On the Two-Dimensional Transport of Stratospheric Trace Gases in Isentropic Coordinates

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ABSTRACT

A zonally averaged model of stratospheric tracer transport is formulated in isentropic coordinates. There are some conceptual and computational advantages, as well as some disadvantages, in adopting the potential temperature, instead of pressure, as the vertical coordinate. The main disadvantage is that the "density" (mass per unit coordinate volume) in isentropic coordinates is no longer a constant as in the pressure coordinate system under the hydrostatic approximation. However, it can be shown that this density effect is almost negligible in the calculation of the mean diabatic circulation and the eddy advective transports. What is gained by adopting the new formulation is a conceptually simpler picture of the interplay of diabatic and adiabatic processes in the transport of tracers. Mean diabatic heating (cooling) forces a direct rising (descending) mean mass flow. Along the streamlines of this mean mass circulation, tracers are advected in the mean. These surfaces slope downward and poleward in the lower stratosphere. In addition to advection, tracers are also dispersed from their mean path by transient adiabatic processes in a direction parallel to the local isentropic surface. As a result, the lines of mean constant tracer mass mixing ratio slope less steeply than the mean streamlines, but more steeply than the isentropic surfaces. The effect of eddy transport on chemically reacting minor constituent gases is also discussed.

1. Introduction

In the stratosphere, dynamics, radiation and photochemistry are coupled in a complicated feedback loop. The motion field which transports the trace elements is itself driven, to a large extent, by the diabatic (radiative) heating and cooling caused by the absorption of solar radiation and the emission of longwave radiation, processes which depend strongly on the concentration of some trace elements such as ozone and carbon dioxide. The incorporation of these interactions in two-dimensional (zonally averaged) models has been impeded by the inevitable presence of eddy fluxes. The common practice of parameterizing the eddy fluxes using mixing-length theory (Reed and German, 1965) fails to fully take into account such a feedback process, since usually the same set of empirical parameters is used, even when the sources and sinks are altered. The readers are referred to Harwood (1980) for a review of two-dimensional (2-D) models incorporating the so-called K-theory for the eddy fluxes and to the MAP report (Mahlman et al., 1981) for a more comprehensive review and discussion of the current developments.

It is well-known by now that the zonally averaged meridional circulation in the stratosphere is thermally *indirect* over the winter latitudes, where eddy activity is predominant (Miyakoda, 1963; Reed et al., 1963; Vincent, 1968). Instead of rising motion in regions of net heating and subsidence where the atmosphere cools, the observed zonally averaged circulation in the lower stratosphere consists of upward motion in the winter polar region and over the tropics, and descending motion over the middle latitudes. The explanation, as given by Newell (1963), is that the stratosphere acts more or less like a refrigerator, *driven*, to a large extent, by upward-propagating waves from the troposphere, whose effect on the mean circulation overwhelms the thermally direct diabatic circulation forced *in situ*. It has also been recognized that conservative tracers are transported neither by the observed (Eulerian) mean circulation, nor by the eddy fluxes alone, but the trajectories appear to approximately follow the diabatic circulation, which is a small residue of the two (Brewer, 1949; Dobson, 1956; Newell, 1963; Hunt and Manabe, 1968; Mahlman, 1969; Mahlman and Moxim, 1978; Dunkerton, 1978). Therefore, it is not surprising to find that in model calculations of tracer transport using the *observed* zonal mean circulation, a large, usually parameterized eddy flux transport is needed, with the net transport obtained as a small difference of the two large terms. This peculiarity of most existing two-dimensional models is a constant source of numerical inaccuracy, in addition to the lack of physical basis for the diffusion type of parameterization currently in use (Mahlman, 1975; Clark and Rogers, 1978; Plumb, 1979; Matsuno, 1980).

Since the separation of the eddy from the mean is simply a matter of convention and depends entirely on how the mean is taken, as was first indicated by Mahlman (1969), it has been suggested that perhaps with a judiciously chosen averaging procedure, the role played by the eddies in the transport of species can be drastically reduced. This feasibility is amply demonstrated by Andrews and McIntyre (1978). By taking the Lagrangian mean, which is the average of a quantity at a "displaced" location, instead of the conventional Eulerian mean, which is taken with respect to fixed coordinate points, the eddy fluxes disappear entirely from the averaged transport equation. The transport is found to be caused by the advective Lagrangian mean meridional circulation. It should be noted, however, that the *eddy problem* is not eliminated with the use of this new formalism because, in principle, one needs to have a knowledge of the eddy displacement field to be able to perform the averaging in accordance with the definition of the Lagrangian mean. [See McIntyre (1980) for a discussion of the practical problems facing the application of the theory of Lagrangian mean to the tracer transport problem.] Nevertheless, the conceptual simplicity afforded by the theory of generalized Lagrangian mean has been very useful in the interpretation of results from three-dimensional models (Kida, 1977; Dunkerton, 1978; Matsuno and Nakamura, 1979; Hsu, 1980; Matsuno, 1980). Further development of the theory is eagerly awaited, especially in the practical area of interpretation of Lagrangian mean results with observational data (Danielsen, 1981). In the meantime, those working with data conventionally taken and analysed may prefer a more conventional Eulerian formulation.

A step in this direction is the development of the theory of the residual mean (or the so-called transformed Eulerian mean) circulation (Andrews and McIntyre, 1976; Boyd, 1976; Dunkerton, 1978; Edmon et al., 1980; Holton, 1980; Matsuno, 1980; Dunkerton et al., 1981; Palmer, 1981). The specific application of this theory to the problem of tracer transports is clearly discussed by Holton (1981). In this formulation a residual circulation, which is the difference between the Eulerian mean circulation and the eddy-induced circulation, is defined so that no eddy terms appear explicitly in the heat or species transport equation, provided that the species is conservative and the eddy field is steady and adiabatic. When these conditions are violated, as is the case in the real atmosphere, the full eddy problem still exists (in the sense that the eddy transport tensor is full[1]),

though now presumably the large cancellations between mean and eddy transports have been removed.

The presence of eddies, either explicitly or implicitly, in the averaged species equation is not the only obstacle in the formulation of a consistent 2-D model. A second (computational) difficulty arises because the mean quantities themselves are coupled. In particular, the diabatic mean flow velocities and the mean temperature field are present simultaneously in the energy equation in height or pressure coordinates. As a consequence, a consistent determination of even the residual (diabatic) circulation requires the simultaneous solution of three nonlinear prognostic[2], plus two linear diagnostic equations (e.g., the calculation of Holton and Wehrbein, 1980). Such an expensive undertaking appears to be excessive (especially for diagnostic purposes) because, as far as the species transport is concerned, only the mean diabatic meridional circulation is needed. One is tempted to simply drop the mean temperature time change and advection terms in the energy equation, thus decoupling the mean temperature field and the mean zonal velocity field from the mean diabatic meridional circulation. This is the approach taken by Dunkerton (1978). The procedure suggested by him is extremely simple: the diabatic heating rates directly give the diabatic vertical velocity, and the meridional velocity is then found by solving a nondivergent continuity equation. The results obtained by Dunkerton are encouraging, although many of the approximations involved are *ad hoc* and not easily justifiable. In isentropic coordinates, however, Dunkerton's procedure is the natural procedure for obtaining the mean circulation. This is due to the fact that the diabatic heating rate *is* the vertical velocity in the isentropic coordinate system, with no eddy fluxes or temperature advections appearing. The mean meridional diabatic velocity is found from this vertical velocity *via* the continuity equation. The mean zonal flow can be evolved, in time, by solving one linear prognostic equation. The mean temperature is obtainable diagnostically as the vertical derivative of the Montgomery stream function, whose meridional derivative is proportional to the mean zonal flow (see Section 5 for more details). Of course, even this much simpler

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[1] Some of the eddy transport components may turn out to be small, but no assessment of their magnitudes is available at this time. As we will discuss later, such an assessment is more easily made in isentropic coordinates (the diffusion tensor turns out to be dominated by one component in isentropic coordinates, while

[2] One prognostic equation, the northward momentum equation, can be approximated by a geostrophic diagnostic equation: see Section 5 for more detail.

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calculation is not needed if one is interested only in diagnosing the 2-D transport of species and not in the general zonally averaged circulation of the. atmosphere.

The zonal mean circulation calculated in isentropic coordinates turns out to be the mean diabatic circulation; the large eddy driven mean circulation that is present in height or pressure coordinates is mostly absent in the isentropic coordinate system. This mean circulation is thermally direct, in the sense that the motion is upward in regions of net diabatic heating and downward in the part of the atmosphere that cools. Furthermore, this direct circulation is found to be in the same direction as tracer transport trajectories.

In order for these results to be easily interpretable with conventionally taken data, we choose to present the formulation here in Eulerian zonal mean, though the theory can also be formulated in Lagrangian mean as well. In the Eulerian approach, eddy fluxes are inevitably present in the species transport equation. These eddy fluxes are expressed in terms of gradients of the mean species concentration using the procedure of linearization of the perturbation species equation. It is found that transient eddies disperse tracers from their mean path of advection by the diabatic circulation. If we assume that eddy events, arising from planetary and gravity waves propagated up from the troposphere, occur on a short enough time scale so that the process can be regarded as quasi-adiabatic, then the dispersion occurs predominantly in one direction only—horizontally along the isentropes. This dispersion process tends to smooth out the gradients of tracer concentration on the isentropes created by the mean diabatic advection. This situation is depicted schematically in Fig. 1, which shows a streamline for the mass circulation sloping downward and poleward at a steeper angle than the isentropes, with particles on the streamline dispersing in a direction parallel to the isentropes.

Though there is, as yet, no rigorous justification for such an assumption of separation of time scales, with the mean meridional mass circulation in isentropic coordinates determined by longer-term systematic diabatic effects, while the eddy dispersion is caused by shorter duration adiabatic processes, the results from such an assumption show a mechanism of tracer transport that seems to be consistent with the observed behavior of tracer movements (Danielsen et al., 1962; Danielsen, 1968; Newell, 1963).

As a wave disturbance propagates through the stratosphere, it displaces the isentropic surfaces up and down, relative to a constant-pressure surface. If the process is quasi-adiabatic, the fluid particles on an isentrope not only change their pressure in response to the wave disturbance, but also change their temperature; this occurs in such a way that the product of the temperature T and pressure p to the power
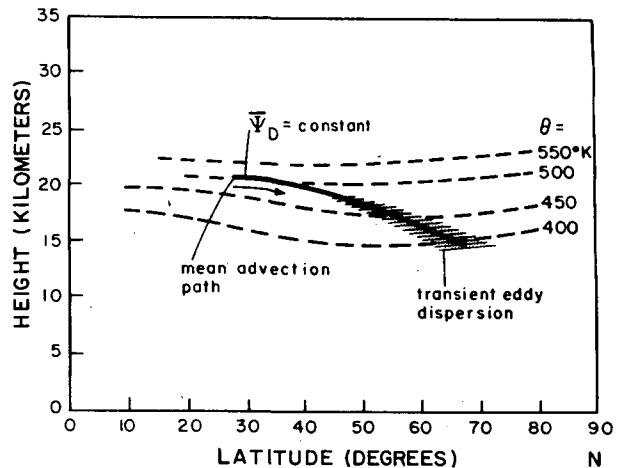

FIG. 1. Schematic diagram depicting an advection path (heavy line) by the diabatic mean circulation and dispersion by the transient eddies (thin lines with shading).

of R/c_p remains unchanged. Therefore, if one uses the potential temperature $\theta = T(p_{00}/p)^{R/c_p}$, as the vertical coordinate, then, with respect to such a coordinate system, there would be no vertical eddy displacements, though, in general, there would be horizontal eddy displacements associated with the passage of a wave disturbance. It is this ability of the isentropic coordinates to "follow" the vertical motion of adiabatic disturbances that endows these coordinates with a quasi-Lagrangian property. They are not true Lagrangian coordinates because the horizontal displacements are not followed. Nevertheless, the approximate absence of vertical eddy displacements reduces the number of eddy "diffusion" terms in the species transport equation by a factor of four, while at the same time retaining the advantage of an Eulerian system in being able to utilize conventional radiosonde data. Since radiosonde measurements are actually taken in the x, y, p system, with the height z inferred (instead of measured) from the temperature, it is as easy to infer the potential temperature as it is to deduce the height, with p and T measured by these instruments. Consequently, the isentropic coordinate system x, y, θ can easily be adopted and, in fact, has been generally used in meteorology, though to a lesser extent than the isobaric system (see Bleck, 1973; for a historical account of the rivalry between the proponents of isentropic versus isobaric coordinates). In the past, the isentropic coordinate system has been used to take advantage of the fact that for adiabatic motions the governing equations become two-dimensional (see Kasahara, 1974; Dutton, 1976). The often-encountered difficulties[3] occur

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[3] Similar difficulties also exist in pressure coordinates, with isobaric surfaces intersecting the lower boundary and with the frequent formation of "fronts" (see Dutton, 1976).
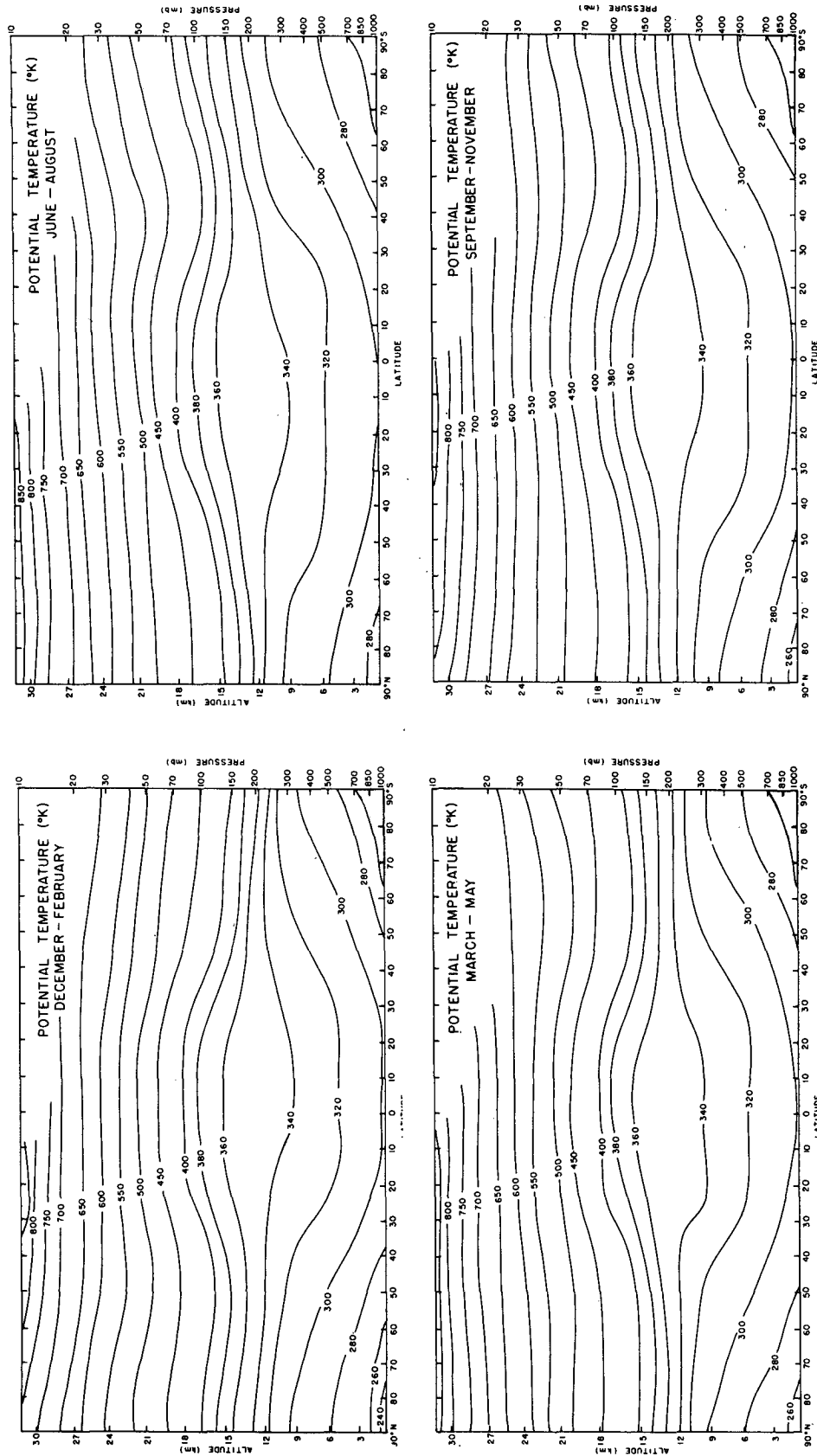
FIG. 2. Potential temperature distribution for each of the four seasons, taken from Newell *et al.* (1974).

near the Earth's surface, with isentropes intersecting the surface, and with the occasional occurrence of adiabatic layers, where θ ceases to be a monotonically increasing function of height. Though these technical difficulties can be overcome with hybrid coordinate systems such as that of Deaven (1976), Friend et al. (1977) and Uccellini et al. (1979)[4], the problem does not present itself in the stratosphere, where the co-ordinate system is adopted in the present application. As seen from Fig. 2 (taken from Newell et al., 1974), the mean potential temperature in the stratosphere is a monotonically increasing function of height and is almost horizontal over most latitudes for all sea-sons. For our purposes, we point out that the "tro-popause" ($\sim$350 K isentrope) is a nearly horizontal surface for all seasons, which separates the tropo-sphere, with its poleward-decreasing potential tem-perature, from the stratosphere. We shall be interested in the region above 350 K. The governing equations in isentropic coordinates are summarized in Appen-dix A. (Symbols are defined in Appendix F.)

