1980.
- McIntyre, M. E., Towards a Lagrangian-mean description of stratospheric circulations and chemical transports, *Philos. Trans. R. Soc. London Ser. A*, *296*, 129–148, 1980.
- Miller, C., D. L. Filkin, A. J. Owens, J. M. Steed, and J. P. Jesson, A two-dimensional model of stratospheric chemistry and transport, *J. Geophys. Res.*, *86*, 12,039–12,065, 1981.
- Molenkamp, C. R., Accuracy of finite-difference methods applied to the advection equation, *J. Appl. Meteorol.*, *7*, 60–61, 1968.
- Murcray, D. G., D. B. Barker, J. N. Brooks, A. Goldman, and W. J. Williams, Seasonal and latitudinal variation of the stratospheric concentration of HNO_3 , *Geophys. Res. Lett.*, *2*, 223–225, 1975.
- Murgatroyd, R. J., and F. Singleton, Possible meridional circulation in the stratosphere and mesosphere, *Q. J. R. Meteorol. Soc.*, *87*, 125–135, 1961.
- Newell, R. W., The circulation of the upper atmosphere, *Sci. Am.*, *210*, 62–74, 1963.
- Plumb, R. A., Eddy fluxes of conserved quantities by small-amplitude waves, *J. Atmos. Sci.*, *36*, 1699–1704, 1979.
- Pyle, J. A., A calculation of possible depletion of ozone by chlorofluoromethanes using a two-dimensional model, *Pure Appl. Geophys.*, *118*, 355–377, 1980.
- Pyle, J. A., and C. F. Rogers, A modified diabatic circulation model for stratospheric tracer transport, *Nature*, *287*, 711–714, 1980a.
- Pyle, J. A., and C. F. Rogers, Stratospheric transport by stationary planetary waves—The importance of chemical processes, *Q. J. R. Meteorol. Soc.*, *106*, 421–446, 1980b.
- Reed, R. J., and K. E. German, A contribution to the problem of stratospheric diffusion by large-scale mixing, *Mon. Weather Rev.*, *93*, 313–321, 1965.
- Reed, R. J., J. Wolfe, and H. Nishimoto, A spectral analysis of the energetics of the stratospheric sudden warming of early 1957, *J. Atmos. Sci.*, *20*, 256–275, 1963.
- Roache, P. J., *Computational Fluid Dynamics*, Hermosa, Albuquerque, N. Mex., 1982.
- Rogers, C. F., and J. A. Pyle, Stratospheric tracer transport: A modified diabatic circulation model, *Q. J. R. Meteorol. Soc.*, *110*, 219–237, 1984.
- Rood, R. B., and M. R. Schoeberl, A mechanistic model of Eulerian, Lagrangian mean and Lagrangian ozone transport by steady planetary waves, *J. Geophys. Res.*, *88*, 5200–5218, 1983.
- Smolarkiewicz, P. K., A simple positive definite advection scheme with small implicit diffusion, *Mon. Weather Rev.*, *111*, 479–487, 1983.
- Solomon, S., and R. R. Garcia, On the distribution of nitrogen dioxide in the high-latitude stratosphere, *J. Geophys. Res.*, *88*, 5229–5239, 1983.
- Sze, N. D., R. G. Isaacs, M. Ko, and M. B. McElroy, Atmospheric studies related to aerospace activities and remote sensing technology, 3, *NASA Tech. Rep.*, *3410*, 1981.
- Tuck, A. F., A comparison of one-, two-, and three-dimensional model representations of stratospheric gases, *Philos. Trans. R. Soc. London Ser. A*, *290*, 477–498, 1979.
- Tung, K. K., On the two-dimensional transport of stratospheric trace gases in isentropic coordinates, *J. Atmos. Sci.*, *39*, 2330–2355, 1982.
- Tung, K. K., Modeling of tracer transport in the middle atmosphere, in *Dynamics of the Middle Atmosphere*, edited by J. R. Holton and T. Matsuno, pp. 412–444, Terra Scientific Publishing, Tokyo, Japan, 1984.
- Vupputuri, R. K. R., The structure of the natural stratosphere and the impact of chlorofluoromethanes on the ozone layer investigated in a 2-D time dependent model, *Pure Appl. Geophys.*, *117*, 448–485, 1978.
- Wehrbein, W. M., and C. B. Leovy, An accurate radiative heating and cooling algorithm for use in a dynamic model of the middle atmosphere, *J. Atmos. Sci.*, *39*, 1532–1564, 1982.
- Wei, M. Y., D. R. Johnson, and R. D. Townsend, Seasonal distributions of diabatic heating during the first GARP global experiment, *Tellus*, *35A*, 241–255, 1983.
- Wilcox, R. W., G. D. Nastrom, and A. D. Belmont, Periodic variations of total ozone and of its vertical distribution, *J. Appl. Meteorol.*, *16*, 290–298, 1977.
- World Meteorological Organization/National Aeronautics and Space

Administration, The stratosphere 1981: Theory and measurements, *Ozone Res. and Monit. Proj. Rep. 11*, World Meteorol. Organ., Geneva, 1981.

K. K. Tung, Department of Mathematics, Massachusetts Institute of Technology, Cambridge, MA 02139.

M. K. W. Ko, N. D. Sze, and D. K. Weisenstein, Atmospheric and Environmental Research, Inc., 840 Memorial Drive, Cambridge, MA 02139.

(Received April 30, 1984;
revised September 24, 1984;
accepted October 1, 1984.)

