Diabatic cross-isentropic dispersion in the lower stratosphere

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Abstract.

A significant contribution to vertical dispersion of tracers in the stratosphere arises from the variability in the diabatic heating of air parcels. Air parcels starting on a given isentropic surface experience different time histories of diabatic heating, which causes vertical dispersion across isentropic surfaces. We refer to this process as "diabatic cross-isentropic dispersion," or "diabatic dispersion" for brevity. The present study investigates diabatic dispersion in the lower stratosphere by computing parcel trajectories initialized uniformly over the 500 K surface on 1 Jan 1993. Parcels are followed for two months using analyzed winds and diabatic heating rates computed from analyzed temperatures. Two independent datasets and radiation schemes are used.

Diabatic dispersion depends on the statistics of the large-scale horizontal eddy motion as well as on the spatial structure, be it steady or time varying, of the diabatic heating field. The trajectory statistics suggest that the polar vortex, winter hemisphere surf zone, tropics, and extratropical summer hemisphere are, to varying extents, isolated from each other by eddy transport barriers. The character and magnitude of the diabatic dispersion for parcels that stay within each of these four regions is distinctly different. The diabatic dispersion in both the surf zone and southern hemisphere extratropics is initially advective, with potential temperature variance $\langle \delta\theta(t)^2 \rangle$ increasing like t^2 as time t increases. After about one month, on the 500 K isentropic surface, the dispersion becomes diffusive in the sense that $\langle \delta\theta(t)^2 \rangle \sim 2K_{\theta\theta}t$ with a diffusivity $K_{\theta\theta}$ in the range 2–6 $\mathrm{K}^2\,\mathrm{day}^{-1}$, roughly equivalent to $K_{zz} \sim$ 0.1–0.2 $\mathrm{m}^2\mathrm{s}^{-1}$. The emergence of a diffusive regime is discussed in terms of loss of memory of diabatic heating along parcel paths, as measured by the decay of the Lagrangian autocorrelation function. Diabatic dispersion within the tropics and polar vortex over the two month period is more than an order of magnitude smaller, and is less clearly diffusive. Some parcels do not remain within any of the four regions defined above, and show entirely different dispersion characteristics. The diabatic dispersion of parcels moving poleward out of the tropics

into either hemisphere is faster than either differential advection or diffusion, and is consistent with a transient shear dispersion model in which $\langle \delta\theta(t)^2 \rangle$ increases like t^3. For the total ensemble of all parcels, the potential temperature variance increases like t^2, consistent with global-scale differential advection by the mean diabatic circulation. This is inconsistent — at least on the two-month timescale considered here — with a one-dimensional diffusive model of vertical dispersion.

1. Introduction

There are well-established advantages to considering stratospheric transport in an isentropic formalism, distinguishing between adiabatic motion along isentropes and diabatic motion across isentropes. The gross characteristics of the observed transport of stratospheric tracers across isentropic surfaces may be explained by considering the vertical advective transport due to the Eulerian zonal-mean circulation in isentropic coordinates, hereafter the "mean diabatic circulation". The sense of this circulation is, in the lower stratosphere, upward in the tropics and downward at middle and high latitudes. However, for a complete picture of tracer transport, especially in the lower stratosphere, it is necessary to take account of more than vertical advection by the mean diabatic circulation. An ensemble of air parcels starting on a given isentropic surface will have different large-scale horizontal trajectories and therefore will generally experience different histories of diabatic vertical velocity. The ensemble will therefore be dispersed across isentropic surfaces. We shall refer to this large-scale process as "diabatic cross-isentropic dispersion", hereafter "diabatic dispersion" for brevity. Additionally, there may be cross-isentropic dispersion due to local small-scale mixing across isentropic surfaces by sporadic three-dimensionally turbulent layers [e.g., *Dewan*, 1981].

Such eddy transport across isentropic surfaces may have important implications, e.g. for the chemical effects of pollutants in the atmosphere. Aircraft exhaust emissions affect ozone chemistry, and the effects vary strongly with altitude. Therefore, mechanisms that permit upward transport of pollutants from their emission region are of particular interest. According to a transport model containing only advection by the mean diabatic circulation, aircraft exhaust emitted in the middle and high latitude lower stratosphere would simply descend from the emission region into the troposphere. In reality, however, the effect of large-scale dispersion and small-scale mixing might be to allow some of the exhaust to rise above the emission region. Similar considerations are relevant also, for instance, to debates about what happens to chemically processed polar-vortex and sub-vortex air and the consequences for midlatitude ozone depletion [e.g. *Albritton et al.*, 1994; *McIntyre*, 1995, *Jones and Kilbane-Dawe*, 1997].

Diabatic dispersion is likely to be different in different geographical regions. There is much evidence from theoretical and modeling studies, and from observations, that rapid global transport on isentropic surfaces is hindered by the presence of eddy transport barriers such as that between the winter polar vortex and the surf zone [e.g., *McCormick et al.*, 1983; *Juckes and McIntyre*, 1987] and that between the surf zone and tropics [e.g. *Hitchman et al.*, 1994; *McIntyre*, 1990; *Chen et al.*, 1994; *Polvani et al.*, 1995]. These barriers are manifested by sharp gradients in volcanic aerosols, chemical tracers or potential vorticity (hereafter PV) [e.g. *Grant et al.*, 1996; *Schoeberl et al.*, 1992; *Murphy et al.*, 1993]. It has been argued that a subtropical eddy transport barrier is necessary for dynamical reasons, for consistency with the modest easterly winds and easterly accelerations observed in the equatorial quasi-biennial oscillation (hereafter QBO) [*McIntyre*, 1990]. The likely existence, and interannual persistence, of these subtropical barriers has led to suggestions that one-dimensional, global-scale, vertically diffusive models of long-lived chemical tracer behavior in the stratosphere [*Holton*, 1986; *Mahlman et al.*, 1986; *Plumb and Ko*, 1992] would be more aptly replaced by "tropical pipe" or "tropical leaky pipe" models [*Plumb*, 1996].