2. The mean meridional mass circulation

Let X be the concentration per unit mass of a par-ticular species under consideration and S be the net rate of production, also per unit mass, of that species. The species equation can be written, in any coordi-nate system, as

$$\frac{d}{dt} X = S. \tag{2.1}$$

Let (u, v, w) be the velocities in the physical (i.e., geometrical) coordinate system (x, y, z) and $(u, v, \dot{\theta})$ be the corresponding velocities[5] in the isentropic system (x, y, θ). The substantial derivative can be specialized to either system as

$$\frac{d}{dt} = \left(\frac{\partial}{\partial t} + u \frac{\partial}{\partial x} + v \frac{\partial}{\partial y}\right)_z + w \frac{\partial}{\partial z} \tag{2.2}$$

or

$$\frac{d}{dt} = \left(\frac{\partial}{\partial t} + u \frac{\partial}{\partial x} + v \frac{\partial}{\partial y}\right)_\theta + \dot{\theta} \frac{\partial}{\partial \theta}. \tag{2.3}$$

The chief advantage of using the isentropic coordi-nate system is that the "vertical velocity" $\dot{\theta} \equiv (d/dt)\theta$ is directly determinable from a specification of the diabatic heating rate.

This relationship is expressed in the form of an energy equation, from the first law of thermodynam-ics:

$$\rho \frac{d}{dt} \ln\theta = \frac{q}{T}, \tag{2.4}$$

where ρ is the conventional density, mass per unit physical volume $dxdydz$, and q is the diabatic heating rate, also per unit physical volume, divided by c_p, the specific heat of air at constant pressure. Eq. (2.4) pro-vides a direct relationship between the diabatic heat-ing rate q, and the vertical "velocity," $\dot{\theta} \equiv (d/dt)\theta$, in isentropic coordinates. Let

$$\rho_\theta \equiv \rho \frac{\partial(x, y, z)}{\partial(x, y, \theta)} = \rho \frac{\partial z}{\partial \theta} \tag{2.5}$$

be the "density" in isentropic coordinates, i.e., mass per unit "volume" $dxdyd\theta$. Eq. (2.4) then implies that the "mass flow rate" in isentropic coordinates

$$W \equiv \rho_\theta \dot{\theta},$$

is related directly to the diabatic heating through

$$W = q/\Gamma. \tag{2.6}$$

In Eq. (2.6), Γ is the usual static stability parameter defined as in Holton (1972),

$$\Gamma \equiv \frac{T\partial\theta}{\theta\partial z} \approx \frac{\partial T}{\partial z} + \frac{g}{c_p}. \tag{2.7}$$

The simple form of (2.6) should be contrasted to the corresponding equation expressed in height (or sim-ilarly in pressure) coordinates, which is

$$\rho\left(\frac{\partial}{\partial t} + u \frac{\partial}{\partial x} + v \frac{\partial}{\partial y}\right)T + (\rho w)\Gamma = q. \tag{2.8}$$

That is, the diabatic heating is not only associated with a vertical mass flow ρw, but also with a hori-zontal temperature advection. Since the latter has to be solved as well, Eq. (2.8) is considerably more com-plicated than (2.6), in addition to the fact that the presence of quadratic terms in (2.8) introduces ad-ditional eddy heat fluxes when that equation is zon-ally averaged, though this latter problem can be cir-cumvented using the transformed Eulerian mean mentioned previously.

It should be pointed out, however, that our Eq. (2.6) is not as simple as its form seems to imply. This is because, strictly speaking, Γ is a function of the temperature, and so W cannot be deduced directly from q even if the latter is completely specified, un-less the temperature is solved as well. This compli-cation can be eliminated if the common practice of replacing Γ by its radiative equilibrium value

$$\Gamma^{(0)} \equiv \frac{T_e}{\theta_e} \frac{\partial\theta_e}{\partial z},$$

is adopted. This approximation is used in almost all current stratospheric dynamics calculations. Accord-ing to Holton (1975), this assumption appears to be necessary for the purpose of obtaining proper ap-proximate quadratic energy integrals and seems to be a satisfactory approximation in the stratosphere. We

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[4] Bleck (1978) found some problems relating to the stability of the scheme proposed by Uccellini et al. (1979). The other schemes mentioned above were found to perform satisfactorily.

[5] The horizontal velocities u and v in $(u, v, \dot{\theta})$ are here understood to be measured on constant θ-surfaces.

have examined this commonly used approximation in more detail in Appendix B, and found that it can be justified for the zonally averaged equation but not the eddy energy equation. That is,

$$\bar{W} = \frac{\bar{q}}{\Gamma^{(0)}} \qquad (2.9)$$

can be shown to be a good approximation to the zonal average of Eq. (2.6). The eddy energy equation will be treated separately later.

With the mean vertical flow $\bar{W}$ determined directly from the diabatic heating *via* Eq. (2.9), we now proceed to deduce the meridional circulation through the mass conservation equation, which is, in isentropic coordinates,

$$\frac{\partial}{\partial t} \rho_\theta + \frac{\partial}{\partial x} U + \frac{\partial}{\partial y} V + \frac{\partial}{\partial \theta} W = 0. \qquad (2.10)$$

The mass flow rates U, V and W are defined as

$$U \equiv \rho_\theta u, \quad V \equiv \rho_\theta v \cos\varphi, \quad W \equiv \rho_\theta \dot{\theta}. \qquad (2.11)$$

On a sphere with longitude λ and latitude φ, the following relationships have been used:

$$y \equiv a \sin\varphi, \quad dx = a \cos\varphi \, d\lambda. \qquad (2.12)$$

Since the mass conservation equation (2.10) is linear in the quantities involved, no eddy terms will be introduced by an Eulerian zonal average. This yields

$$\frac{\partial}{\partial t} \bar{\rho}_\theta + \frac{\partial}{\partial y} \bar{V} + \frac{\partial}{\partial \theta} \bar{W} = 0. \qquad (2.13)$$

We define a diabatic mean circulation ($\bar{V}_D$, $\bar{W}_D$) as

$$\bar{W}_D \equiv \frac{\bar{q}}{\Gamma^{(0)}}, \qquad (2.14)$$

$$\bar{V}_D \equiv -\int_a^y \frac{\partial}{\partial \theta} \left(\frac{\bar{q}}{\Gamma^{(0)}} \right) dy. \qquad (2.15)$$

From (2.9) one knows that $\bar{W}_D$ is the same as $\bar{W}$, but $\bar{V}_D$ is only the part of the meridional flow that is *directly driven* by the diabatic heating. There is an additional part arising from transient time changes in mean density. In general, one should write

$$\bar{V} = \bar{V}_T + \bar{V}_D. \qquad (2.16)$$

With $\bar{V}_D$ satisfying the divergence-free relation

$$\frac{\partial}{\partial y} \bar{V}_D + \frac{\partial}{\partial \theta} \bar{W}_D = 0, \qquad (2.17)$$

one can see from (2.13) that $\bar{V}_T$ is given by

$$\frac{\partial}{\partial t} \bar{\rho}_\theta + \frac{\partial}{\partial y} \bar{V}_T = 0. \qquad (2.18)$$

It is seen from (2.18) that $\bar{V}_T$ should be small in a long-term (e.g., seasonal) time average. It is further argued, in Appendix B, that $\bar{V}_T$, as given by (2.18),

is negligible compared to $\bar{V}_D$ even for subseasonal events. Consequently, we have

$$\bar{W} \approx \bar{W}_D \qquad (2.19)$$

and

$$\bar{V} \approx \bar{V}_D \qquad (2.20)$$

in isentropic coordinates. These approximations permit the mean meridional mass circulation to be deducible from q, in principle, using the simple relations in (2.14) and (2.15). However, in practice the determination of the diabatic heating rate q itself is not an easy matter. Here we shall first consider the simpler task of *diagnosing* the meridional circulation which is consistent with calculated radiative heating rates based on climatological mean conditions. In a later section, we will discuss the procedure for the more involved interactive calculations.

Using the more recent result of Dopplick (1979), who calculated the total radiative heating rate per unit mass, zonally and seasonally averaged, from various radiative and photochemical sources and sinks between 1000 and 10 mb, we have deduced the mean meridional mass circulation diagnostically. The procedure is described in Appendix C and the result is presented in Figs. 3 and 4. Because such diabatic heatings as latent heat release and sensible heating have not been included in the radiative calculation of Dopplick, the result for the diabatic circulation appears to be questionable in the troposphere. Therefore one should concentrate on the features in the stratosphere. Dopplick's calculation terminates at 10 mb. Above 10 mb, the radiative heating rate of Murgatroyd and Goody (1958) and Murgatroyd and Singleton (1961) is used, though it has a much coarser resolution. This circulation is represented by heavy dashed lines in Fig. 3. In Fig. 4, the streamfunction for the meridional mass circulation is depicted. Note the two-cell structure of the diabatic circulation in the lower stratosphere, with rising motion over the tropics and sinking motion over both poles. (As argued later in this paper, this feature of the meridional circulation essentially implies poleward and downward transport of tracers in a trajectory steeper than the isentropic surfaces.) In contrast, the mean circulation in the upper stratosphere and mesosphere consists of a single cell, with ascending motion over the summer pole and descending motion over the winter pole. These features of the mass circulation are consistent with those deduced by Dunkerton (1978). However, the physical interpretation is more direct in isentropic coordinates.

Equilibrium pressure coordinate

In Fig. 4, we have labeled the vertical coordinate also in terms of a pressure-like function defined as

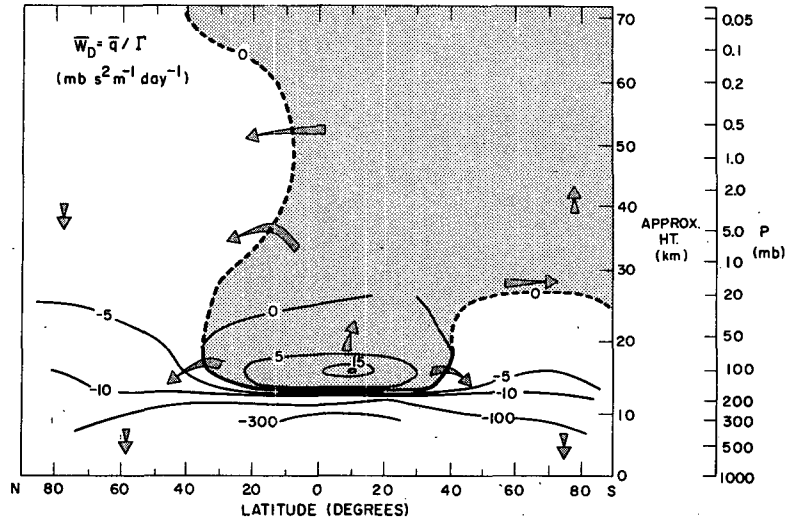$$p_e(\theta) \equiv p_{00} \left(\frac{\tilde{T}_e}{\theta} \right)^{1/\kappa}, \qquad (2.21)$$

FIG. 3. Zonally-averaged vertical mass flow rate of the diabatic circulation, $\bar{W}_D = \bar{q}/\Gamma^{(0)}$, for the northern winter season (December–February). The arrows indicate the direction of the meridional mass flow. The dashed lines refer to data taken from Murgatroyd and Singleton (1961).

with the inverse

$$\theta = \frac{gH}{R}\left(\frac{p_{00}}{p_e}\right)^{\kappa}, \tag{2.22}$$

where $H \equiv R\tilde{T}_e/g$ is the scale-height based on the background equilibrium temperature. The quantity $p_e(\theta)$ has an approximate correspondence with the pressure levels in an isobaric coordinate system. It is the pressure in an atmosphere whose temperature is the same as the globally averaged radiative equilibrium value $\tilde{T}_e$. Hence, p_e shall be called the equilibrium pressure. In existing models using pressure co-

ordinates it may be easier operationally to convert into the isentropic coordinate system based on $p_e(\theta)$ instead of θ.

3. The zonally averaged transport equation

In this section, the equation for the zonally averaged species concentration is derived. Eddy fluxes are inevitably present in the Eulerian average used here. These eddy terms cannot be consistently determined within the framework of two-dimensional theories. The situation is very similar to the well-known clo-
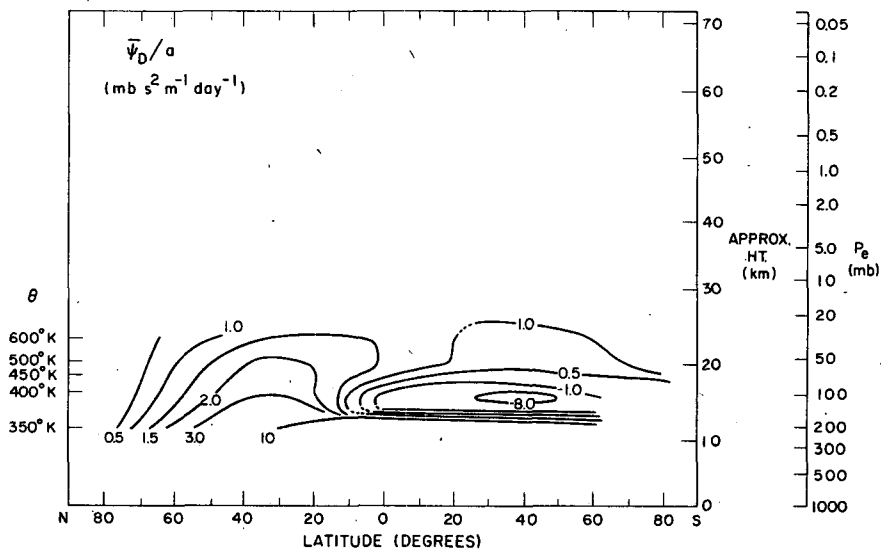

FIG. 4. Streamfunction for the diabatic meridional circulation in isentropic coordinates; the indicated height is approximate.

sure problem in theories of turbulence; no progress appears to be possible unless some *ad hoc* assumptions are adopted. We adopt here the *a priori* assumption that the perturbation quantities are determinable through the *procedure* of asymptotic expansion in powers of perturbation amplitude. This assumption is *a priori* because there is no sufficient reason to believe that in the real atmosphere the perturbation quantities are small compared to the mean. However, having adopted this procedure, the derivations that follow are *systematic* and no *ad hoc* parameters need be introduced. Nevertheless, one should be cautioned against placing too much confidence in the mathematical results obtained this way, as some of the behaviors of large-amplitude disturbances cannot be described in this manner (see Hsu, 1980; McIntyre, 1980).

With the aid of (2.10), the equation for mass conservation, one can rewrite the species equation (2.1) in a flux form as

$$\frac{\partial}{\partial t}(\rho_\theta X) + \frac{\partial}{\partial x}(UX) + \frac{\partial}{\partial y}(VX)$$
$$+ \frac{\partial}{\partial \theta}(WX) = \rho_\theta S, \quad (3.1)$$

which becomes, when the zonal average is taken,

$$\bar{\rho}_\theta \frac{\partial}{\partial t} \bar{X} + \bar{V} \frac{\partial}{\partial y} \bar{X} + \bar{W} \frac{\partial}{\partial \theta} \bar{X} + \frac{\partial}{\partial t} \overline{\rho_\theta' X'}$$
$$+ \frac{\partial}{\partial y}(\overline{V'X'}) + \frac{\partial}{\partial \theta}(\overline{W'X'}) = \overline{(\rho_\theta S)}. \quad (3.2)$$

In arriving at (3.2), the zonally averaged continuity equation (2.13) has been used. The mean quantities appearing in Eq. (3.2) have already been found to be

$$\bar{V} \approx \bar{V}_D \equiv -\int_a^y \frac{\partial}{\partial \theta}(\bar{q}/\Gamma^{(0)})dy, \quad (3.3a)$$

$$\bar{W} \approx \bar{W}_D \equiv \bar{q}/\Gamma^{(0)}. \quad (3.3b)$$

The mean "density" $\bar{\rho}_\theta$ will be assumed to be given approximately by the background radiative equilibrium value (see Appendix B), i.e.,

$$\bar{\rho}_\theta \approx \rho_\theta^{(0)} \approx \rho_{\theta_0}^{(0)}\left(\frac{\theta}{\theta_0}\right)^{-9/2}. \quad (3.3c)$$

[This assumption is not actually necessary, as the mean "density" can be calculated in 2-D models. However, this degree of accuracy does not seem warranted in the present context as far as the transport equation (3.2) is concerned.]

To close the system (3.2), the eddy fluxes are calculated using perturbation theory. The derivation is given in Appendix D. The results are summarized here. The transport of the mean species concentration $\bar{X}$ is governed by the following equation:

$$\bar{\rho}_\theta \frac{\partial}{\partial t} \bar{X} + \left(\bar{V}_D + \bar{V}_E - \frac{\partial}{\partial t}\overline{\rho_\theta'\eta'}\right)\frac{\partial}{\partial y}\bar{X} + \left(\bar{W}_D + \bar{W}_E\right.$$
$$- \frac{\partial}{\partial t}\overline{\rho_\theta'\phi'}\right)\frac{\partial}{\partial \theta}\bar{X} - \frac{\partial}{\partial y}\left(\bar{\rho}_\theta D_{yy}\frac{\partial}{\partial y}\bar{X}\right) - \frac{\partial}{\partial y}$$
$$\times \left(\bar{\rho}_\theta D_{y\theta}\frac{\partial}{\partial \theta}\bar{X}\right) - \frac{\partial}{\partial \theta}\left(\bar{\rho}_\theta D_{\theta y}\frac{\partial}{\partial y}\bar{X}\right) - \frac{\partial}{\partial \theta}\left(\bar{\rho}_\theta D_{\theta\theta}\frac{\partial}{\partial \theta}\bar{X}\right)$$
$$= \bar{\rho}_\theta\bar{S} - \frac{\partial}{\partial y}(\overline{V'\sigma'}) - \frac{\partial}{\partial \theta}(\overline{W'\sigma'}) - \overline{\left(\frac{\partial}{\partial t} + \bar{u}\frac{\partial}{\partial x}\right)\rho_\theta'\sigma'}$$
$$\equiv \bar{\rho}_\theta\bar{P}. \quad (3.4)$$

In (3.4), we have defined

$$\bar{V}_E \equiv -\frac{\partial}{\partial \theta}\left[\overline{\eta'W'} - \frac{1}{2}\bar{\rho}_\theta\frac{\partial}{\partial t}\overline{\eta'\phi'}\right], \quad (3.5a)$$

$$\bar{W}_E \equiv \frac{\partial}{\partial y}\left[\overline{\eta'W'} - \frac{1}{2}\bar{\rho}_\theta\frac{\partial}{\partial t}\overline{\eta'\phi'}\right], \quad (3.5b)$$

$$D_{yy} \equiv \frac{1}{2}\frac{\partial}{\partial t}\overline{\eta'\eta'},$$

$$D_{y\theta} \equiv \frac{1}{2}\frac{\partial}{\partial t}\overline{\eta'\phi'} = D_{\theta y},$$

$$D_{\theta\theta} \equiv \frac{1}{2}\frac{\partial}{\partial t}\overline{\phi'\phi'}. \quad (3.6)$$

It is seen that the advective transport of $\bar{X}$ consists of 1) the diabatically forced direct circulation ($\bar{V}_D$, $\bar{W}_D$), 2) an eddy-flux-induced flow ($\bar{V}_E$, $\bar{W}_E$) and 3) an additional advection caused by the correlation between "density" perturbation and particle displacements: $(-(\partial/\partial t)\overline{\rho_\theta'\eta'}, -(\partial/\partial t)\overline{\rho_\theta'\phi'})$. This last term is absent in isobaric coordinates. In height coordinates, however, a similar term should also exist but is often neglected. The "diffusive" terms, that involve the symmetric components of the diffusion tensor, are seen to arise entirely from transient disturbances, since the D's in (3.6) are all in the form of explicit time derivatives. It should be pointed out that even though the D-terms in Eq. (3.4) have the form of diffusion, they act physically as diffusion only if the following conditions are met (Matsuno, 1980):

$$D_{yy}D_{\theta\theta} \geq 0; \quad D_{yy}D_{\theta\theta} \geq D_{y\theta}^2.$$

However, for convenience, we will continue to use the word "diffusion" to refer to these D-terms. The D's would vanish in models that assume steady or periodic wave disturbances [e.g., Clark and Rogers (1978); Pyle and Rogers (1980b)]. On the right-hand side of Eq. (3.4), there are some new terms involving the zonal asymmetries in the *source* term:

$$-\frac{\partial}{\partial y}\overline{V'\sigma'} - \frac{\partial}{\partial \theta}\overline{W'\sigma'} - \overline{\left(\frac{\partial}{\partial t} + \bar{u}\frac{\partial}{\partial x}\right)\rho_\theta'\sigma'},$$

where σ' is defined from

$$\left(\frac{\partial}{\partial t} + \bar{u}\frac{\partial}{\partial x}\right)\sigma' = S'.$$

These terms are difficult to calculate, and have been considered only very recently. They are found by Tuck (1979) and Pyle and Rogers (1980b) to be important in isobaric coordinates. We will discuss them later in Section 4.

Eq. (3.4), then, is the equation governing the 2-D transport of a species. When more than one species are involved in a chemical reaction, each one of the species is transported in the manner described by Eq. (3.4). Their chemistry combines these individual equations through their source terms.

a. Adiabatic eddy model

In terms of the number of eddy transport terms that are present in the species transport equation, Eq. (3.4) is not simpler than the corresponding equation in either height or pressure coordinates under the transformed Eulerian interpretation. However (as detailed in Appendix E), many of the eddy transport terms appearing in Eq. (3.4) are actually small in isentropic coordinates and can be approximately neglected in first-order models. This is the case when the eddies are associated with disturbance events of short durations. These eddies can be regarded approximately as adiabatic if their residence time in the atmospheric layer of interest is shorter than the time it takes for the disturbance to gain or lose appreciable heat to the environment. Under such a circumstance, the disturbance conserves its potential temperature (i.e., $\dot{\theta} \approx 0$) and consequently no "vertical" particle displacement occurs in isentropic coordinates. Setting ϕ' and W' to be zero in Eq. (3.4) then leads to the following simplified equation:

$$\bar{\rho}_\theta \frac{\partial}{\partial x}\bar{X} + \left(\bar{V}_D - \frac{\partial}{\partial t}\overline{\rho_\theta'\eta'}\right)\frac{\partial}{\partial y}\bar{X} + \bar{W}_D\frac{\partial}{\partial \theta}\bar{X}$$
$$- \frac{\partial}{\partial y}\left(\bar{\rho}_\theta D_{yy}\frac{\partial}{\partial y}\bar{X}\right) = \bar{\rho}_\theta \bar{P}. \quad (3.7)$$

Note that of the four components of the diffusion tensor, only one, the horizontal diffusion along an isentropic surface arising from the horizontal gradient of mean concentration, remains. No comparable simplification can be achieved in height or pressure coordinates, unless the disturbances are assumed to be steady (or strictly periodic), which is difficult to justify for the real atmosphere.

It is shown in Appendix B that the horizontal eddy advection induced by density perturbations, $-(\partial/\partial t)\overline{\rho_\theta'\eta'}$, appearing in (3.7) is generally small compared to the mean advection $\bar{V}_D$ for both quasi-geostrophic waves and gravity waves, and so can be ignored in first-order models. Eq. (3.7) reduces to the following simple form:

$$\bar{\rho}_\theta \frac{\partial}{\partial t}\bar{X} + \bar{V}_D\frac{\partial}{\partial y}\bar{X} + \bar{W}_D\frac{\partial}{\partial \theta}\bar{X}$$
$$- \frac{\partial}{\partial y}\left(\bar{\rho}_\theta D_{yy}\frac{\partial}{\partial y}\bar{X}\right) = \bar{\rho}_\theta \bar{P}. \quad (3.8)$$

The transport of an inert tracer, as described by Eq. (3.8) with $\bar{P} = 0$, consists of advection by the diabatic mean mass circulation ($\bar{V}_D$, $\bar{W}_D$) and dispersion ("diffusion") by the transient eddies along isentropic surfaces.

In the absence of transient eddy dispersion (i.e., $D_{yy} = 0$), the equilibrium solution of Eq. (3.8) for inert tracers is (see Appendix C)

$$\bar{X} = F(\bar{\Psi}_D), \quad (3.9)$$

where $\bar{\Psi}_D$ is the streamfunction for the mean mass circulation (see Fig. 4). Eq. (3.9) implies that the lines of constant $\bar{X}$ would tend to coincide with those of $\bar{\Psi}_D$, in the absence of eddy dispersion. In the winter latitudes below 30 km, the mean vertical velocity on the isentropic surfaces is downward, since that region is radiatively cooled, while the velocity is upward in the tropical region due to radiative heating. The lines of constant $\bar{\Psi}_D$ for the thermally direct circulation ($\bar{V}_D$, $\bar{W}_D$) therefore slope at a greater downward angle than the isentropes. This feature of the direction of advection is evident in Fig. 4. The result in Eq. (3.9), therefore, implies that the tracer should be transported poleward and downward *at a greater slope than the surfaces of constant potential temperature*. The present simple and direct explanation of this feature of tracer transport should be compared with the rather complicated arguments in height or pressure coordinates (e.g., Hunt and Manabe, 1968; Wallace, 1978). This feature is *qualitatively* consistent with most observations of tracer concentration in this part of the atmosphere, though the steady-state slopes of tracer mass mixing ratios calculated here appear to be too steep near the polar region. Fig. 5, taken from Newell (1963), suggests a poleward and downward transport of ozone (which can be treated as a conservative tracer below 25 km) in northern latitudes, where the mean vertical velocity in pressure or height coordinates is observed to be *upward*. It is obvious that a model in height or pressure coordinates, with only advective transports by the observed mean Eulerian velocities, is not even qualitatively acceptable, while a model in isentropic coordinates, with only advective transports by the mean mass circulation, is qualitatively correct. It is on this basis that we speculate that the effects of eddy transports are of secondary importance away from the poles in the present formulation.

For quantitative purposes, however, the effect of dispersion by transient eddies has to be considered. The presence of dispersion (or "diffusion") tends to lessen the *gradient* of the tracer concentration created
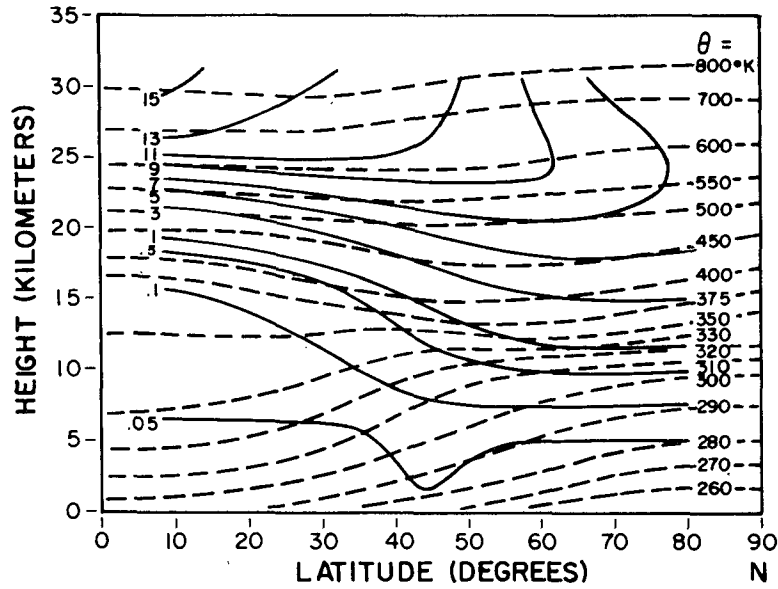
FIG. 5. The observed ozone mass mixing ratio (micrograms per gram of air) for January and February. Dashed lines are potential temperature surfaces (K). Taken from Newell (1963).

on the isentropic surfaces by the diabatic advective transport, making the slope of constant tracer mass mixing ratio less steep. These two competing effects of tracer transport, depicted schematically in Fig. 1, have previously been suggested by Mahlman et al. (1981).

b. Comparison with Dunkerton's Lagrangian mean transport equation

Dunkerton (1978) treated the case of steady conservative eddy fields. Under these same conditions our Eq. (3.4) reduces to

$$\bar{\rho}_\theta \frac{\partial}{\partial t} \bar{X} + (\bar{V}_D + \bar{V}_E) \frac{\partial}{\partial y} \bar{X}$$

$$+ (\bar{W}_D + \bar{W}_E) \frac{\partial}{\partial \theta} \bar{X} = 0, \quad (3.10)$$

for an inert tracer. Eq. (3.10) implies that the tracer is simply advected by the mean flow

$$\left[\frac{1}{\bar{\rho}_\theta} (\bar{V}_D + \bar{V}_E), \frac{1}{\bar{\rho}_\theta} (\bar{W}_D + \bar{W}_E) \right].$$

These advective velocities can be shown to be approximately equal to the Lagrangian mean flow of Andrews and McIntyre (1978), under the same assumptions used by Dunkerton (i.e., small-amplitude steady waves). That is, if one defines the generalized Lagrangian mean of θ to be

$$\bar{\theta}^L \equiv \frac{1}{2\pi} \int_0^{2\pi} \theta(x + \xi', y + \eta', \theta + \phi') d\lambda, \quad (3.11)$$

then by Taylor-expanding the integrand in (3.11), assuming small wave displacements, one finds

$$\bar{\rho}_\theta \bar{\theta}^L \approx \bar{\rho}_\theta \left[\bar{\theta} + \overline{\xi' \frac{\partial}{\partial x} \theta'} + \overline{\eta' \frac{\partial}{\partial y} \theta'} + \overline{\phi' \frac{\partial}{\partial \theta} \theta'} \right]$$

$$= \bar{\rho}_\theta \bar{\theta} + \overline{\rho'_\theta \theta'} + \frac{\partial}{\partial y} (\overline{\bar{\rho}_\theta \eta' \theta'}) + \frac{\partial}{\partial \theta} (\overline{\bar{\rho}_\theta \phi' \theta'}). \quad (3.12)$$

The first two terms on the right-hand side (rhs) of Eq. (3.12) are $\bar{\rho}_\theta \bar{\theta} = \bar{W}$. The last term vanishes for the steady waves under consideration. The third term is simply $\bar{W}_E$. Thus

$$\bar{\theta}^L \approx \frac{1}{\bar{\rho}_\theta} (\bar{W}_D + \bar{W}_E), \quad (3.13)$$

and similarly

$$\bar{v}^L \cos\varphi \approx \frac{1}{\bar{\rho}_\theta} (\bar{V}_D + \bar{V}_E). \quad (3.14)$$

We have now shown that the advective velocities in isentropic coordinates are approximately the Lagrangian mean velocities.

Dunkerton, by replacing the Lagrangian mean diabatic heating rate $\bar{q}^L$ by the Eulerian mean $\bar{q}$ in the energy equation, in effect ignored $\bar{W}_E$ compared to $\bar{W}_D$, with the final result that the advective Lagrangian mean flow is taken to be approximated by the Eulerian diabatic circulation, i.e.,

$$(\bar{v}^L \cos\varphi, \bar{\theta}^L) \approx \left(\frac{1}{\bar{\rho}_\theta} \bar{V}_D, \frac{1}{\bar{\rho}_\theta} \bar{W}_D \right). \quad (3.15)$$

In isentropic coordinates, the approximation in (3.15)

follows from the assumption that the *eddy* disturbances are quasi-adiabatic. The present arguments, then, constitute a somewhat more systematic justification of Dunkerton's procedure for *steady* adiabatic wave disturbances of small amplitude, but the use of isentropic coordinates is essential for our arguments.