The present work aims to quantify, using meteorological analyses, diabatic dispersion in the lower stratosphere and to investigate its dependence both on the spatial characteristics of the diabatic heating field and on the large-scale horizontal eddy motion, within each of the regions separated by eddy transport barriers. In the extratropical regions, for example, the vertical dispersion occurs largely as a result of horizontal mixing across a large latitudinal gradient in diabatic heating. This is closely analogous to the effect of molecular diffusion on the dispersion of a dye in pipe flow [*Taylor*, 1953]. Fast diffusion across the radial gradient in the axial pipe flow results in dispersion along the axis of the pipe; this axial dispersion is an example of what is generically referred to as "shear dispersion." In the present context, the diabatic vertical velocity corresponds to the axial flow and the horizontal mixing to the radial diffusion.

Because diffusive models are frequently used to parameterize transport processes, often without sufficient justification, another aim is to develop a more quantitative understanding of the conditions under which one expects diabatic dispersion to resemble simple diffusion, at least in an ensemble mean, with a well defined diffusion coefficient and a displacement variance proportional to time t.

In this paper we confine attention to the diabatic dispersion of parcels initialized on the 500 K potential temperature surface, and followed over the period January and February 1993. We quantify the dispersion by considering the time evolution of the Lagrangian potential temperature variance. The potential temperature is a natural vertical coordinate for the study of irreversible vertical dispersion because it excludes from the analysis the vertical displacements associated with reversible undulations of isentropic surfaces, which can be both large and rapid but are irrelevant for present purposes.

Section 2 gives details of the trajectory and heating calculations and summarizes characteristics of the transport and radiative heating field that are relevant for the vertical dispersion problem. Section 3 defines subensembles of parcels that are confined to different geographical regions and discusses the Lagrangian mean motion for each subensemble. Diabatic dispersion is quantified in Section 4 by considering the time evolution of the potential temperature variance separately for each subensemble. There are some regions for which the vertical dispersion resembles simple diffusion, and others where it clearly does not. Section 5 presents a theoretical analysis that discusses the connections between such simple-diffusive dispersion and the loss of memory of diabatic heating along parcel trajectories, as quantified in terms of the relevant Lagrangian autocorrelation function (LAF). This loss of memory occurs over the integral timescale τ_L associated with the LAF. Over timescales much longer than τ_L, the cross-isentropic parcel displacements can be regarded as composed of a Lagrangian-mean drift plus a random walk with independent steps, and the dispersion of the ensemble becomes diffusive. Estimates are given for τ_L, which in the extratropics appears to be well defined and of order 10 days. Section 6 is a discussion and summary of the main results, which are shown to be more consistent with the tropical-leaky-pipe models than with the global-scale, vertically diffusive models.

2. Heating Rate and Trajectory calculations

Because diabatic dispersion arises from the exposure of air parcels to different radiative heating rates, it is most readily studied using trajectory models. Here "model" means a computational procedure using a radiation scheme together with horizontal winds from meteorological analyses — which of course depend on forecast models as well as on observations — to estimate the air-parcel trajectories. Results are presented in this paper that are based on calculations with two different trajectory models. This allows a cross check of the results, and therefore gives greater confidence in any conclusions. Both models include horizontal advection along isentropes using analyzed winds and cross-isentropic advection using radiative heating rates calculated from analyzed temperature fields.

The first trajectory model is the Goddard Space Flight Center model (hereafter GSFC) [*Schoeberl and Sparling*, 1995], which uses GSFC analyzed winds for the horizontal advection along isentropes, and, for the cross-isentropic advection, heating rates computed from the GSFC analyzed temperatures using the Goddard radiation scheme described in [*Rosenfield et al.*, 1994] and references therein. The GSFC analysis is a stratospheric version of the tropospheric GSFC/GEOS-1 assimilation system [*Schubert et al.* 1993]. Calculations using this model will be referred to as GSFC calculations.

The second trajectory model is an adaptation of that of *Fisher et al.* [1993] which uses UK Meteorological Office (hereafter UKMO) wind and temperature analyses [*Swinbank and O'Neill*, 1994]. Heating rates are calculated using the MIDRAD radiative heating algorithm of *Shine* [1987]. A correction has been applied to the MIDRAD heating rates to ensure the mass fluxes through an isentropic surface balance any increase or decrease of mass above the surface. (*Shine* [1989] gives a more detailed discussion of reasons for applying such a correction.) Calculations using this second model will be referred to as UKMO calculations. Figure 1 is a comparison of the GSFC and UKMO zonal mean diabatic heating rates on 1 February 1993, which are typical for the period of interest. In each case, the longitudinal variability in the heating rate has been superimposed on the zonal mean. Here, and in what follows, potential temperatures will be written as K and temperature written as °K. In much of what follows heating rates are expressed as a rate of change of potential temperature (K day^{-1}). It will be made explicit where heating rates are expressed as a rate of change of temperature (°K day^{-1}).

Both radiative heating calculations give a large midlatitude gradient in the heating in both northern and southern hemispheres, and much of the diabatic dispersion in midlatitudes arises from latitudinal motion across this gradient. The gradients in radiative heating are shallow within the polar vortex, and as we show later, the dispersion is weak there. Large differences between GSFC and UKMO radiation schemes are apparent in the tropics, where different predictions for diabatic dispersion are therefore to be expected. The GSFC zonal mean heating rate varies little in the tropics, in contrast to the UKMO heating rates, which have a strong local minimum at the equator.

There are two factors which may contribute to differences between the GSFC and UKMO heating rates in the tropics. In the UKMO calculation, the climatological effects of clouds were included while the GSFC calculations were carried out for clear sky conditions. Preliminary calculations indicate that upper tropospheric cirrus could lower the heating rates locally by as much as 0.5 K day^{-1} near the equator at the 50 mb level. Another contributing factor might be differences in the temperature profiles used in the two schemes, for example during this time period the GSFC tropical tropopause temperature is 4–5°K warmer than the UKMO tropical tropopause. We also note that the GSFC and UKMO analyses have different residual vertical mass fluxes in the lower stratosphere, particularly in the tropics where the differences are similiar to the differences in the heating rates shown in Figure 1 [*Coy and Swinbank*, 1996]. The disagreement between the two radiative codes in the tropics, together with uncertainties in the tropical winds, means that the diabatic dispersion in the tropics can be discussed only semi-quantitatively in this paper. We therefore put less weight on the results, but include them for completeness.