Under the same assumption of steady waves, it can also be shown that the source terms on the rhs of the species transport equation can be interpreted as a Lagrangian zonal mean. In Appendix D (D18), we have shown that the source term on the rhs of Eq. (3.4) can be expressed as

$$\bar{\rho}_\theta \bar{P} = \bar{\rho}_\theta \left[\bar{S} + \overline{\xi' \frac{\partial}{\partial x} S'} + \overline{\eta' \frac{\partial}{\partial y} S'} + \overline{\phi' \frac{\partial}{\partial \theta} S'} \right]$$

$$- \frac{\partial}{\partial t} \left[\frac{\partial}{\partial y} (\overline{\bar{\rho}_\theta \eta' \sigma'}) + \frac{\partial}{\partial \theta} (\overline{\bar{\rho}_\theta \phi' \sigma'}) + \overline{\rho_\theta' \sigma'} \right],$$

which becomes, for steady waves

$$\bar{\rho}_\theta \bar{S} + \bar{\rho}_\theta \left[\overline{\xi' \frac{\partial}{\partial x} S'} + \overline{\eta' \frac{\partial}{\partial y} S'} + \overline{\phi' \frac{\partial}{\partial \theta} S'} \right]$$

$$= \bar{\rho}_\theta \bar{S}^L. \quad (3.16)$$

This result gives additional support that the Eulerian mean diabatic circulation $\left(\frac{1}{\rho_\theta} \bar{V}_D, \frac{1}{\rho_\theta} \bar{W}_D \right)$ approximates the Lagrangian mean transport ($\bar{v}^L \cos\varphi$, $\bar{\theta}^L$), as our Eq. (3.4) becomes, under the approximations used by Dunkerton and using (3.16),

$$\frac{\partial}{\partial t} \bar{X} + \frac{\bar{V}_D}{\bar{\rho}_\theta} \frac{\partial}{\partial y} \bar{X} + \frac{\bar{W}_D}{\bar{\rho}_\theta} \frac{\partial}{\partial \theta} \bar{X} = \bar{S}^L, \quad (3.17)$$

which has the same form as the corresponding species transport equation under the Lagrangian zonal averaging of Andrews and McIntyre (1978).

As pointed out by Matsuno (1980), the neglect of transient eddies by Dunkerton eliminates the dispersive transports which might otherwise be present. Numerical simulation of ozone transport in isobaric coordinates, using only the diabatic mean circulation of Dunkerton in the Eulerian formulation (Pyle and Rogers, 1980a), is found to poorly predict the observed ozone concentration.

c. Nonsteady disturbance field

As the assumption of steady (or periodic) wave fields cannot be justified in the real atmosphere, the transport equation should be more complicated than Eq. (3.17). However, as suggested earlier, maximum simplification can still be obtained if the adiabaticity assumption can be justified (see Appendix E). It is interesting to note here that, even in the presence of a nonsteady eddy disturbance field, the vertical Eu-

lerian mean velocity in isentropic coordinates is still the same as the Lagrangian mean, provided the eddy field is adiabatic and of small amplitude. That is, with $\bar{\theta}' \approx 0$, $\phi' \approx 0$, Eq. (3.12) implies

$$\bar{\theta}^L = \bar{\theta}. \quad (3.18)$$

Eq. (3.18) therefore illustrates the quasi-Lagrangian nature of the isentropic coordinate system. It is not a true Lagrangian system, as the horizontal eddy displacements are not followed. In fact, the Eulerian and Lagrangian mean horizontal advections differ by a transient eddy dispersion term:

$$\bar{v}^L \cos\varphi = \frac{1}{\bar{\rho}_\theta} \bar{V} + \frac{1}{\bar{\rho}_\theta} \frac{\partial}{\partial y} (\bar{\rho}_\theta D_{yy}). \quad (3.19)$$
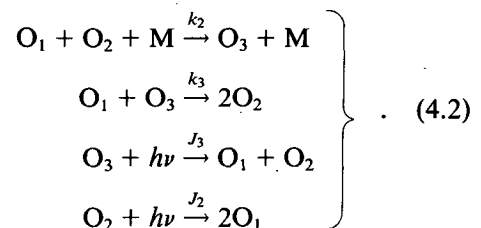
4. Chemically reacting species

We will now discuss the procedure for calculating the mean and eddy source terms in the species transport equation for a system of chemically reacting minor constituents. The relation between the eddy source fluxes and the dynamical diffusion coefficients will be illustrated. We will emphasize the point that within the framework of small amplitude perturbation theory, the diffusion coefficients are determined by the *dynamics* of the atmosphere and consequently should remain the same for all minor chemical species within the same atmosphere. The apparent discrepancy between this statement and the result of Pyle and Rogers (1980b), will be shown to be merely due to the different definitions used. The discussions in this section do not depend on the adiabatic eddy assumption adopted earlier.

A chemical scheme involving N participating minor species can be written in the form

$$\frac{d}{dt} X_i = S_i, \quad i = 1, 2, \ldots, N. \quad (4.1)$$

These equations are generally coupled because the production rate of the ith species usually depends not only on the concentration of that species, but also other species reacting with it. For illustrative purposes, let us consider an extremely simple example of the "classical" theory of ozone photochemistry involving the oxygen allotropes only [see Craig (1950)]:

$$\left. \begin{array}{c} O_1 + O_2 + M \xrightarrow{k_2} O_3 + M \\ O_1 + O_3 \xrightarrow{k_3} 2O_2 \\ O_3 + h\nu \xrightarrow{J_3} O_1 + O_2 \\ O_2 + h\nu \xrightarrow{J_2} 2O_1 \end{array} \right\} . \quad (4.2)$$

From this scheme, one can easily deduce that the production rate of the ozone, for example, is

$$\frac{d}{dt} X_{O_3} = S_{O_3}$$

$$= k_2 X_{O_1} X_{O_2} X_M - k_3 X_{O_1} X_{O_3} - J_3 X_{O_3}. \quad (4.3)$$

(The reaction and photodissociation rates are appropriately scaled by ρ for the *mass* mixing ratio used.) Similarly for atomic oxygen,

$$\frac{d}{dt} X_{O_1} = S_{O_1} = 2 J_2 X_{O_2} + J_3 X_{O_3}$$

$$- k_2 X_{O_1} X_{O_2} X_M - k_3 X_{O_1} X_{O_3}. \quad (4.4)$$

Since O_2 and M (air) are not minor species, their concentrations are assumed to be known and so can be absorbed into the rate "constants", which are also assumed to be known. Writing X_1 for X_{O_1} and X_2 for X_{O_3}, the reactions can be put into the form of (4.1) as

$$\frac{d}{dt} X_1 = S_1 = 2\hat{J}_2 + J_3 X_2 - \hat{k}_2 X_1 - k_3 X_1 X_2, \quad (4.5a)$$

$$\frac{d}{dt} X_2 = S_2 = \hat{k}_2 X_1 - k_3 X_1 X_2 - J_3 X_2. \quad (4.5b)$$

This simple example illustrates how the species equations are coupled through their source terms. (Due to its short lifetime, the radical species O_1 is often assumed to be in photochemical equilibrium. Since our purpose here is to illustrate the formal procedure for coupled equations, this approximation is not introduced at this point.)

Returning to the general case of N minor species, one can easily see that the perturbation production rate S_i' is of the form

$$S_i' = \sum_{j=1}^{N} A_{ij} X_j',$$

$$i = 1, 2, \ldots, N, \quad A_{ij} \equiv \left. \frac{\partial S_i}{\partial X_j} \right|_{X_k = \bar{X}_k}, \quad (4.6)$$

to first order in perturbation amplitudes. The A_{ij}'s are zonal mean quantities and in general are rational algebraic functions of the mean species concentrations. [It is assumed here that the rate "constants" are mean quantities. This is in general not true because some of the reaction rates are sensitive to *local* temperature change. Modifications to (4.6) due to such temperature fluxes will be considered at the end of this section.] For the example in Eq. (4.5), linearization gives

$$S_1' = J_3 X_2' - \hat{k}_2 X_1' - k_3 \bar{X}_1 X_2' - k_3 \bar{X}_2 X_1',$$

$$S_2' = \hat{k}_2 X_1' - J_3 X_2' - k_3 \bar{X}_1 X_2' - k_3 \bar{X}_2 X_1',$$

and hence

$$A_{11} = -\hat{k}_2 - k_3 \bar{X}_2, \quad A_{12} = J_3 - k_3 \bar{X}_1,$$

$$A_{21} = \hat{k}_2 - k_3 \bar{X}_2, \quad A_{22} = -J_3 - k_3 \bar{X}_1.$$

The expression in (4.6) for S_i' will be needed for the calculation of the eddy flux

$$\overline{\mathbf{V}'X_i'} \equiv (\overline{V'X_i'}, \overline{W'X_i'}),$$

since the perturbation concentration X_i' is given by (see Appendix D)

$$D_0 X_i' = -v' \cos\varphi \frac{\partial}{\partial y} \bar{X}_i - \dot{\theta}' \frac{\partial}{\partial \theta} \bar{X}_i + S_i', \quad (4.7)$$

where $D_0 \equiv \frac{\partial}{\partial t} + \bar{u} \frac{\partial}{\partial x}$. It was found that the solution to (4.7) is

$$X_i' = -\eta' \frac{\partial}{\partial y} \bar{X}_i - \phi' \frac{\partial}{\partial \theta} \bar{X}_i + \sigma', \quad (4.8)$$

with the displacement fields defined by

$$\eta' = D_0^{-1} v' \cos\varphi,$$

$$\phi' = D_0^{-1} \dot{\theta}',$$

$$\sigma_i' = D_0^{-1} S_i'. \quad (4.9)$$

The inverse D_0^{-1} of the operator D_0 is defined formally according to:

$$D_0^{-1} D_0 = 1.$$

Since the operator D_0 is the time derivative following the mean zonal flow, the inverse operator D_0^{-1} is then the time integration along the mean zonal flow. Substituting (4.8) into the expression for eddy fluxes, we get

$$\overline{\mathbf{V}'X_i'} = -\bar{\rho}_\theta \mathbf{K} \cdot \nabla \bar{X}_i + \overline{\mathbf{V}'\sigma'}. \quad (4.10)$$

The **K**-matrix

$$\mathbf{K} = \begin{bmatrix} K_{yy} & K_{y\theta} \\ K_{\theta y} & K_{\theta\theta} \end{bmatrix}$$

is the so-called eddy transport tensor whose elements have been defined in (D.8) in Appendix D. (For adiabatic eddies, $K_{y\theta} = K_{\theta y} = K_{\theta\theta} = 0$. We have used D_{yy} to stand for K_{yy} in Section 3.) It is important to point out that the elements of the **K**-matrix are expressible exclusively in terms of η' and ϕ', the *air* displacements in the horizontal and vertical directions, respectively, and therefore should be independent of the concentration of the *minor* chemical constituents. Once parameterized and validated for one species, the same set of K's should, in principle at least, be applicable to other species. Pyle and Rogers (1980b) defined their **K**-matrix to include the source term [the last term in (4.10)] in it as well. Therefore, it is not surprising that they concluded that their **K**-matrix depends strongly on species concentration.

Since the perturbations in the species production rate ultimately arise from the eddy air motions, one should, in principle, and with a knowledge of the chemical scheme involved, be able to reduce the eddy terms in the source fluxes to dynamical eddy dis-

placements. Using (4.6) and (4.9), we find that the last term in (4.10) can be expressed as

$$\overline{V'\sigma_i'} = \sum_{j=1}^{N} A_{ij} \overline{V'D_0^{-1}X_j'} .$$

With (4.8) for X_j', this further becomes

$$\overline{V\sigma_i'} = -\bar{\rho}_\theta \sum_{j=1}^{N} A_{ij} \mathbf{K}^{(1)} \cdot \nabla \bar{X}_j + \sum_{j=1}^{N} A_{ij} \overline{V'D_0^{-1}\sigma_j'} . \quad (4.11)$$

The $\mathbf{K}^{(1)}$ matrix in (4.11) is defined such that its elements are

$$\left. \begin{array}{ll} K_{yy}^{(1)} \equiv \dfrac{1}{\bar{\rho}_\theta} \overline{V'D_0^{-1}\eta'}, & K_{\theta\theta}^{(1)} \equiv \dfrac{1}{\bar{\rho}_\theta} \overline{W'D_0^{-1}\phi'} \\[2mm] K_{y\theta}^{(1)} \equiv \dfrac{1}{\bar{\rho}_\theta} \overline{V'D_0^{-1}\phi'}, & K_{\theta y}^{(1)} \equiv \dfrac{1}{\bar{\rho}_\theta} \overline{W'D_0^{-1}\eta'} \end{array} \right\} . \quad (4.12)$$

This procedure can be repeated indefinitely. The final result can be written in a compact form if we define $\mathbf{A}$ to be the $N \times N$ square matrix whose (ij)th element is A_{ij}, σ' and $\bar{X}$ each to be the $1 \times N$ column matrix whose ith element is σ_i' and $\bar{X}_i$, respectively. This leads to the following expression for $\overline{V'\sigma_i'}$:

$$\overline{V'\sigma'} = -\bar{\rho}_\theta [\mathbf{A} \cdot (\mathbf{K}^{(1)} \cdot \nabla)\bar{X} + \mathbf{A} \cdot \mathbf{A} \cdot (\mathbf{K}^{(2)} \cdot \nabla)\bar{X}$$

$$+ \mathbf{A} \cdot \mathbf{A} \cdot \mathbf{A} \cdot (\mathbf{K}^{(3)} \cdot \nabla)\bar{X} + \cdots]$$

$$= -\bar{\rho}_\theta \sum_{n=1}^{\infty} (\mathbf{A})^{(n)} \cdot (\mathbf{K}^{(n)} \cdot \nabla)\bar{X}, \quad (4.13)$$

and hence (4.10) becomes

$$\overline{V'X'} = -\bar{\rho}_\theta \sum_{n=0}^{\infty} (\mathbf{A})^n (\mathbf{K}^{(n)} \cdot \nabla)\bar{X}. \quad (4.14)$$

In these expressions, we have defined $\mathbf{K}^{(n)}$ to be the 2×2 matrix whose elements are defined in the same way as those of $\mathbf{K}^{(1)}$, except with D_0^{-1} replaced by $(D_0^{-1})^n$. Also $\mathbf{K}^{(0)} \equiv \mathbf{K}$ and $(\mathbf{A})^0 \equiv \mathbf{I}$. The factor in front of $\nabla \bar{X}$ should reduce to Pyle and Rogers' $\mathbf{K}$-matrix for stationary planetary waves.

Provided the series converges, the expression in (4.14) then generalizes the K-theory for conservative tracers to chemically reacting minor species participating in an N-reaction scheme. It expresses the eddy fluxes of each species in terms of the mean gradients of the participating species. It clearly separates the coefficients into a factor $\mathbf{K}^{(n)}$ that depends on the dynamics of eddy air displacements only (and so is independent of the chemistry) and another part $(\mathbf{A})^n$ which is a function of the *mean* species concentrations and rate constants only. Given the chemical scheme, $(\mathbf{A})^n$ is, in principle, known. Given the dynamics, $\mathbf{K}^{(n)}$ can either be calculated or parameterized. This is done once for all species within the same atmosphere.

For practical calculations, it seems that (4.14) can be severely truncated, provided that the chemical life-

time of the species involved is not too short [see Pyle and Rogers (1980b) for the case of steady planetary-wave eddies]. (When the chemical lifetime is short, a separate photochemical equilibrium approximation can be used instead.) The rationale is that the operator D_0^{-1} appearing in $\mathbf{K}^{(n)}$ is an integration operator, and its acting on a perturbation displacement field would have the effect of smoothing the latter. It appears, then, that each successive application of the operator would tend to reduce the magnitude of the eddy term further. For a first-order model, it is recommended that the series in Eq. (4.13) be truncated beyond the $n = 0$ and $n = 1$ terms. This retains the first order effect of eddy transport on chemistry:

$$\overline{V'X_i'} \approx -\bar{\rho}_\theta \mathbf{K} \cdot \nabla \bar{X}_i - \bar{\rho}_\theta \sum_{j=1}^{N} A_{ij} \mathbf{K}^{(1)} \cdot \nabla \bar{X}_j ,$$

$$i = 1, 2, \ldots, N. \quad (4.15)$$

For adiabatic eddies, (4.15) can be simplified further because the only nonvanishing element in $\mathbf{K}$ is K_{yy} $= \dfrac{1}{2} \dfrac{\partial}{\partial t} \overline{\eta'\eta'} \equiv D_{yy}$, and the only nonvanishing element in $\mathbf{K}^{(1)}$ is

$$K_{yy}^{(1)} = \frac{1}{\bar{\rho}_\theta} \overline{V'D_0^{-1}\eta'} \doteq \overline{\eta'\eta'} - \frac{\partial}{\partial t} \overline{\eta'D_0^{-1}\eta'}$$

$$= \overline{\eta'\eta'} \equiv D_{yy}^{(1)}.$$

So

$$\overline{V'X_i'} \approx -\bar{\rho}_\theta D_{yy} \frac{\partial}{\partial y} \bar{X}_i - \bar{\rho}_\theta D_{yy}^{(1)} \sum_{j=1}^{N} A_{ij} \frac{\partial}{\partial y} \bar{X}_j$$

and

$$\overline{W'X_i'} \approx 0. \quad (4.16)$$

With the parameterization of one function $D_{yy}^{(1)}$ in addition to the "diffusion" coefficient D_{yy} needed for tracers, all chemically reacting species can be considered in this manner. The fact that the two parameters are in theory related, i.e.,

$$D_{yy} = \frac{\partial}{\partial t} D_{yy}^{(1)},$$

may help simplify the process of parameterization, but in statistical approximations D_{yy} and $D_{yy}^{(1)}$ should be treated as two distinct functions, as the processes dominating the statistics of a function may be very different from the processes dominating the statistics of its derivative.

We now turn to the calculation of the source term in the right-hand side of the species transport equation (3.4):

$$\bar{\rho}_\theta \bar{P}_i \equiv \bar{\rho}_\theta \bar{S}_i - \left(\frac{\partial}{\partial t} + \bar{u} \frac{\partial}{\partial x} \right) \overline{\rho_\theta' \cdot \sigma_i'}$$

$$- \frac{\partial}{\partial y} (\overline{V'\sigma_i'}) - \frac{\partial}{\partial \theta} (\overline{W'\sigma_i'}). \quad (4.17)$$

Despite its appearance, the first term on the right-hand side of (4.17) contains some important eddy terms. For the simple example used earlier, we have [from (4.5)]

$$\bar{S}_2 = -J_3\bar{X}_2 + \hat{k}_2\bar{X}_1 - k_3\bar{X}_1\bar{X}_2 - k_3\overline{X_1'X_2'};$$

the last term, $\bar{S}_2{}^E \equiv -k_3\overline{X_1'X_2'}$, depends on the eddy dynamics. In general, with only quadratic terms in eddy correlation retained, the eddy terms in $\bar{S}_i$, denoted by $\bar{S}_i{}^E$, can be written in the form

$$\bar{S}_i{}^E = \sum_{j=1}^{N}\sum_{k=1}^{N} B_{ijk}\overline{X_j'X_k'}$$

$$\doteq \sum_{j=1}^{N}\sum_{k=1}^{N} B_{ijk}\left[\overline{\eta'\eta'}\frac{\partial}{\partial y}\bar{X}_j\frac{\partial}{\partial y}\bar{X}_k + \overline{\phi'\phi'}\frac{\partial}{\partial\theta}\bar{X}_j\frac{\partial}{\partial\theta}\bar{X}_k\right.$$
$$\left. + \overline{\eta'\phi'}\left(\frac{\partial}{\partial y}\bar{X}_j\frac{\partial}{\partial\theta}\bar{X}_k + \frac{\partial}{\partial\theta}\bar{X}_j\frac{\partial}{\partial y}\bar{X}_k\right)\right]. \quad (4.18)$$

Again, severe truncation has been used for the source terms. In (4.18), B_{ijk} is known from chemistry, and $E_{yy} \equiv \overline{\eta'\eta'} = D_{yy}{}^{(1)}$, $E_{\theta\theta} \equiv \overline{\phi'\phi'}$ and $E_{y\theta} \equiv \overline{\eta'\phi'}$ need to be parameterized (but independently of chemistry). For adiabatic waves, only E_{yy} is nonzero, and so

$$\bar{S}_i{}^E \approx D_{yy}{}^{(1)}\sum_{j=1}^{N}\sum_{k=1}^{N} B_{ijk}\frac{\partial}{\partial y}\bar{X}_j\frac{\partial}{\partial y}\bar{X}_k. \quad (4.19)$$

The last two terms in (4.17) have already been treated. The result is

$$-\frac{\partial}{\partial y}(\overline{V'\sigma_i'}) - \frac{\partial}{\partial\theta}(\overline{W'\sigma_i'})$$

$$\approx \nabla\cdot[\bar{\rho}_\theta\sum_{j=1}^{N} A_{ij}\mathbf{K}^{(1)}\cdot\nabla\bar{X}_j]. \quad (4.20)$$

The remaining term $-\left(\dfrac{\partial}{\partial t} + \bar{u}\dfrac{\partial}{\partial x}\right)\overline{\rho_\theta'\cdot\sigma_i'}$ is new (it disappears in isobaric coordinates), but can be treated the same way, giving

$$-\overline{\left(\frac{\partial}{\partial t} + \bar{u}\frac{\partial}{\partial x}\right)\rho_\theta'\cdot\sigma_i'}$$

$$\approx \sum_{j=1}^{N} A_{ij}\left[F_y\frac{\partial}{\partial y}\bar{X}_j + F_\theta\frac{\partial}{\partial\theta}\bar{X}_j\right], \quad (4.21)$$

where $F_y \equiv \overline{D_0\rho_\theta'\cdot D_0^{-1}\eta'}$ and $F_\theta \equiv \overline{D_0\rho_\theta'\cdot D_0^{-1}\phi'}$. For adiabatic waves, only F_y is nonzero. However, as we have argued previously, the density perturbation is small for geostrophic planetary waves. For gravity waves, ρ_θ' and η' are 90° out of phase, and so F_y is small even for unstable waves. Thus, we will neglect the term in (4.21). All the eddy correlation terms in the mean production rate in (4.17) have now been considered. The result is complicated, but in isentropic coordinates the number of terms can be re-

duced by a factor of four when the quasi-adiabatic approximation is adopted. It is instructive here to synthesize all the results up to this point and apply them to a simple example.

a. An example

We use, again, the simple example considered earlier in (4.2) involving oxygen allotropes. (This example should only be used for illustrating the procedure involved. The photochemical equilibrium assumption for the radical oxygen should be applied in practice from the beginning to simplify the equations involved.) The zonally averaged species equations are [from (4.5)]:

$$\frac{\partial}{\partial t}\bar{X}_1 + \frac{\bar{V}_D}{\bar{\rho}_\theta}\frac{\partial}{\partial y}\bar{X}_1 + \frac{\bar{W}_D}{\bar{\rho}_\theta}\frac{\partial}{\partial\theta}\bar{X}_1 - \frac{1}{\bar{\rho}_\theta}\frac{\partial}{\partial y}\left(\bar{\rho}_\theta D_{yy}\frac{\partial}{\partial y}\bar{X}_1\right)$$

$$= \bar{P}_1 = 2\hat{J}_2 + J_3\bar{X}_2 - \hat{k}_2\bar{X}_1 - k_3\bar{X}_1\bar{X}_2$$

$$- D_{yy}{}^{(1)}k_3\frac{\partial}{\partial y}\bar{X}_1\frac{\partial}{\partial y}\bar{X}_2 + \frac{1}{\bar{\rho}_\theta}\frac{\partial}{\partial y}\left\{\bar{\rho}_\theta D_{yy}{}^{(1)}\right.$$

$$\times\left[(-\hat{k}_2 - k_3\bar{X}_2)\frac{\partial}{\partial y}\bar{X}_1 + (J_3 - k_3\bar{X}_1)\frac{\partial}{\partial y}\bar{X}_2\right]\right\} \quad (4.22)$$

and

$$\frac{\partial}{\partial t}\bar{X}_2 + \frac{\bar{V}_D}{\bar{\rho}_\theta}\frac{\partial}{\partial y}\bar{X}_2 + \frac{\bar{W}_D}{\bar{\rho}_\theta}\frac{\partial}{\partial\theta}\bar{X}_2 - \frac{1}{\bar{\rho}_\theta}\frac{\partial}{\partial y}\left(\bar{\rho}_\theta D_{yy}\frac{\partial}{\partial y}\bar{X}_2\right)$$

$$= \bar{P}_2 = \hat{k}_2\bar{X}_1 - k_3\bar{X}_1\bar{X}_2 - J_3\bar{X}_2 - D_{yy}{}^{(1)}k_3\frac{\partial}{\partial y}\bar{X}_1$$

$$\times\frac{\partial}{\partial y}\bar{X}_2 + \frac{1}{\bar{\rho}_\theta}\frac{\partial}{\partial y}\left\{\bar{\rho}_\theta D_{yy}{}^{(1)}\left[(\hat{k}_2 - k_3\bar{X}_2)\frac{\partial}{\partial y}\bar{X}_1\right.\right.$$

$$\left.\left. + (-J_3 - k_3\bar{X}_1)\frac{\partial}{\partial y}\bar{X}_2\right]\right\}. \quad (4.23)$$

Eqs. (4.22) and (4.23) are a highly coupled set, but they can, in principle, be solved when a parameterization of D_{yy} and $D_{yy}{}^{(1)}$ is adopted. (Furthermore, for this particular example, the equations can be simplified greatly if the photochemical equilibrium assumption for O_1 mentioned earlier is adopted in the beginning.) The extra terms in $\bar{P}_1$ and $\bar{P}_2$ arising from eddy dynamics [terms multiplying $D_{yy}{}^{(1)}$] have not been commonly treated in photochemical calculations. They are expected to play an important role in determining the mean concentrations $\bar{X}_1$ and $\bar{X}_2$. Thus, there is considerable room for improving the prediction by treating the eddy dynamics more consistently in the source terms, without having to adopt more sophisticated chemical schemes.

b. Modification due to eddy fluctuations in the reaction rates

The photochemical reaction rates, the k's and J's in Eq. (4.5), have previously been taken to be zonal

mean quantities. Here, we shall consider the modification to our formulation arising from eddy fluctuations in the reaction rates. These terms have not been treated previously in existing 2-D models, but their contribution needs to be assessed in the future.

Suppose that the reaction rates are predominantly sensitive to local temperature T. Then the perturbation source term S_i' should have the following form [cf. Eq. (4.6)] according to small amplitude perturbation theory:

$$S_i' = \sum_{j=1}^{N} A_{ij}X_j' + B_iT', \qquad (4.24)$$

where A_{ij} is the same as before, i.e.,

$$A_{ij} \equiv \frac{\partial S_i}{\partial X_j}\bigg|_{X_k=\bar{X}_k, T=\bar{T},}$$

and the extra term B_iT' is due to local temperature fluctuation, with

$$B_i \equiv \frac{\partial S_i}{\partial T}\bigg|_{X_k=\bar{X}_k, T=\bar{T}.}$$

With the extra temperature variation term in (4.24), the eddy flux of the species is seen to be [cf. Eq. (4.14)]

$$\overline{V'X_i'} = -\bar{\rho}_\theta[\sum_{n=0}^{\infty} (\mathbf{A})^n \cdot (\mathbf{K}^{(n)} \cdot \nabla)\bar{\mathbf{X}}]_i$$

$$+ B_i\overline{V'D_0^{-1}T'}. \qquad (4.25)$$

The modification, the second term on the rhs of Eq. (4.25) is the product of a term B_i that depends on the chemistry of the zonally averaged quantities with zonally averaged temperature, and a part $\overline{V'D_0^{-1}T'}$ that depends on the dynamics of atmospheric air perturbations. We find that (4.16) now becomes

$$\overline{V'X_i'} \approx -\bar{\rho}_\theta D_{yy}\frac{\partial}{\partial y}\bar{X}_i - \bar{\rho}_\theta D_{yy}^{(1)}$$

$$\times \sum_{j=1}^{N} A_{ij}\frac{\partial}{\partial y}X_j + B_iG_y \Bigg\} \qquad (4.26)$$

$$\overline{W'X_i'} \approx 0$$

One additional parameter,

$$G_y \equiv \overline{V'D_0^{-1}T'},$$

has been introduced in (4.26) and has to be parameterized along with D_{yy} and $D_{yy}^{(1)}$.

5. Interactive 2-D model of radiation and dynamics

Qualitatively, the structure of the mean diabatic circulation deduced in Section 2 (see Figs. 3 and 4) is seen to be consistent, from angular momentum considerations, with the observed zonal wind distribution without the need for large eddy momentum fluxes. Below 30 km, the two-cell meridional circulation should produce westerly zonal winds in both the winter and summer midlatitudes because the fluid from the equatorial region carries excess (and hence westerly) angular momentum when it is transported to higher latitudes by the diabatic circulation. Above 30 km, the one-cell pole-to-pole diabatic circulation is seen to produce easterlies in the summer hemisphere and westerlies in the winter hemisphere by the same angular momentum consideration. This situation should be contrasted with the Eulerian zonal mean circulation in pressure or height coordinates, where in the high latitude lower stratosphere, the presence of an indirect mean meridional circulation in a westerly region would violate the angular momentum principle unless a large eddy momentum flux is invoked. It is, therefore, apparent that it is conceptually simpler to study zonal mean circulations in isentropic coordinates. Furthermore, since the eddies in this formulation are relegated to a secondary role, the model is less critically dependent on the particular parameterization adopted for the eddy fluxes. This property of the present 2-D model is important when interaction between radiation and dynamics is allowed.

Our previous quantitative calculations of the mean meridional circulation are only diagnostic because we have regarded the radiative heating rates as given. In fact, we have used the heating rates calculated by previous authors, using climatological distributions of temperature field and ozone concentration. In an interactive model, these quantities should be internally calculated. It has, so far, not been done consistently in two-dimensional models. With the simplification obtained in our present formulation, the zonally averaged general circulation and temperature distribution in the stratosphere and mesosphere can be economically calculated and coupled to the radiative and photochemical calculations. The procedure for such calculations will be briefly outlined here; the actual numerical computation is beyond the scope of this paper.

Starting with an initial distribution of source and sinks for the radiative heating q, one determines, diagnostically, the initial meridional circulation in the manner described in Section 2, i.e., from the following two approximate equations

$$\bar{W} = \bar{q}/\Gamma^{(0)}, \qquad (5.1)$$

$$\frac{\partial}{\partial y}(\bar{V}) + \frac{\partial}{\partial \theta}\bar{W} = 0. \qquad (5.2)$$

To step forward in time, the averaged zonal momentum equation is used:

$$\frac{\partial}{\partial t}\bar{U} + \frac{\partial}{\partial y}\left(\frac{1}{\bar{\rho}_\theta}\bar{U}\bar{V}\right) + \frac{\partial}{\partial \theta}\left(\frac{1}{\bar{\rho}_\theta}\bar{U}\bar{W}\right)$$

$$- f\bar{V}/\cos\varphi = \mathcal{P} + \mathcal{M} + \mathcal{F}. \qquad (5.3)$$

This equation is obtained by multiplying the zonal momentum equation (A.3) by ρ_θ and taking the zonal average. Here $\mathcal{F}$ is the eddy damping term and we have defined the eddy pressure torque $\mathcal{P}$ to be

$$\mathcal{P} \equiv -\overline{\rho_\theta \frac{\partial}{\partial x} \Phi} \qquad (5.4)$$

and the eddy momentum flux convergence $\mathcal{M}$ to be

$$\mathcal{M} \equiv \frac{\partial}{\partial y} (\overline{\rho_\theta u^* v^*} \cos\varphi) + \frac{\partial}{\partial \theta} (\overline{\rho_\theta u^* \dot{\theta}^*}), \quad (5.5)$$

where, following the notation of Gallimore and Johnson (1981a), the asterisk denotes deviation from the mass-weighted zonal mean, i.e.,

$$h^* \equiv h - \overline{\rho_\theta h}/\overline{\rho_\theta}. \qquad (5.6)$$

The momentum flux terms in Eq. (5.5) can be interpreted as mass averaged momentum fluxes. In isobaric coordinates, the "density" is a constant, $\mathcal{M}$ is then the regular eddy momentum flux convergence and $\mathcal{P}$ vanishes. In isentropic coordinates, however, the pressure torque may be an important mechanism for eddy forcing of the mean flow (see Gallimore and Johnson, 1981b). We will return in a moment to a discussion of the parameterization of these eddy terms.

[Incidentally, in (5.3) the eddy forcing term $\mathcal{P} + \mathcal{M}$ can be written in a form analogous to the Eliassen–Palm flux divergence (Andrews, 1982, personal communication), i.e., for a hydrostatic atmosphere, we have, using (A.5) and (A.8):

$$\mathcal{P} = \frac{1}{g} \overline{\frac{\partial p}{\partial \theta} \frac{\partial}{\partial x} \Phi} = \frac{1}{g} \frac{\partial}{\partial \theta} \left(\overline{p \frac{\partial}{\partial x} \Phi} \right) - \frac{c_p}{g} p_{00} \frac{\partial}{\partial x} \overline{\left(\frac{p}{p_{00}} \right)^{k+1}}$$

$$= \frac{1}{g} \frac{\partial}{\partial \theta} \left(\overline{p \frac{\partial}{\partial x} \Phi} \right).$$

Therefore,

$$\mathcal{P} + \mathcal{M} = \frac{\partial}{\partial y} (\overline{\rho_\theta u^* v^*} \cos\varphi)$$

$$+ \frac{\partial}{\partial \theta} \left(\overline{\rho_\theta u^* \dot{\theta}^*} + \frac{1}{g} \overline{p \frac{\partial}{\partial x} \Phi} \right).$$

Once $\bar{U}$ is predicted, the meridional momentum equation

$$f \cos\varphi \bar{U} \approx -\overline{\rho_\theta} \frac{\partial}{\partial y} \bar{\Phi}, \qquad (5.7)$$

can be used to diagnostically calculate the Montgomery streamfunction $\bar{\Phi}$. Eq. (5.7) is the geostrophic approximation to the zonal flow, and its two terms represent the dominant balance in the zonally averaged meridional momentum equation (Gallimore and Johnson, 1981a).