Both trajectory models were used to follow the trajectories of parcels for two months, starting on January 1, 1993. Parcels were initialized uniformly in area over the 500 K isentropic surface (∼20–22 km altitude). Only small differences were found in the parcel statistics when up to half of the parcels were uniformly deleted; this verifies that the large number of parcels used (∼10,300) in each integration is sufficient to ensure convergence of the statistics. We also note that while the computed trajectories are three dimensional, we shall only be concerned here with the projection of the motion on the latitude-height (ϕ, θ) plane.

The UKMO and GSFC parcel distributions at the end of the two month period are compared in Figure 2. The figure clearly demonstrates both advection by the mean circulation and diabatic dispersion. There is ascent in the tropics, descent at the poles and a mean descent in the midlatitudes in both hemispheres. In both calculations, the diabatic dispersion at high northern latitudes in the polar vortex is weak and the largest dispersion is in midlatitudes in the northern hemisphere and in middle and high latitudes in the southern hemisphere. Note that the GSFC parcel distribution appears smoother than the UKMO distribution, which shows small scale clustering. This could be due to differences in the temporal resolution of the winds, which is 6 hours in the GSFC analyses, and 24 hours in the UKMO. The main difference between the two integrations is in the tropics, where there is less ascent and greater dispersion in the UKMO integration than the GSFC integration. The parcel distribution in the UKMO tropics also shows an equatorial dip. These differences reflect the structure of the zonal mean radiative heating rates shown in Figure 1.

The dispersion of parcels is consistent with the mechanism noted earlier of diabatic dispersion arising from parcels wandering across gradients in diabatic heating. The diabatic dispersion is especially large in the northern winter surf zone, where both the zonal mean latitudinal gradients in radiative heating and the horizontal parcel excursions are large. Parcels within the polar vortex and at low latitudes have very different dispersion characteristics compared to those at midlatitudes. This reflects the inhibition of quasi-horizontal transport across the vortex edge and across the edge of the tropics. Most of the parcels that are in these regions on day 60 have been there throughout the two months and, because latitudinal gradients in heating and latitudinal particle excursions are relatively small, the diabatic dispersion is small. The behavior in the southern hemisphere is different again. Here the dispersion is large, as might be expected from the strong latitudinal gradients in diabatic heating, but there is no clear boundary in dispersion characteristics between middle and high latitudes, reflecting the fact that there is no effective transport barrier. The strong latitudinal variation in the transport and dispersion characteristics of ensembles of parcels has also been noted by *Eluskiewicz et al.* [1995] in a similiar trajectory analysis using winds and heating rates from the GFDL SKYHI general circulation model.

Figure 3 shows some examples of the parcel trajectories projected onto the ϕ-θ

plane. A few trajectories have been highlighted for clarity. This figure illustrates the confinement of certain ensembles of parcels to different regions, as shown more clearly below in §3. For example, some parcels are confined to the tropics while others remain in the northern or southern hemisphere extratropics. GSFC trajectories that descend below 500 K rarely enter the region between about 25S and 15N, consistent with the heating rates shown in Figure 1a and with the isolation of the tropics from midlatitudes. The UKMO trajectories show a weaker tropical ascent and even descend below 500 K near the equator. Again, this is consistent with the heating rate profile in Figure 1b. In addition, the region of mean upwelling is considerably wider in the UKMO tropics, extending from about 30S to 20N. These results highlight the differences in the UKMO and GSFC residual circulations [*Coy and Swinbank*, 1996], which, as we show later, give rise to differences in diabatic dispersion. Figure 3 also illustrates the large latitudinal excursions of parcels in the surf zone and SH extratropics in both calculations. From this it is clear that parcels in the extratropics see a range of radiative heating rates as they wander across the latitudinal gradient in the heating rate profile.

3. Trajectory Ensembles

The presence of transport barriers such as the edge of the winter polar vortex, means that air parcels do not move freely on an isentropic surface over the whole globe, but tend to be confined to certain regions. This limits the range of radiative heating conditions experienced by parcels along their paths, thus parcels in different regions have distinct dispersion characteristics, as has been noted in the discussion of Figure 2. To take account of the barriers, parcels have been divided into a number of ensembles. This section describes the parcel ensembles chosen, quantifies the ensemble mean motions, and characterizes some of the aspects of the latitudinal motion that affect the diabatic dispersion.

The transport barriers give rise to four distinct regions identifiable in the trajectories: the polar vortex, the surf zone, the tropics, and the southern hemisphere extratropics. These naturally define four ensembles, each comprising parcels that stay in one of these regions throughout the two-month period. The transport barriers, however, are not impermeable and we therefore define additional ensembles containing parcels that move from one region to another. A number of different criteria have been used to assign air parcels to ensembles. Some criteria are based on PV values calculated at parcel positions using the analyzed fields, others on the latitudes visited by the parcels. Essentially, ensembles will be defined according to whether the parcels remain within a given latitude or PV range for the entire two month integration.

The polar vortex ensemble may be satisfactorily defined using PV values, since the vortex edge is clearly marked by strong gradients in PV. In the GSFC calculation, the value of PV at the vortex edge on the starting day January 1st is found using the method of *Nash et al.* [1996]. Parcels having PV greater than this value on the starting day ($\sim$45 PVU with 1 PVU=10^{-6}m^{2}s^{-1}°K kg^{-1}) are assigned to the vortex ensemble.