With $\bar{\Phi}$, the temperature field can be diagnosed

easily:

$$\delta \bar{T} = \frac{1}{c_p} \theta \frac{\partial}{\partial \theta} \bar{\Phi}. \qquad (5.8)$$

The quantity $\delta \bar{T}$ is the deviation of the zonal mean temperature from the radiative equilibrium temperature T_e. Therefore the total mean temperature is $\bar{T} = T_e + \delta \bar{T}$. Knowing $\bar{T}$, one can then calculate the new value of $\bar{q}$, using also the predicted values of the radiatively active minor constituent gases, whose distributions are consistently calculated using Eq. (3.8) discussed earlier. Compared to that of Gallimore and Johnson (1981b), the simpler procedure outlined here is a result of our neglecting the time rate of change of mean density in the continuity equation (see Section 2), which makes the determination of $\bar{V}$ diagnostic instead of prognostic.

Parameterization of eddy forcing of the zonal mean flow

As in any zonally averaged model of the general circulation, the eddy forcing terms on the rhs of the momentum equation (5.3) must be parameterized. It is well-known that these eddy forcing terms play an important part in the maintenance of the observed zonal flow in the mesosphere (Leovy, 1964; Holton and Wehrbein, 1980; Lindzen, 1981). If the eddy forcing terms are dropped from Eq. (5.3), the Coriolis torque arising from the diabatic circulation, i.e., the $f \bar{V}$ term, will create an acceleration of the zonal flow with a magnitude on the order of 10^2 (m s^{-1}) day^{-1} or larger. In the models of Leovy, and Holton and Wehrbein (in isobaric coordinates), the eddy momentum flux terms are parameterized as a simple Rayleigh friction acting to decelerate the magnitude of the zonal mean flow. Recently, Lindzen (1981) gave an alternative parameterization by including the deposition of wave momentum by breaking gravity waves and the enhanced turbulent eddy diffusion resulting from these breaking waves. The same physical mechanism and analogous parameterization should also apply to the isentropic coordinate system. In the stratosphere however, the eddy momentum flux is expected to be only of secondary importance.

The pressure torque term has no counterpart in isobaric coordinates. It has the interpretation as the Coriolis torque of the *geostrophic* circulation

$$\mathcal{P} \equiv -\overline{\rho_\theta \frac{\partial}{\partial x} \Phi} = -f \bar{V}_g/\cos\varphi, \qquad (5.9)$$

where

$$\bar{V}_g \equiv \overline{\rho_\theta v_g} \cos\varphi, \quad f v_g = \frac{\partial}{\partial x} \Phi.$$

Because the geostrophic flow $\bar{V}_g$ generally is in the same direction as the total flow $\bar{V}$ away from equatorial regions, this pressure torque term in isentropic coordinates probably tends to reduce the magnitude of the acceleration due to the Coriolis torque. Since

$$f\bar{V} + \mathcal{P}\cos\varphi = f(\bar{V} - \bar{V}_g) \equiv f\bar{V}_a, \quad (5.10)$$

only the *ageostrophic* part of the diabatic meridional circulation is effective in accelerating the mean zonal flow; this is presumably smaller than the acceleration due to the total meridional flow. With only a slight deviation from the one proposed by Gallimore and Johnson (1981b), we suggest the parameterization that $\bar{V}_g$ is proportional to $\bar{V}$, i.e.,

$$\mathcal{P} = fk(y, \theta)\bar{V}/\cos\varphi. \quad (5.11)$$

In extratropical regions in the lower atmosphere, there is some indication (see Gallimore and Johnson, 1981a,b) that $\bar{V}$ is close to $\bar{V}_g$, and hence k should be close to one, while in the tropics, k should be small. k should also decrease somewhat with increasing altitude in the upper stratosphere and mesosphere, to accommodate the increasing importance of eddy damping ($\mathcal{M} + \mathcal{F}$) due to breaking gravity waves mentioned earlier. With (5.11), the zonal momentum equation becomes

$$\frac{\partial}{\partial t}\bar{U} + \frac{\partial}{\partial y}\left(\frac{1}{\rho_\theta}\bar{U}\bar{V}\right) + \frac{\partial}{\partial\theta}\left(\frac{1}{\rho_\theta}\bar{U}\bar{W}\right)$$
$$- f[1 - k(y, \theta)]\bar{V}/\cos\varphi = \mathcal{M} + \mathcal{F}. \quad (5.12)$$

The large acceleration of the Coriolis torque is now diminished by a factor $[1 - k(y, \theta)]$. Through such a simple parameterization, the pressure torque is allowed to play a direct role in counteracting the Coriolis torque, thus helping to maintain the zonal mean flow at reasonable (observed) values.

6. Conclusion

We have endeavored in this paper to formulate a self-consistent zonally averaged model of transports of minor constituent gases in the stratosphere. This is done by adopting the *procedure* of small-amplitude perturbation expansion, the adequacy of which is yet to be assessed for the real atmosphere.

The use of isentropic coordinates does not, by itself, simplify the species transport equation. It does, however, provide a framework for assessing the importance of various terms in that equation. It clearly separates the long-term systematic diabatic process of mean advection from the process of transient eddy dispersion. If the eddy dispersion process in the stratosphere can be assumed to be quasi-adiabatic, then it can be shown that the dispersion occurs predominantly in one direction—along the isentropes. The number of eddy transport terms is thus reduced by a factor of four. The resulting transport equation for a tracer with mean mass mixing ratio $\bar{X}$ is then of the simple form

$$\bar{\rho}_\theta\frac{\partial}{\partial t}\bar{X} + \bar{V}_D\frac{\partial}{\partial y}\bar{X} + \bar{W}_D\frac{\partial}{\partial\theta}\bar{X} = \frac{\partial}{\partial y}\left(\bar{\rho}_\theta D_{yy}\frac{\partial}{\partial y}\bar{X}\right).$$

The mean diabatic mass circulation $(\bar{V}_D, \bar{W}_D)$ is

directly related to the mean diabatic heating rate $\bar{q}$:

$$\bar{W}_D = \bar{q}/\Gamma^{(0)},$$

$$\frac{\partial}{\partial y}\bar{V}_D = -\frac{\partial}{\partial\theta}\bar{W}_D.$$

The streamfunction for this mean circulation determines the mean path along which the tracer is advected, and plays the role of the "mixing path" in the mixing-length theory of Reed and German (1965) with the following important differences:

1) While in Reed and German's formulation the mixing path is fixed by the relative magnitudes of the four *ad hoc* diffusion coefficients, the mean advection path in the present formulation is directly determined by the radiative heating and cooling of the atmosphere. This relationship between radiation and dynamics of transport allows some degree of interactive feedback for the radiatively active minor species, such as ozone and carbon dioxide, whose distribution in the stratosphere determines the distribution of radiative heating and cooling which drive the diabatic circulation that initially affects the species distribution.

2) Instead of mixing along the "mixing path," the presence of transient eddies disperses ("mixes") the tracer, predominantly along the isentropes, which slope less steeply poleward and downward than the advection path in the lower stratosphere.

The dispersion term is quantitatively important near the polar regions, where the streamlines are much steeper than the isentropes. Away from the poles, the eddy dispersion term is less effective because the gradients on the isentropic surfaces of the species concentration created by the mean advection are small. Though this term can be neglected in crude models of tracer transport, without causing a qualitative error (the same cannot be said for the traditional mixing-length model), its effect should be included in a quantitative model. For this purpose, a parameterization of D_{yy} is needed. Because the large and systematic effects of eddy transport that are present in height or pressure coordinates have been removed in the present isentropic coordinate formulation, there is some reason to believe that D_{yy} is due mostly to incoherent irreversible processes and, as such, is suitable for a "turbulence" type of parameterization.

APPENDIX A

Dynamical Equations in Isentropic Coordinates

A brief listing of the dynamical equations in isentropic coordinates is given here. The reader is referred to Holton (1972), Kasahara (1974) and Dutton (1976) for more details.

The first law of thermodynamics is

$$\frac{d}{dt}\ln\theta = \frac{Q}{T}, \tag{A1}$$

which can be rewritten as

$$\dot{\theta} = \frac{\theta}{T}Q, \tag{A2}$$

where $\dot{\theta} \equiv (d/dt)\theta$ is the vertical "velocity". The horizontal momentum equations are:

$$\frac{d}{dt}u - fv = -\frac{\partial}{\partial x}\Phi, \tag{A3}$$

$$\frac{d}{dt}v + fu = -\frac{1}{\cos\varphi}\frac{\partial}{\partial y}\Phi. \tag{A4}$$

The Montgomery streamfunction Φ is related to the pressure through the hydrostatic relation

$$\frac{\partial}{\partial\theta}\Phi = c_p\left(\frac{p}{p_{00}}\right)^\kappa. \tag{A5}$$

The pressure is related to the temperature via the definition

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

and so (A5) can also be written as

$$c_pT = \theta\frac{\partial}{\partial\theta}\Phi. \tag{A6}$$

The equation for conservation of mass is

$$\frac{\partial}{\partial t}\rho_\theta + \frac{\partial}{\partial x}(\rho_\theta u) + \frac{\partial}{\partial y}(\rho_\theta v\cos\varphi) + \frac{\partial}{\partial\theta}(\rho_\theta\dot{\theta}) = 0, \tag{A7}$$

where $\rho_\theta \equiv \rho\partial z/\partial\theta$ is the "density" in isentropic coordinates. The hydrostatic relation can also be written in terms of ρ_θ as

$$g\rho_\theta = -\frac{\partial}{\partial\theta}p. \tag{A8}$$

This provides an equation relating ρ_θ and p. There

are now seven equations, (A2)–(A8), for seven unknowns, u, v, $\dot{\theta}$, ρ_θ, p, T and Φ.

In Section 2, the "density"-weighted quantities have been defined:

$$U \equiv \rho_\theta u, \quad V \equiv \rho_\theta v\cos\varphi, \quad W \equiv \rho_\theta\dot{\theta}. \tag{A9}$$

In terms of these, the equation for conservation of mass can be rewritten

$$\frac{\partial}{\partial t}\rho_\theta + \frac{\partial}{\partial x}U + \frac{\partial}{\partial y}V + \frac{\partial}{\partial\theta}W = 0. \tag{A10}$$

The energy equation is expressed in terms of W and $q \equiv \rho Q$ as

$$W = q/\Gamma, \tag{A11}$$

where

$$\Gamma \equiv \frac{T}{\theta}\frac{\partial\theta}{\partial z} \tag{A12}$$

is the static stability parameter.

APPENDIX B

Some Approximations on Static Stability and Density

The static stability parameter is defined as

$$\Gamma \equiv \frac{T}{\theta}\frac{\partial}{\partial z}\theta \doteq \frac{\partial T}{\partial z} + \frac{g}{c_p}. \tag{B1}$$

The zonally averaged Γ is

$$\bar{\Gamma} = \left(\overline{\frac{\partial T}{\partial z}}\right) + \frac{g}{c_p}; \tag{B2}$$

$$\Gamma' \equiv \Gamma - \bar{\Gamma} = \left(\frac{\partial T}{\partial z}\right)'. \tag{B3}$$

In the stratosphere, the mean vertical temperature gradient $(\partial T/\partial z)$ is ~ 2 K per kilometer, while the constant g/c_p, the so-called adiabatic lapse rate, is ~ 10 K per kilometer. The quantity $\bar{\Gamma}$ is thus seen to be dominated by a large part that does not vary. This seems to justify the approximation of replacing $[(\partial/\partial z)T]$ by a background equilibrium value, $[(\partial/\partial z)T]_e$, or even a meridionally averaged background value $[(\overline{\partial/\partial z})T]_e$. The more drastic approximation of replacing Γ by the adiabatic lapse rate g/c_p has sometimes been used for the stratosphere, though this will not be done here.

Again, because the adiabatic lapse rate g/c_p in $\bar{\Gamma}$ is large, the perturbation static stability Γ' is much smaller than the mean static stability $\bar{\Gamma}$ in the stratosphere, as only rarely can a disturbance create more than a couple degrees of temperature change per kilometer. This observation, however, does not justify the neglect of Γ' in the perturbation energy equation despite the fact that this is a common practice in dynamic meteorology. Starting with the exact equation (A11),

$$W\Gamma = q, \tag{B4}$$

taking the zonal average of (B4) yields

$$\bar{W}\bar{\Gamma} + \overline{W'\Gamma'} = \bar{q}. \qquad (B5)$$

The perturbation equation is obtained by subtracting (B5) from (B4) and retaining leading orders in perturbation amplitude

$$W'\bar{\Gamma} + \bar{W}\Gamma' = q'. \qquad (B6)$$

Note that if Γ' were neglected in Γ, the second term in (B6) would not have appeared. In a formal asymptotic expansion, the second term in Eq. (B6) should be of the same order as the first and there is no *a priori* reason for neglecting it.

Eq. (B6) implies that W' is composed of two parts:

$$W' = W'_D + W'_A, \qquad (B7)$$

a diabatic vertical perturbation flow given by

$$W'_D = q'/\bar{\Gamma}, \qquad (B8)$$

and an adiabatic part arising from perturbations in the static stability parameter:

$$W'_A = -\bar{W}\Gamma'/\bar{\Gamma}. \qquad (B9)$$

The perturbation vertical velocity in isentropic coordinates is in general nonzero unless the atmosphere is strictly adiabatic, i.e., $q' = 0$ *and* $\bar{q} = 0$.

Returning to the mean equation (B5), we now want to show that the term $\overline{W'\Gamma'}$ can be neglected compared to $\bar{W}\bar{\Gamma}$. The ratio of these two terms is

$$\left| \overline{\left(\frac{\Gamma'}{\bar{\Gamma}}\right)\left(\frac{W'}{\bar{W}}\right)} \right| = \left| \overline{\left(\frac{\Gamma'}{\bar{\Gamma}}\right)\left(\frac{W'_D}{\bar{W}}\right)} + \overline{\left(\frac{\Gamma'}{\bar{\Gamma}}\right)\left(\frac{W'_A}{\bar{W}}\right)} \right|$$

$$= O\left(\left| \overline{\left(\frac{\Gamma'}{\bar{\Gamma}}\right)\left(\frac{q'}{\bar{q}}\right)} - \overline{\left(\frac{\Gamma'}{\bar{\Gamma}}\right)\left(\frac{\Gamma'}{\bar{\Gamma}}\right)} \right| \right), \qquad (B10)$$

where the definitions (B7), (B8) and (B9) have been used. As $|\Gamma'/\bar{\Gamma}|$ is small, the second term on the rhs of (B10) is much smaller than unity, while the same is true for the first term unless $|q'/\bar{q}|$ is larger than order one. Assuming this is not so, in order to be consistent with our perturbation procedure, we conclude that the ratio in (B10) is small and so the mean energy equation can be approximated by

$$\bar{W} = \bar{q}/\bar{\Gamma}. \qquad (B11)$$

Eq. (2.9) is then obtained with the further approximation mentioned previously:

$$\bar{\Gamma} \approx \Gamma^{(0)}. \qquad (B12)$$

a. Some comments on the density field in isentropic coordinates

Unlike the "density" field in pressure coordinates

$$\rho_p \equiv \rho \frac{\partial z}{\partial p},$$

which in a hydrostatic atmosphere is a constant $(-g^{-1})$, the "density" field ρ_θ defined in (2.5) is more variable. However, there is some reason to believe that ρ_θ is much less variable than its counterpart ρ in height coordinates. We will attempt to show here that the fluctuations in ρ_θ can approximately be ignored as far as the tracer transport is concerned. First we will argue that the time rate of change of mean density, $(\partial/\partial t)\bar{\rho}_\theta$, is so small that $\bar{V}_T$ is negligible compared to $\bar{V}_D$ under normal conditions; consequently only the diabatic circulation enters into the mean transport of species.

Under the hydrostatic approximation, one has

$$\rho_\theta = -\frac{1}{g}\frac{\partial}{\partial \theta} p. \qquad (B13)$$

Therefore, from (2.18)

$$\frac{\partial}{\partial y}\bar{V}_T = -\frac{\partial}{\partial t}\bar{\rho}_\theta = g^{-1}\frac{\partial}{\partial \theta}\frac{\partial}{\partial t}\bar{p}. \qquad (B14)$$

On the other hand, (2.14) and (2.17) imply

$$\frac{\partial}{\partial y}\bar{V}_D = -\frac{\partial}{\partial \theta}(\bar{q}/\Gamma^{(0)}). \qquad (B15)$$

From Fig. 3, one can infer that the changes in the mean diabatic heating rate in the lower stratosphere are such that $g\bar{q}/\Gamma^{(0)}$ varies by ~ 10 mb per day over a layer of roughly $\Delta\theta \approx 20$ K. Since the zonally averaged pressure field $\bar{p}$ seldom changes by such a large magnitude, especially in the stratosphere, it appears that one always has

$$\left| \frac{\partial}{\partial \theta}\frac{\partial}{\partial t}\bar{p} \right| \ll \left| \frac{\partial}{\partial \theta}(g\bar{q}/\Gamma) \right|. \qquad (B16)$$

This, together with the boundary condition $\bar{V}_T = \bar{V}_D = 0$ at $y = a$, then implies that

$$\left| \frac{\partial}{\partial y}\bar{V}_T \right| \ll \left| \frac{\partial}{\partial y}\bar{V}_D \right| \quad and \quad |\bar{V}_T| \ll |\bar{V}_D|. \qquad (B17)$$

(B17) permits the great simplification of using the divergence-free continuity equation for the determination of the meridional mean circulation, yielding directly

$$\left.\begin{array}{c} \bar{V} \approx \bar{V}_D \\ \bar{W} \approx \bar{W}_D \end{array}\right\}. \qquad (B18)$$

It should be emphasized that without approximation (B17), the use of isentropic coordinates loses the advantage of being more direct than the formulation in pressure coordinates in the calculation of meridional circulation. This is because the quantity $(\partial/\partial t)\bar{\rho}_\theta$ in the continuity equation is not directly determinable from the radiative heating rates. If its contribution cannot be neglected, its calculation would have involved the simultaneous solution of a coupled system of equations.

From the definitions for Γ and ρ_θ, it can be shown that

$$\overset{\circ}{\rho_\theta} = \rho T/(\theta\Gamma),$$

which, for an ideal gas, is

$$\rho_\theta = \frac{p}{\theta R\Gamma}. \qquad (B19)$$

Using the hydrostatic relation (B13), we then have

$$\theta\frac{\partial}{\partial\theta}p + \frac{g}{R\Gamma}p = 0. \qquad (B20)$$

In a "log θ" coordinate,

$$\hat{z} \equiv \ln\left(\frac{\theta}{\theta_0}\right),$$

(B20) can be "solved" to yield

$$p = p_0 \exp\left\{-\int_0^{\hat{z}}\frac{g}{R\Gamma}d\hat{z}\right\}. \qquad (B21)$$

(B19) then implies that the "density" ρ_θ also decreases with increasing potential temperature in a stable atmosphere with $\Gamma > 0$:

$$\rho_\theta = \rho_{\theta_0}\left(\frac{\Gamma_0\theta_0}{\Gamma\theta}\right)\exp\left\{-\int_0^{\hat{z}}\frac{g}{R\Gamma}d\hat{z}\right\}. \qquad (B22)$$

For the background reference "density" $\rho_\theta^{(0)}$, we replace Γ by $\Gamma^{(0)}$ to get

$$\rho_\theta^{(0)} = \rho_{\theta_0}^{(0)}\left(\frac{\Gamma_0^{(0)}\theta_0}{\Gamma^{(0)}\theta}\right)\exp\left\{-\int_0^{\hat{z}}\frac{g}{R\Gamma^{(0)}}d\hat{z}\right\}. \qquad (B23)$$

An approximate expression for $\rho_\theta^{(0)}$ can be obtained if $\Gamma^{(0)}$ is treated as a constant (e.g., $\sim g/c_p$). In this case

$$\rho_\theta^{(0)} \approx \rho_{\theta_0}^{(0)}\left(\frac{\theta}{\theta_0}\right)^{-[g/(R\Gamma^{(0)})-1]}. \qquad (B24)$$

A very simple expression for the background "density" can be obtained as

$$\rho_\theta^{(0)}/\rho_{\theta_0}^{(0)} = \left(\frac{\theta}{\theta_0}\right)^{-9/2}, \qquad (B25)$$

where we have taken $\Gamma^{(0)}$ to be g/c_p, and $\kappa = R/c_p$ to be 2/7. Similarly, the background pressure is given by

$$p^{(0)}/p_0^{(0)} = \left(\frac{\theta}{\theta_0}\right)^{-1/\kappa} = \left(\frac{\theta}{\theta_0}\right)^{-7/2} \qquad (B26)$$

Note that (B26) is approximate (It is exact only for an isothermal atmosphere), while (2.21) for $p_e(\theta)$ is an exact definition.

b. On the horizontal advection caused by density perturbation

We compare here the contribution to the advective transport by the density perturbation to the mean

advection. Referring to Eq. (3.7), we wish to show that the term $(\partial/\partial t)\overline{\rho_\theta'\eta'}$ is small compared to $\bar{V}_D$, for perturbations caused either by quasi-geostrophic waves or by gravity waves. The estimates are very rough, but they should be sufficient for our purpose.

The perturbation continuity equation can be obtained by linearizing Eq. (A7):

$$\left(\frac{\partial}{\partial t} + \bar{u}\frac{\partial}{\partial x}\right)\rho_\theta' + \bar{\rho}_\theta\left[\frac{\partial}{\partial x}u' + \frac{\partial}{\partial y}(v'\cos\varphi)\right]$$

$$+ \frac{\partial}{\partial\theta}(\dot{\theta}'\bar{\rho}_\theta) = 0. \qquad (B27)$$

The last term on the lhs of Eq. (B27) is small for quasi-adiabatic waves. The second term, in brackets, represents the horizontal divergence, and is therefore also small for quasi-geostrophic waves. More specifically, to the lowest order in Rossby number Ro, the second term is zero. Thus we have from (B27),

$$\rho_\theta'/\bar{\rho}_\theta = O(\text{Ro}) \qquad (B28)$$

for quasi-geostrophic waves.

Assessing the approximate orders of magnitude, we have

$$\frac{\partial}{\partial t}\overline{\rho_\theta'\eta'} = O\left(\text{Ro}\bar{\rho}_\theta\frac{\partial}{\partial t}\overline{\eta'}\right) = O(\text{Ro}V') \lesssim O(\text{Ro}\bar{V}_D).$$

It therefore appears that

$$\frac{\partial}{\partial t}\overline{\rho_\theta'\eta'}/\bar{V}_D \lesssim O(\text{Ro}), \qquad (B29)$$

which is small for quasi-geostrophic waves.

For gravity waves, the horizontal convergence term is no longer small. However, the density perturbation ρ_θ' usually tends to be 90° out of phase with the meridional displacement η', with the result that the correlation $\overline{\rho_\theta'\eta'}$ is probably small. To see this, we note that the perturbation horizontal velocities are related to the perturbation Montgomery streamfunction approximately by

$$\left(\frac{\partial}{\partial t} + \bar{u}\frac{\partial}{\partial x}\right)u' = -\frac{\partial}{\partial x}\Phi', \qquad (B30)$$

$$\left(\frac{\partial}{\partial t} + \bar{u}\frac{\partial}{\partial x}\right)v' = -\frac{\partial}{\partial y}\Phi'. \qquad (B31)$$

(Due to the presumably smaller horizontal scale of the gravity waves, the spherical geometry and horizontal shear of the zonal flow are neglected.) Therefore,

$$\left(\frac{\partial}{\partial t} + \bar{u}\frac{\partial}{\partial x}\right)\left(\frac{\partial}{\partial x}u' + \frac{\partial}{\partial y}v'\right)$$

$$= -\left(\frac{\partial^2}{\partial x^2}\Phi' + \frac{\partial^2}{\partial y^2}\Phi'\right) \qquad (B32)$$

and so for quasi-adiabatic waves,

$$\frac{1}{\bar{\rho}_\theta}\left(\frac{\partial}{\partial t} + \bar{u}\frac{\partial}{\partial x}\right)^2 \rho'_\theta = \left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)\Phi'. \quad \text{(B33)}$$

Using (B31) and the definition for η', one has

$$\left(\frac{\partial}{\partial t} + \bar{u}\frac{\partial}{\partial x}\right)^2 \eta' = -\frac{\partial}{\partial y}\Phi' \quad \text{(B34)}$$

and so (B33) becomes

$$\frac{1}{\bar{\rho}_\theta}\left(\frac{\partial}{\partial t} + \bar{u}\frac{\partial}{\partial x}\right)^2 \frac{\partial}{\partial y}\rho'_\theta = -\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)\left(\frac{\partial}{\partial t} + \bar{u}\frac{\partial}{\partial x}\right)^2 \eta'.$$

Hence,

$$\frac{1}{\bar{\rho}_\theta}\frac{\partial}{\partial y}\rho'_\theta = -\left(\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}\right)\eta'. \quad \text{(B35)}$$

Therefore for gravity waves of the form

$$\text{function}(t, \theta) \cdot e^{ikx+ily}, \quad \text{(B36)}$$

Eq. (B35) implies that ρ'_θ is $90°$ out of phase with η'. Hence,

$$\overline{\rho'_\theta \eta'} \approx 0. \quad \text{(B37)}$$

Note that this result holds for transient, growing as well as periodic waves. We have not had to assume that the gravity waves are of the form $e^{ik(x-ct)+ily}$.

APPENDIX C

Diagnostic Calculation of the Diabatic Circulation

For a nondivergent circulation $(\bar{V}_D, \bar{W}_D)$, there exists a streamfunction $\bar{\psi}_D$ such that

$$-\frac{\partial}{\partial\theta}\bar{\psi}_D = \bar{V}_D, \quad \text{(C1)}$$

$$\frac{\partial}{\partial y}\bar{\psi}_D = \bar{W}_D. \quad \text{(C2)}$$

Since in the isentropic coordinate system one has

$$\bar{W}_D = \bar{q}/\Gamma^{(0)}, \quad \text{(C3)}$$

the streamfunction is found from (C2) to be

$$\bar{\psi}_D = \int_a^y \bar{q}/\Gamma^{(0)}dy, \quad \text{(C4)}$$

where the y-integration is to be performed on surfaces of constant potential temperature.

A conservative tracer advected by the meridional circulation $(\bar{V}_D, \bar{W}_D)$ satisfies

$$\bar{\rho}_\theta\frac{\partial}{\partial t}\bar{X} + \bar{V}_D\frac{\partial}{\partial y}\bar{X} + \bar{W}_D\frac{\partial}{\partial\theta}\bar{X} = 0. \quad \text{(C5)}$$

It is easy to verify that the steady-state solution to Eq. (C5) is

$$\bar{X} = F(\bar{\psi}_D). \quad \text{(C6)}$$

Therefore the isopleths of $\bar{X}$ are the same as the iso-

pleths of $\bar{\psi}_D$. In other words, lines of constant $\bar{\psi}_D$ are also the lines of constant $\bar{X}$.

For a diagnostic study, we take the radiative heating rates calculated by Dopplick (1979) and deduce $\bar{W}_D$ and $\bar{\psi}_D$ in the following manner. Since the published values are for the seasonal average of $\bar{Q}$, these will be used here, though Dopplick also provides tabulated monthly values of $\bar{Q}$ on request. To calculate $\bar{W}_D$, the vertical diabatic mass flow rate, the heating rate per unit volume is needed. Since this is not available, the approximate expression $\bar{q} \doteq \bar{\rho}\bar{Q}$ is used. This quantity is then estimated by first calculating $\bar{\rho}$ using the ideal gas law

$$\bar{\rho} = p/R\bar{T},$$

with the zonally averaged temperature field $\bar{T}$ taken from Fig. 4 of Dopplick (1972) at each pressure level. With $\bar{Q}$ provided by Dopplick, the quantity $\bar{q}$ is deduced at each pressure level. The static stability parameter

$$\Gamma^{(0)} \approx \frac{98.1}{9.96}\ \text{K km}^{-1} + \frac{\partial\bar{T}}{\partial z}$$

is calculated here using the temperature data mentioned above and height z deduced approximately from the pressure. The ratio

$$\bar{W}_D = \bar{q}/\Gamma^{(0)}$$

is depicted in Fig. 3 as a function of pressure and latitude. The pressure coordinate is used here because that is the coordinate in which Dopplick's data are expressed.

To deduce the streamfunction $\bar{\psi}_D$, one needs to re-express $\bar{W}_D$ in isentropic coordinates. This conversion cannot be done exactly with Dopplick's data. The approximate procedure adopted here is to use the temperature data from Fig. 4 of Dopplick (1972) and calculate the potential temperature as

$$\theta \approx \bar{T}\left(\frac{p_{00}}{p}\right)^\kappa.$$

With θ thus calculated, we obtain values of $\bar{W}_D$ at various constant potential temperature surfaces, and $\bar{\psi}_D$ is then obtained from

$$\bar{\psi}_D/a = \int_{\pi/2}^\varphi (\bar{q}/\Gamma)_\theta \cos\varphi\, d\varphi,$$

with the integration performed using the trapezoidal rule. The main uncertainty in the streamfunction obtained from Dopplick's data occurs near the tropopause where large vertical gradients exist. Due to the lack of resolution in this region it is almost impossible to decide whether a given constant streamline should continue northward or southward. These uncertain lines are indicated in Fig. 4 with dashed lines. A second difficulty is encountered near the south pole, where the boundary condition $\bar{V}_D = 0$ cannot be satisfied. This is a problem common to all

analyses dealing with imperfect data, whether in pressure or isentropic coordinates. To overcome this problem, Dunkerton (1978) chose to alter the data at other latitudes slightly to make $\bar{V}_D = 0$ at both poles. No such alteration to Dopplick's values is done here. We simply recognize the fact that the calculated value for the streamfunction is unreliable near the south pole due to the fact that any error in the data accumulates in the integral. Only the north polar boundary condition is enforced by starting the integration for the streamfunction from $\varphi = \pi/2$.

Dopplick (1972, 1979) provided no information above 10 mb. From Murgatroyd and Singleton (1961), it is known that the diabatic circulation in the upper stratosphere and mesosphere consists of a single cell with rising motion in the south polar region and sinking motion in the north. This feature is indicated in Fig. 3 with heavy dashed lines. It is obvious that these two sets of data are incompatible with each other. Nevertheless, the qualitative nature of the physical situation seems to be clear: the two-cell structure of the diabatic circulation in the lower stratosphere changing into a single-cell structure appears to be correct and consistent with other tracer studies (see Dunkerton, 1978).

A main deficiency in this diagnostic study is recognized as the lack of radiation data $\bar{q}$ calculated and presented *in isentropic coordinates*. Hopefully this situation will change as the isentropic coordinate system becomes more commonly used.

APPENDIX D

The Zonally-Averaged Species Transport Equation

To find X' we linearize Eq. (2.1) about a mean zonal state to yield

$$\left(\frac{\partial}{\partial t} + \bar{u}\frac{\partial}{\partial x}\right)X' + v'\cos\varphi\,\frac{\partial}{\partial y}\,\bar{X}$$

$$+ \,\theta'\frac{\partial}{\partial\theta}\,X' = S',\quad (D1)$$

where the meridional advection of X' has been neglected compared to the zonal advection of the same quantity. With the following definition of displacement fields:

$$\left(\frac{\partial}{\partial t} + \bar{u}\frac{\partial}{\partial x}\right)\eta^r \equiv v'\cos\varphi,\quad (D2)$$

$$\left(\frac{\partial}{\partial t} + \bar{u}\frac{\partial}{\partial x}\right)\phi' \equiv \theta',\quad (D3)$$

$$\left(\frac{\partial}{\partial t} + \bar{u}\frac{\partial}{\partial x}\right)\sigma' \equiv S',\quad (D4)$$

Eq. (D1) can be solved approximately; the result is

$$X' = -\eta'\frac{\partial}{\partial y}\,\bar{X} - \phi'\frac{\partial}{\partial\theta}\,\bar{X} + \sigma'.\quad (D5)$$

In arriving at (D5) from (D1), we have made the assumption that the time variation of the mean concentration can be taken to be much slower than the time variation of the perturbation quantities and so the mean concentration has been taken to be quasi-steady when the perturbation quantities are calculated.

For conservative tracers, (D5) reduces to the form assumed by the mixing-length theory (see Reed and German, 1965; Green, 1970), though the definition of the displacement fields is slightly different. Similar results have previously been obtained by Plumb (1979), Matsuno (1980), Holton (1980, 1981) and Danielsen (1981).