Alternatively, the vortex ensemble may be defined to be those parcels whose PV value, Q, remains above some fixed value $Q = Q^*$ throughout the period considered. The value of Q^* is chosen to maximize the proportion of parcels for which $Q > Q^*$ initially and throughout the period considered. The value of Q^* thus defined from the UKMO winds was found to be $\sim$35 PVU. These two different ways of defining the vortex ensemble give similiar results for the dispersion within the vortex.

In order to assign parcels to the surf zone, tropical, and southern hemisphere extratropical ensembles it is necessary to identify the tropical transport barriers. The locations of these tropical barriers are not clearly defined by analyzed PV values. PV gradients are much weaker in the tropics than at the edge of the polar vortex, and furthermore PV fields calculated from analyzed fields are less reliable in the tropics than those at higher latitudes [e.g. *Waugh*, 1997]. Two methods have been used to assign parcels to the different ensembles outside the vortex. The first takes account of the range of latitudes visited by parcels throughout the two-month period of interest. Figure 3 shows that parcels with ascent and dispersion characteristics typical of the tropics lie between 25S and 15N (GSFC) and between 30S and 20N (UKMO). Parcels that remain between these latitudes are assigned to the tropical ensemble. It is worth noting that the southern hemisphere easterly jet, which is likely to correspond to a transport barrier [*Bowman and Hu*, 1996] is located at 20–25S in the GSFC winds during this time period. GSFC parcels that remain south of 25S are assigned to the southern hemisphere extratropical ensemble (hereafter the "SH ensemble"), and those that remain north of

15N, but outside the vortex, are assigned to the surf zone ensemble.

The second method used for defining ensembles is similar to that described above for defining the polar vortex ensemble in the UKMO calculation. Approximate values of the PV corresponding to the northern and southern edges of the tropics can be found, just as for the vortex edge, though such values are less clearly defined than for the vortex edge. Parcels are assigned to a given ensemble if their PV remains within the range defined by the corresponding boundaries for the entire two months. In the UKMO integration, for example, the tropics extend from ~ -26PVU to ~ 19PVU. The relative zonal symmetry of the PV contours in the tropics means that the assignment of parcels to ensembles using either the latitude or PV give similar results for the ensemble mean motion and the ensemble dispersion. Thus in the following, results are presented for ensembles defined with respect to latitude, with the exception of the vortex ensemble. In addition, only the GSFC results will be shown explicitly; similiarities and differences with UKMO results will be pointed out as necessary.

It is worth restating that the transport barriers at the edges of the tropics are not as strong or as well defined as the vortex edge barrier. There are a range of latitudes (or PV contours) that might be chosen to define the edges of the tropics. The sensitivity of the diabatic dispersion to the choice of boundary is discussed in §4.

In addition to the main ensembles where parcels are confined to one region, two additional ensembles are defined. The first is the "poleward" ensemble, which consists of parcels that are initialized in the GSFC tropical region $[25S, 15N]$ but that do not remain there. Some of these parcels briefly visit the midlatitudes and return to the tropics, others cluster near the tropical barriers, and still others penetrate well into midlatitudes. Parcels in this ensemble, on the average, move poleward out of the tropics. The second is the "equatorward" ensemble, which consists of parcels initialized outside of the tropical region that enter the tropics at some time. Some of these cluster near the tropical barriers and move back and forth across them, others visit low latitudes, rise, and return to the midlatitudes. Corresponding ensembles can also be defined using PV values.

Having defined the ensembles it is now possible to quantify the ensemble mean motions. Figure 4 shows the ensemble mean trajectories for the GSFC integration.

The numbers near each trajectory indicate the fraction of parcels contributing to each ensemble. Note that the poleward and equatorward branches have been decomposed into northern and southern hemisphere sub-branches in order to look at hemispheric differences. The tropical ensemble has a mean ascent rate of about 0.75 K day^{-1} ($\sim 0.4°\mathrm{K\,day}^{-1}$ or $0.4\mathrm{mm\,s}^{-1}$), which agrees well with tropical ascent rates inferred from HALOE water vapor observations [*Mote et al.*,1996]. The surf zone ensemble and SH ensemble have similar mean descent rates of about 0.4 K day^{-1} ($\sim 0.3\mathrm{mm\,s}^{-1}$) and the vortex ensemble a mean descent rate of 0.7 K day^{-1} ($\sim 0.4\mathrm{mm\,s}^{-1}$) (see also [*Rosenfield et al.* 1994]). While there is little variation in the mean latitude in the SH, surf zone and tropics, there is a small southward drift of the vortex mean trajectory, which is associated with the weakening of the vortex. We expect little net transport for the global ensemble, and the mean trajectory for all parcels, shown enclosed by a circle, verifies this.

The Lagrangian mean trajectory in the UKMO surf zone is similiar to that found in the GSFC integration, but the mean descent in the UKMO SH extratropics is about half that found for the GSFC analysis. In addition, the mean ascent of about 0.25 K day^{-1} ($\sim 0.15\mathrm{mm\,s}^{-1}$) in the UKMO tropics is significantly less than in the GSFC tropics. This is as expected from the differences in the two heating rate profiles shown in Fig. 1. Little net transport for the global ensemble was also found in the UKMO calculation.

Some indication of the latitudinal extent of each ensemble is shown by the three horizontal lines that intersect each ensemble mean trajectory in figure 4. These lines indicate the standard deviation of latitude within the ensemble at different times and are centered on the ensemble mean latitude at those times. The statistics of the latitudinal displacements away from the mean latitude are nearly time independent in most cases. Statistical studies of the latitudinal displacements of individual parcels within the surf zone for example indicate that parcels that start in the same latitude band spread nearly uniformly across the entire surf zone in about one week or less. This is consistent with the idea that tracers are likely to be effectively homogenized within the surf zone.

The statistics for the poleward ensemble are however not stationary in time, and in this case the variance of latitudinal displacements increases approximately linearly with time for most of the two month period, indicating that the latitudinal dispersion is diffusive. This is important in interpreting the diabatic dispersion of these parcels and a relevant theoretical model for the dispersion is discussed in §5.

The pattern of ensemble mean trajectories and the proportions of parcels in each ensemble shown in Fig. 4 illustrates the "semipermeability" of the tropical barrier. It appears somewhat more difficult for parcels to enter the tropical region than to leave, consistent with the Brewer-Dobson circulation in the lower stratosphere. 73% of the parcels initialized outside the vortex and north of 15N remain north of 15N for the entire 2 months, and 66% of parcels initialized south of 25S remain in that region. However, only 29% of parcels initialized in the tropical region remain there, with 41% leaving the region by crossing the 15N boundary.

4. Quantifying the Diabatic Dispersion

The diabatic dispersion about each of the Lagrangian mean paths discussed earlier can be characterized and quantified by considering the time evolution of the variance of potential temperature within each ensemble. In a purely Lagrangian context, it is standard to refer to the dispersion as "advective" when the displacement variance increases quadratically with time and as "diffusive" when the variance increases linearly with time [e.g. *Van Kampen*, 1981]. The former occurs when, for example, the dispersion is dominated by large scale differential advection, as discussed below. The latter occurs, for example, in the case of shear dispersion mentioned in §1. In addition, we will identify the dispersion as due to "transient shear dispersion" when the variance increases cubically with time [*Tennekes and Lumley*, 1972]. Some further aspects of these three different types of dispersion are considered in §5, including a discussion of the less well-known transient shear dispersion (see also *Young et al.*, [1982, §5] and references). At this stage the regimes simply provide a useful framework in which to quantify the dispersion. In particular in the diffusive regime we can define a diffusion coefficient, following convention, as half the time rate of change of the variance. We discuss the relevance to tracer transport in §6.

In what follows, we use angle brackets $\langle\cdot\rangle$ to indicate an ensemble mean, and the ensemble variance of a Lagrangian quantity $\psi(t)$ will be denoted by $\langle\delta\psi(t)^2\rangle \equiv \langle[\psi(t) - \langle\psi(t)\rangle]^2\rangle$. The time evolution of the potential temperature variance

$\langle \delta\theta(t)^2 \rangle$ for each ensemble in the GSFC integration is plotted in Figure 5. Also shown is $\langle \delta\theta(t)^2 \rangle$ for the entire set of parcels. The different ensembles show a variety of dispersive behavior, which can be seen clearly by comparison with the dotted straight lines proportional to t, t^2 and t^3.

Before considering the ensembles individually we briefly consider the dispersion of the entire global ensemble of parcels. The variance $\langle \delta\theta(t)^2 \rangle$ increases quadratically with time, being dominated by large-scale differential advection associated with the vertical separation of parcels in the rising and descending branches of the mean diabatic circulation. This is further discussed in the final section.

The dispersion for the surf zone ensemble is initially advective ($\langle \delta\theta(t)^2 \rangle \sim t^2$) but becomes diffusive ($\langle \delta\theta(t)^2 \rangle \sim t$) on a timescale of about one month. The same is true for the SH extratropics. Numerical estimates for the diffusivities depend to some extent on how the tropical boundaries are defined and will be discussed in the next section. The diffusive limit is reached sooner for the surf zone ensemble than for the global ensemble because the horizontal motion within the surf zone is unconstrained by transport barriers and the parcels move rapidly across the latitudinal gradient in heating rate. The UKMO integration gives similar diffusion coefficients in the surf zone, but somewhat smaller diffusivities in the SH extratropics, probably because the southern tropical boundary is closer to the south pole and the parcels experience a smaller range of heating rates.

Dispersion within the polar vortex is similar in the UKMO and GSFC integrations, is more than an order of magnitude smaller than in the surf zone and SH ensembles, and is not clearly described by a simple power law. This is due to the fact that the statistics of diabatic heating experienced by parcels in the vortex is more nonstationary than in the other cases. The motion of parcels within the polar vortex is sensitive to the large intermittent distortions of the vortex which break the time translation invariance of the statistics. In the GSFC calculations shown here, there is also an increase in the vortex dispersion towards the end of the two month period. This occurs because parcels were assigned to the vortex based only on their initial PV. Some vortex parcels near the vortex edge have escaped to lower latitudes by vortex erosion or weakening of the barrier. A vortex ensemble defined using the parcel PV throughout the integration does not show the same increase in dispersion.

In both GSFC and UKMO calculations the diabatic dispersion in the tropics is smaller than in the midlatitudes. In addition, the dispersion in the GSFC tropics shown in Fig. 5 is well-approximated by the power law $\langle \delta\theta(t)^2 \rangle \sim t^{3/2}$ over most of the period, therefore not reaching the diffusive regime on this time scale. This non-integer power law might well be a long-lived transient connecting the advective and diffusive dispersion regimes. One might be tempted, from the power law, to suppose that the underlying process is a "fractional Brownian motion" [*Shlesinger et al.*, 1995], but we note that there is no physical basis for this here.

The tropical dispersion in the UKMO integration is larger than the GSFC tropical dispersion and the approximate $t^{3/2}$ behavior is not found. Instead, the UKMO tropical dispersion becomes diffusive towards the end of the integration period, with a diffusivity of about 0.1 $\mathrm{K}^2\,\mathrm{day}^{-1}$ ($0.003\,\mathrm{m}^2\mathrm{s}^{-1}$). This is well below the upper bound of 0.03–0.09$\mathrm{m}^2\mathrm{s}^{-1}$ estimated from the "taperecorder" signal in tropical water vapor [*Mote et al.*, 1996]. The larger UKMO tropical dispersion, relative to that in the GSFC tropics, and the fact that it becomes diffusive after about 2 months, are consistent with the greater spatial variability in the UKMO tropical diabatic heating rates discussed earlier.