Using (D5), we find the flux terms to be expressible as

$$\overline{V'X'} = -\bar{\rho}_\theta K_{yy}\frac{\partial}{\partial y}\,\bar{X} - \bar{\rho}_\theta K_{y\theta}\frac{\partial}{\partial\theta}\,\bar{X} + \overline{V'\sigma'},\quad (D6)$$

$$\overline{W'X'} = -\bar{\rho}_\theta K_{\theta y}\frac{\partial}{\partial y}\,\bar{X} - \bar{\rho}_\theta K_{\theta\theta}\frac{\partial}{\partial\theta}\,\bar{X} + \overline{W'\sigma'}.\quad (D7)$$

In (D6) and (D7), the coefficients are defined as

$$\left. \begin{array}{ll} K_{yy} \equiv \dfrac{1}{\bar{\rho}_\theta}\,\overline{V'\eta'}, & K_{\theta\theta} \equiv \dfrac{1}{\bar{\rho}_\theta}\,\overline{W'\phi'} \\[2ex] K_{y\theta} \equiv \dfrac{1}{\bar{\rho}_\theta}\,\overline{V'\phi'}, & K_{\theta y} \equiv \dfrac{1}{\bar{\rho}_\theta}\,\overline{W'\eta'} \end{array} \right\}.\quad (D8)$$

To express the K's in terms of the displacements η' and ϕ' only, we note that

$$V' = (\rho_\theta v\cos\varphi)' \approx \bar{\rho}_\theta v'\cos\varphi + \bar{v}\cos\varphi\rho_\theta'.\quad (D9)$$

Therefore,

$$\frac{1}{\bar{\rho}_\theta}\,V' = \left(\frac{\partial}{\partial t} + \bar{u}\frac{\partial}{\partial x}\right)\eta' + \bar{v}\cos\varphi\,\frac{\rho_\theta'}{\bar{\rho}_\theta}.\quad (D10)$$

From the continuity equation, one can show that $\rho_\theta'/\bar{\rho}_\theta$ is of the magnitude $(\partial/\partial y)\eta'$; therefore, the last term in (D10) is of the magnitude of the meridional advection of η' and should be dropped when compared to the zonal advection retained in the first term on the rhs of (D10). This is to be consistent with the degree of approximation in (D1). Therefore we have

$$\frac{1}{\bar{\rho}_\theta}\,V' \approx \left(\frac{\partial}{\partial t} + \bar{u}\frac{\partial}{\partial x}\right)\eta'\quad (D11)$$

and similarly,

$$\frac{1}{\bar{\rho}_\theta}\,W' \approx \left(\frac{\partial}{\partial t} + \bar{u}\frac{\partial}{\partial x}\right)\phi'.\quad (D12)$$

Using these two expressions, one then has, from (D8)

$$\left. \begin{array}{ll} K_{yy} = \dfrac{\partial}{\partial t}\,\tfrac{1}{2}\overline{\eta'\eta'}, & K_{\theta\theta} = \dfrac{\partial}{\partial t}\,\tfrac{1}{2}\overline{\phi'\phi'} \\[2ex] K_{y\theta} = \dfrac{\partial}{\partial t}\,\overline{\eta'\phi'} - \dfrac{1}{\bar{\rho}_\theta}\,\overline{\eta'W'}, & K_{\theta y} = \dfrac{1}{\bar{\rho}_\theta}\,\overline{\eta'W'} \end{array} \right\}.\quad (D13)$$

Substituting these back into Eq. (3.2), one then obtains the transport equation

$$
\bar{\rho}_\theta \frac{\partial}{\partial t} \bar{X} + \left(\bar{V} - \frac{\partial}{\partial t} \overline{\rho'_\theta \eta'} \right) \frac{\partial}{\partial y} \bar{X} + \left(\bar{W} - \frac{\partial}{\partial t} \overline{\rho'_\theta \phi'} \right) \frac{\partial}{\partial \theta} \bar{X}
$$

$$
- \frac{\partial}{\partial y} \left(\bar{\rho}_\theta K_{yy} \frac{\partial}{\partial y} \bar{X} \right) - \frac{\partial}{\partial y} \left(\bar{\rho}_\theta K_{y\theta} \frac{\partial}{\partial \theta} \bar{X} \right)
$$

$$
- \frac{\partial}{\partial \theta} \left(\bar{\rho}_\theta K_{\theta y} \frac{\partial}{\partial y} \bar{X} \right) - \frac{\partial}{\partial \theta} \left(\bar{\rho}_\theta K_{\theta\theta} \frac{\partial}{\partial \theta} \bar{X} \right) = \bar{\rho}_\theta \bar{S}
$$

$$
+ \overline{\rho'_\theta S'} - \frac{\partial}{\partial y} (\overline{V'\sigma'}) - \frac{\partial}{\partial \theta} (\overline{W'\sigma'}) - \frac{\partial}{\partial t} (\overline{\rho'_\theta \sigma'}) = \bar{\rho}_\theta \bar{S}
$$

$$
- \left(\frac{\partial}{\partial t} + \bar{u} \frac{\partial}{\partial x} \right) \overline{\rho'_\theta \sigma'} - \frac{\partial}{\partial y} (\overline{V'\sigma'}) - \frac{\partial}{\partial \theta} (\overline{W'\sigma'}). \quad (D14)
$$

The eddy transports in (D14) consist of both the processes of advection and "diffusion". Separating out a nondivergent eddy-induced mean advective transport, one can rewrite (D14) in the form adopted in Eq. (3.4).

The source terms of the rhs of Eq. (D14) can be expressed in a slightly different form with the use of the perturbation continuity equation:

$$
\left(\frac{\partial}{\partial t} + \bar{u} \frac{\partial}{\partial x} \right) \rho'_\theta + \frac{\partial}{\partial x} (\bar{\rho}_\theta u') + \frac{\partial}{\partial y} (\bar{\rho}_\theta v' \cos\varphi)
$$

$$
+ \frac{\partial}{\partial \theta} (\bar{\rho}_\theta \theta') = 0. \quad (D15)
$$

Defining the meridional and vertical displacements as before and the zonal displacement ξ' according to

$$
\left(\frac{\partial}{\partial t} + \bar{u} \frac{\partial}{\partial x} \right) \xi' = u' + \left[\frac{\partial}{\partial y} \bar{u} \cdot \eta' + \frac{\partial}{\partial \theta} \bar{u} \cdot \phi' \right], \quad (D16)
$$

one then obtains from (D15) the following solution for ρ'_θ:

$$
\rho'_\theta = - \left[\frac{\partial}{\partial x} (\bar{\rho}_\theta \xi') + \frac{\partial}{\partial y} (\bar{\rho}_\theta \eta') + \frac{\partial}{\partial \theta} (\bar{\rho}_\theta \phi') \right]. \quad (D17)
$$

Thus the right-hand side of Eq. (D14) becomes

$$
\bar{\rho}_\theta \bar{S} - \left(\frac{\partial}{\partial t} + \bar{u} \frac{\partial}{\partial x} \right) \overline{\rho'_\theta \sigma'} - \frac{\partial}{\partial y} (\overline{V'\sigma'}) - \frac{\partial}{\partial \theta} (\overline{W'\sigma'})
$$

$$
= \bar{\rho}_\theta \bar{S} + \bar{\rho}_\theta \left[\overline{\xi' \frac{\partial}{\partial x} S'} + \overline{\phi' \frac{\partial}{\partial y} S'} + \overline{\phi' \frac{\partial}{\partial \theta} S'} \right]
$$

$$
- \frac{\partial}{\partial t} \left[\overline{\rho'_\theta \sigma'} + \frac{\partial}{\partial y} (\overline{\bar{\rho}_\theta \eta' \phi'}) + \frac{\partial}{\partial \theta} (\overline{\bar{\rho}_\theta \phi' \sigma'}) \right]. \quad (D18)
$$

This form will be useful for comparing with Lagrangian means.

APPENDIX E

Quasi-Adiabatic Eddies

Atmospheric wave motions are in general nonadiabatic. We shall divide the disturbance field into two categories according to the origin of diabatic forcing:

1) Waves forced *in situ* in the stratosphere,
2) Waves remotely forced (mainly in the troposphere) that propagate into the region under consideration.

a. Locally forced waves

The zonally asymmetric wave field forced *in situ* in the stratosphere and mesosphere has its origin mainly in the daily cycle of solar insolation principally in the semi-diurnal mode. These harmonic waves can be written approximately in the form

$$
W'_d = \text{Re}[A(y, \theta) e^{i(s\lambda + \omega t)}], \quad (E1)
$$

where s is the zonal wavenumber and ω is the frequency. Waves of this form do not contribute to "diffusion" despite their relatively high frequency, even though the "diffusion" terms derived in the last section are all in the form of explicit time derivatives. This is because time derivatives of zonal means of harmonic waves vanish. (Though the cross correlations between, for example, the semi-diurnal and diurnal modes, can still contribute to the "diffusion" coefficients, these terms are presumably small in amplitude, because the diurnal mode is weaker than the semi-diurnal mode in the stratosphere.) Their contribution to the transport of tracer is mainly advective in nature, through the flux term $\overline{\eta'_d W'_d}$. However, owing to the small amplitude of thermal tides in the stratosphere, these eddy advection terms appear to be negligible compared to the mean diabatic advection, though the effect of tides may become significant in the mesosphere.

b. The remotely forced waves

The remotely forced planetary and gravity waves, on the other hand, can have significant amplitudes in the stratosphere because of their large energy source in the troposphere and their amplitude increase with decreasing density in their upward propagation. Away from their sources of forcing (e.g., topography, latent heat release from cumulus clouds), their upward propagation can be described reasonably well by almost-adiabatic processes [see e.g., Matsuno (1970) for planetary waves]. The waves are considered diabatic if they can exchange appreciable heat with the medium as they propagate through it.

Let Δt be the residence time, i.e., the length of time a wave spends in the region of the atmosphere of interest. The photochemically active layer of the atmosphere is $\sim$30 km thick (between 25 km and 55 km in altitude). Thus for the problem at hand, we have

$$
\Delta t \approx O\left(\frac{30 \text{ km}}{c_g} \right), \quad (E2)
$$

where c_g is the vertical group velocity of the wave under consideration. Though there is still some uncertainty, a reasonable measure of a wave's diabaticity appears to be the fractional change of the wave's potential temperature due to diabatic processes. This quantity will be estimated as

$$|\Delta\theta/\theta| = O(\dot{\theta}\Delta t/\theta). \qquad (E3)$$

Since $\dot{\theta}$ is usually of the same order as $\bar{\dot{\theta}} = \bar{Q}/\bar{T}$, we have

$$|\Delta\theta/\theta| = O(\bar{Q}\Delta t/\bar{T}). \qquad (E4)$$

Using Dopplick's (1979) data, below 30 km the mean diabatic heating rate $\bar{Q}$ during winter is on the order of 1 K day^{-1} and the mean temperature $\bar{T}$ is $\sim$220 K. Therefore, (E4) implies that

$$|\Delta\theta/\theta| = O(\Delta t/220 \text{ days}). \qquad (E5)$$

The residence time calculated using (E2) is generally approximately two weeks for the stationary planetary waves, and less than two days for gravity waves. [Exceptions to (E6) may occur, especially for waves that encounter critical levels in the atmosphere. We will assume, however, that most of the wave energy of tropospheric origin lies in low frequencies and so most of the disturbances do not have a critical level in the stratosphere.] Therefore it appears that

$$|\Delta\theta/\theta| \ll 1. \qquad (E6)$$

Accordingly, we shall assume that the *eddies* are approximately adiabatic as far as *long-term* transport is concerned. (A more careful analysis, which is not done here, should involve a comparison of various terms in the transport equation to ascertain if the transport terms arising from vertical displacements are actually small.)

Maximum advantage of this quasi-adiabatic property of the waves can be taken, if one adopts the isentropic coordinate system in tracer transport calculations. As a wave propagates through the stratosphere, the isentropes are displaced up and down relative to a fixed height. [For planetary-scale disturbances, the isentrope displacement often occurs in a seesaw pattern (Hsu, 1981).] Since the potential temperature of the fluid is not appreciably affected by the passage of a wave, a fluid particle stays close to its original isentropic surface during the eddy event, even though that surface is itself being displaced up and down by the wave. In isentropic coordinates, this observation implies that the disturbance does not induce a "vertical" displacement. Hence,

$$W' \approx 0, \quad \phi' \approx 0. \qquad (E7)$$

There are, however, some eddy displacements *along* the isentropic surfaces induced by the upward-propagating wave. This is probably due mainly to the buoyancy effect acting on fluid particles on a displaced isentropic surface. As a part of an isentrope bulges downward in response to the wave disturbance, the fluid particles on that part of the surface acquire a positive buoyancy because they have come from a higher altitude, where the density is lower. These fluid particles will have an initial tendency to move upward. They do so by moving up the slope along the same isentropic surface. The restoring forces (e.g., gravity, β-effect) will then set up an oscillation along that surface. It is through such a mechanism that quasi-adiabatic waves forced in the troposphere propagate to the stratosphere. Unlike other coordinate systems, the wave displacement of such a disturbance in isentropic coordinates occurs in one direction (horizontal) only.

APPENDIX F

List of Symbols

p — pressure

p_e — radiative equilibrium pressure $[\equiv p_{00}(\tilde{T}_e/\theta)^{1/\kappa}]$

q — diabatic heating rate per unit volume divided by c_p

u — eastward velocity

v — northward velocity

w — vertical velocity $[\equiv dz/dt]$

y — northward horizontal coordinate $[=a \sin\varphi$ on a sphere with radius a and latitude $\varphi]$

x — eastward horizontal coordinate $[dx = a \times \cos\varphi d\lambda$ on a sphere where λ is longitude]

z — height

D_0 — $\equiv \dfrac{\partial}{\partial t} + \dfrac{\bar{u}}{a \cos\varphi}\dfrac{\partial}{\partial \lambda}$

H — the scale height of the atmosphere $[\equiv R\tilde{T}_e/g]$

Q — diabatic heating rate per unit mass divided by c_p (K per day)

S — net rate of production of X, source per unit mass

T — temperature

T_e — background radiative equilibrium temperature

$\tilde{T}_e$ — meridional average of T_e, function of vertical coordinate only: $\tilde{T}_e = \tilde{T}_e(\theta)$; in practice, usually taken to be the zonal and meridional average of the observed temperature field

U — $\rho_\theta u$ (eastward mass flow rate in isentropic coordinates)

V — $\rho_\theta v \cos\varphi$ (northward mass flow rate in isentropic coordinates)

W — $\rho_\theta\dot{\theta}$ (upward mass flow rate in isentropic coordinates)

$\bar{V}_E$ — advective northward mass flow induced by the zonal eddies

$\bar{W}_E$ — advective vertical mass flow induced by the zonal eddies

$\bar{W}_D$ — $\equiv \bar{q}/\Gamma^{(0)}$

$\bar{V}_D$ — $\equiv -\displaystyle\int_a^y \dfrac{\partial}{\partial\theta} \bar{W}_D dy$

$$\bar{V}_T \equiv -\int_a^y \frac{\partial}{\partial t}\bar{\rho}_\theta dy$$

X species concentration per unit mass of air, also called mass mixing ratio

θ potential temperature $[T(p_{00}/p)^\kappa$, where $p_{00} = 1000$ mb]

$\dot{\theta}$ $\equiv d\theta/dt$ ("vertical velocity" in an isentropic coordinate system)

κ R/c_p where R is gas constant, c_p is specific heat at constant pressure

ρ density of air, mass per unit volume $dxdydz$

ρ_θ mass of air per unit "volume" $dxdyd\theta$ $[=\rho\partial z/\partial\theta]$

$\rho_\theta^{(0)}$ background radiative equilibrium "density" in isentropic coordinates $[=p_e/(\theta R\Gamma^{(0)})$; also $= -g^{-1}\partial p_e/\partial\theta$ in a hydrostatic atmosphere]

η' northward displacement field

ϕ' vertical displacement field

ξ' eastward displacement field

σ' source "displacement" defined from $D_0\sigma' = S'$

$\Gamma^{(0)}$ static stability parameter

$$\left[\equiv \frac{T_e}{\theta_e}\frac{\partial\theta_e}{\partial z} \approx \frac{\partial T_e}{\partial z} + \frac{g}{c_p}\right]$$

$(\ ^-\ )$ zonal average $\left[= \frac{1}{2\pi}\int_0^{2\pi}(\ \ )d\lambda\right]$

$(\ \ )'$ deviation from the zonal mean $[=(\ \ ) - (\ ^-\ )]$

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