The poleward ensemble of parcels in the GSFC calculation has a dispersion typical of transient shear dispersion ($\langle \delta\theta(t)^2 \rangle \sim t^3$). The dispersion of parcels moving north out of the tropics is similiar to the dispersion of those that move south. The cubic rate of increase results from parcels, initially confined in latitude, spreading out across the latitudinal gradient in heating rate. As the parcels spread, the range of heating rates seen by the ensemble increases with time, and the cubic rate of increase is due in part to this time dependence of the heating rate statistics. (A relevant mathematical model is discussed in more detail in §5.) It is important to note that the UKMO results for this case do not show a cubic rate of increase in the variance, but instead show advective dispersion $\langle \delta\theta(t)^2 \rangle \sim t^2$. The reasons for this disagreement are not related in an obvious way to the differences in the UKMO and GSFC residual circulations mentioned earlier, and seem to depend, rather, on the effects of eddies on parcel trajectories.

The heating rate statistics for parcels in the equatorward ensemble are more stationary than the statistics for the poleward ensemble, and more nearly like the statistics in the surf zone and SH extratropics. Figure 5 shows that the rate of increase of $\langle \delta\theta(t)^2 \rangle$ for parcels in the equatorward ensemble, which rise on the average, is similar to that in the surf zone and SH extratropics, where there is mean descent.

Sensitivity of diffusivity to tropical boundaries

It has been noted previously that the subtropical transport barriers are more difficult to define than at the edge of the polar vortex. In this section we show the sensitivity of the estimated diffusivity to the location chosen for the subtropical barriers. The study is presented with the barrier chosen by latitude. Similar results are found if the ensembles are defined with respect to PV by the method discussed earlier. The northern and southern tropical boundaries are denoted by ϕ_N and ϕ_S, respectively; recall that the SH extratropical ensemble is by definition that group of parcels that remain south of ϕ_S for the entire two months, and the surf zone ensemble is the group outside the polar vortex that remain north of ϕ_N. Initially we concentrate on the dispersion in the GSFC calculation. Up to this point we have only considered ϕ_S=25S and ϕ_N=15N, which we have argued are the most natural choices for the tropical boundaries based on the parcel trajectories in the GSFC calculation (cf. Figs. 2a and 3a).

Figure 6 shows the behavior of the function $\langle \delta\theta(t)^2 \rangle / 2t$ for several values of ϕ_S and ϕ_N. This function increases linearly with time if the dispersion is advective, and in the long time diffusive regime approaches a constant, which we denote by $K_{\theta\theta}$. Its significance will be discussed in the next section. The main point here is to illustrate how restricted transport between the tropics and midlatitudes affects the character of the vertical dispersion.

The change in the rate of increase of $\langle \delta\theta(t)^2 \rangle / 2t$ shows that the time to reach the diffusive regime increases as the tropical boundary is moved equatorward. This occurs because as the boundary moves equatorward, an increasing number of tropical, ascending trajectories are added to the descending extratropical trajectories, and the dispersion therefore becomes more advective. This change in the character of the dispersion occurs to a lesser extent in the northern hemisphere, and we estimate that the dispersion is diffusive only after about three months for parcels moving throughout the entire northern hemisphere outside the vortex ($\phi_N = 0$). The vertical dispersion of parcels moving throughout the entire Southern hemisphere ($\phi_S = 0$) is clearly not diffusive on seasonal timescales. These hemispheric differences most likely reflect the greater permeability of the northern tropical barrier relative to the southern barrier, which results in parcels moving more freely between regions of net heating and cooling compared to the southern hemisphere.

The main conclusion from the UKMO calculations is the same: as the latitude defining the tropical barrier is moved equatorward the dispersion becomes more advective. There are, however, some differences in the details. Firstly, in the SH the dispersion is diffusive only if parcels remaining south of 35S are considered. This reflects the broader tropics in the UKMO calculation compared to the GSFC calculation. Secondly, global and hemispheric dispersion, after two months, is less in the UKMO calculation than in the GSFC calculation. This results from the smaller tropical ascent rate in the UKMO calculation. Because the difference between the tropical ascent and high and middle latitude descent is smaller in the UKMO case, the dispersion due to differential advection is less.

Both UKMO and GSFC calculations lead to estimates for the extratropical diffusivity that are in the range $K_{\theta\theta}$ ~2–6 $\mathrm{K^2\,day^{-1}}$, depending on how the tropical boundaries are defined.

5. Theoretical Models of Dispersion

In this section we review some of the well-established theoretical models for dispersion and comment on how they apply to diabatic dispersion in the stratosphere. Firstly we discuss the conditions under which parcel dispersion is advective or diffusive. Secondly, in a separate subsection, we discuss the case of transient shear dispersion.

The fact that particle dispersion in a variable velocity field becomes diffusive on sufficiently long timescales was first discussed by *Taylor* [1921] in the context of dispersion in homogeneous and stationary turbulent flow and is discussed in standard turbulence textbooks [e.g. *Tennekes and Lumley*, 1972]. The standard ideas are all relevant here, provided that potential temperature is taken to represent displacement and heating rate to represent velocity.

The potential temperature of a parcel is given by

$$\theta(t) = \theta_o + \int_0^t W(t')dt' \tag{1}$$

where $W(t)$ is the parcel vertical velocity in isentropic coordinates, i.e. the diabatic heating rate along the parcel trajectory. All parcels are assumed to have the same initial potential temperature θ_o.

The difference between advective dispersion and diffusive dispersion can be explained in terms of the loss of memory along parcel trajectories. This loss of memory is characterized by the heating rate Lagrangian autocorrelation function (hereafter LAF) $\mathcal{C}_W(t,\tau)$ defined by

$$\mathcal{C}_W(t,\tau) = \frac{\langle W(t)W(t+\tau)\rangle - \langle W(t)\rangle \langle W(t+\tau)\rangle}{\sigma(t)\sigma(t+\tau)} \tag{2}$$

where $\sigma(t) = \langle \delta W^2(t)\rangle^{1/2}$. The decrease in $\mathcal{C}_W(t,\tau)$ as a function of the time lag τ, for different starting times t, is a measure of the rate at which a parcel loses the memory of its initial heating rate.

For the purpose of this discussion, it is convienient to assume that the second order statistics of the process $W(t)$ are time translation invariant. This means that the one-point statistics $\langle W(t)\rangle$ and $\langle \delta W^2(t)\rangle$ are assumed to be time independent, and that the two-point statistic $\mathcal{C}_W(t,\tau)$ depends only on the magnitude of the time lag τ. More generally, a stochastic process is called "stationary" if all statistics are time-translation invariant. If $W(t)$ were Gaussian-distributed, then the stated assumptions would imply stationarity, since a Gaussian distribution is completely characterized by second order statistics.

The time translation invariance of the second order statistics depends on the ensemble. For example, this appears to be an excellent approximation in the SH and is reasonable in the surf zone. (However a substantial fluctuation in the heating rate statistics in the surf zone occurred near day 40 of the integration, which was associated with a minor warming.) Stationarity does not seem to hold within the polar vortex where the parcels are particularly sensitive to the temporally intermittent distortions of the vortex.

If we assume that $W(t)$ is stationary, then $\mathcal{C}_W(t,\tau)$ depends only on the time lag τ and the variance of the potential temperature distribution at time t can be written as

$$\langle \delta\theta(t)^2 \rangle = 2\langle \delta W^2 \rangle t \int_0^t d\tau \; (1 - \tau/t)\mathcal{C}_W(\tau). \tag{3}$$

If $\mathcal{C}_W(\tau)$ tends to zero sufficiently fast as lag time τ increases it is possible to define a correlation time τ_L

$$\tau_L = \int_0^\infty \; \mathcal{C}_W(\tau)\, d\tau \tag{4}$$

Parcel heating rates, or analogously vertical velocities, along a trajectory become nearly independent when separated by a time interval τ_L or longer. On timescales longer than τ_L the cross-isentropic parcel motion can be thought of as a series of statistically independent steps of a random walk. Within the surf zone, τ_L is on the order of the time for a displacement across midlatitudes, e.g. due to a breaking planetary wave.

The nature of the dispersion depends on the overall timescale of the dispersion relative to the correlation time, and the potential temperature variance has the following limiting behavior

$$\begin{aligned} \langle \delta\theta(t)^2 \rangle \;\; &\rightarrow \;\; \langle \delta W^2 \rangle\, t^2 \qquad\qquad t \ll \tau_L \\ &\rightarrow \;\; 2\,\langle \delta W^2 \rangle\, \tau_L\, t \qquad t \gg \tau_L. \end{aligned} \tag{5}$$

Thus the dispersion is advective on timescales much shorter than the correlation time and is diffusive on timescales much longer than the correlation time. In the latter case a diffusivity $K_{\theta\theta}$ may be defined by

$$K_{\theta\theta} \equiv \lim_{t\to\infty} \frac{\langle \delta\theta(t)^2 \rangle}{2\,t} = \langle \delta W^2 \rangle \tau_L \tag{6}$$

provided that the limit exists. We use the term "diffusion" here in the sense normally applied to random walks. A discussion of the conditions under which this is relevant to flux-gradient relationships and tracer transport is delayed until §6. Thus when the diabatic heating rate statistics are stationary, or approximately so, the vertical dispersion is diffusive on timescales that are much longer than the time a parcel retains the memory of diabatic heating along its path. It is this loss of memory of radiative heating along parcel trajectories that gives rise to irreversible vertical dispersion. Note that within the diffusive regime, for the same variance $\langle \delta W^2 \rangle$, the diffusivity increases with the correlation time. This has to do with the "persistence" of the random walk — a random walker who frequently changes direction (small τ_L) does not get very far.

We can now understand why diabatic dispersion appears diffusive in some cases and not in others. Figure 7 shows some examples of $\mathcal{C}_W(\tau)$, which has been obtained by averaging Eq. 2 over t. $\mathcal{C}_W(\tau)$ for the entire ensemble decays slowly and we expect diffusion only on seasonal timescales or longer. The relatively slow decay of $\mathcal{C}_W(\tau)$ for the global ensemble reflects the fact that parcels within regions of net heating or cooling tend to remain separated on the timescale of two months. Diffusive dispersion will result only when parcels forget which of these regions they started in. The decay is more rapid in the surf zone and an estimate based on Eq. 4 gives τ_L=10 days, which is comparable to the time it takes for air parcels to spread throughout the surf zone. The correlation time in the SH is also about 10 days, however in the SH $\mathcal{C}_W(\tau)$ is represented to an excellent approximation by the exponential function e^{-t/τ_L}, indicated by the dashed line in the figure. The simpler behavior of $\mathcal{C}_W(\tau)$ in the southern extratropics relative to the surf zone reflects the greater degree of statistical stationarity of $W(t)$ in the southern hemisphere during this time period.

Transient shear dispersion

The heating rate statistics of parcels in the poleward ensemble are not stationary over the two-month period of interest. We have found that the ensemble variance in the heating rate increases approximately linearly in time over most of the two months for parcels that move either north or south out of the tropics. The latitude variance also increases nearly linearly with time, and as mentioned earlier, the potential temperature variance increases like t^3.

The cubic rate of increase of the variance is consistent with the following simple model, in which parcels perform a random walk in the horizontal across a horizontal gradient in vertical velocity. Let y be a horizontal coordinate and z a vertical coordinate and consider the motion in the (y, z) plane of a parcel advected by the velocity field (v, w)

$$\begin{aligned} dy/dt &= v(y(t)) \qquad y \in (-\infty, \infty) \\ dz/dt &= w(y(t)) \qquad z \in (-\infty, \infty) \end{aligned} \tag{7}$$

For simplicity, we assume that the vertical velocity w is independent of z and the horizontal velocity v is zero mean Gaussian white noise with autocorrelation $\langle v(t)v(t')\rangle = D\delta(|t-t'|)$. The stochastic process $y(t)$ is then a simple random walk ("Wiener process") with autocorrelation

$$\langle y(t)y(t')\rangle = 2D\, min(t, t') \tag{8}$$

and variance $\langle y^2(t)\rangle = 2Dt$. Thus the ensemble motion along the y axis is diffusive with diffusion coefficient D.

As a further simplification, we take $w(y) = \alpha y$, where α is a constant. We can then write the variance of z as

$$\langle z^2(t)\rangle = \alpha^2 \int_0^t dt' \int_0^t dt''\ \langle y(t')y(t'')\rangle\, dt'' \tag{9}$$

Substitution of the expression for the autocorrelation $\langle y(t)y(t')\rangle$ from Eq. 8 and performing the integration gives

$$\langle z^2(t)\rangle = 2\alpha^2 Dt^3/3 \tag{10}$$

Thus the vertical dispersion is proportional to the horizontal diffusivity D and the square of the horizontal shear α and increases like t^3. The rapid dispersion occurs because the variance of the vertical velocity $\langle \delta w^2\rangle$ is not constant as it would be under stationary conditions. Because w$\propto$ y, $\langle \delta w^2\rangle$ also increases linearly with time as parcels diffuse across the vertical velocity gradient. In a bounded domain $y \in [-L, L]$, the variances $\langle y^2\rangle$ and $\langle \delta w^2\rangle$ will level off and eventually the diffusive limit $\langle z^2\rangle \propto t$ will be reached. Thus the dispersion of the poleward ensemble can eventually become diffusive on a timescale that is longer than the time for the parcels to spread uniformly across the midlatitudes. This transient shear dispersion phenomenon has also been discussed in the context of ocean mixing [*Young et al.* 1982].

We can verify that Eq. (10) is approximately satisfied. Calculation of the potential temperature variance for parcels moving north out of the tropics, for example, gives $\langle \delta\theta(t)^2 \rangle \sim 0.001 t^3$ for times $t > 10$ days. Thus from Eq. (10) we expect $2\alpha^2 D/3 \sim 0.001$. From the latitude statistics for this group of parcels, we can derive a horizontal diffusivity $D \sim 5 \times 10^5 \mathrm{m}^2\mathrm{s}^{-1}$ (here, D is the meridional diffusivity). An estimate for the midlatitude heating gradient is $\alpha \sim \partial H/\partial\phi \sim (1.5\ \mathrm{K\ day}^{-1})/(50\ ^\circ$ latitude) (cf Fig. 1). These estimates give a value of $2\alpha^2 D/3 \sim 0.002$, which is consistent with the value of 0.001 found from the potential temperature statistics. Similiar agreement is found for parcels moving south out of the tropics.

6. Discussion

We have presented calculations of parcel trajectories in the lower stratosphere that demonstrate how differences in radiative heating along the trajectories results in vertical dispersion. This "diabatic dispersion" depends on large scale transport barriers that restrict the range of heating rates seen by the parcels. During northern hemisphere winter, the globe is naturally partitioned into four regions: the southern hemisphere extratropics, the northern hemisphere surf zone, the polar vortex and the tropics. Within each of these regions, the dispersion depends on the spatial variability of the diabatic heating and the characteristics of the horizontal motion. We have characterized the dispersion by considering the time evolution of the potential temperature variance for those parcels that are confined to these regions, as well as for those that move between regions.

Two independent calculations give similiar results in the northern hemisphere surf zone and southern hemisphere extratropics. Within these regions, the dispersion can be described as diffusive after 3–4 weeks, with a diffusivity $K_{\theta\theta}$ in the range 2–6 $\mathrm{K}^2\,\mathrm{day}^{-1}$, depending on how the tropical boundaries are defined. In terms of log pressure height, this is equivalent to a diffusivity $K_{zz} \sim 0.1\,\mathrm{m}^2\mathrm{s}^{-1}$, consistent with that found in model simulations [*Plumb and Mahlman*, 1987; *Kida*, 1983].

The dispersion in the UKMO southern extratropics is somewhat smaller, most likely because the region, as defined by transport barriers, is narrower than in the GSFC analysis. The two calculations also give similiar results for the dispersion within the polar vortex. Here, the dispersion after two months is an order of magnitude smaller than in the extratropics, and cannot described as a diffusive process. On the two month timescale, the potential temperature dispersion about the global mean is advective. This reflects the restricted transport between tropical regions of mean upwelling and extratropical regions of mean descent.

In the UKMO tropics the dispersion is an order of magnitude smaller than in the midlatitudes, while in the GSFC tropics it is almost two orders of magnitude smaller. The tropical dispersion should be viewed as a lower bound in both calculations because the effects of clouds were neglected in the GSFC radiative calculation and were included only in a climatological sense in the UKMO calculation. Underlying patches of temporally and spatially varying cloud can give rise to an extra source of variability in the radiative heating and thus to greater diabatic dispersion.

It is not clear whether the discrepancy between the two integrations in the tropics is due primarily to differences in the radiative heating calculations, the temperature profiles, or the horizontal winds and the location of the tropical barriers. These are related, and the differences found here reflect the disagreement in the residual circulations of the GSFC and UKMO analyses in the tropics and subtropics [*Coy and Swinbank*, 1996]. This is the region where the geostrophic constraint is small and where there are relatively few observations and so the analyses are likely to reflect differences in the GCMs used in the assimilation process.

In much of this paper we have emphasized the role of the latitudinal gradient in diabatic heating as the primary source of vertical dispersion. In reality, the zonal and temporal variability in diabatic heating also contribute to the dispersion. Additional studies were performed that were aimed at isolating the effects of zonal and temporal fluctuations. This was done by interpolating time constant or zonal mean heating rates to the horizontal parcel paths, which were not changed, and recomputing the dispersion. The expectation was that the diffusivity $K_{\theta\theta} = \langle \delta \mathrm{W}^2 \rangle \tau_L$ would decrease from that in the full calculation, because of the decrease in $\langle \delta \mathrm{W}^2 \rangle$ due to the removal of zonal or temporal variation. In practice, some examples were found where $K_{\theta\theta}$ increased, presumably because of an increase in τ_L. Such an increase in τ_L might arise due to a decrease in the spatial variability of the diabatic heating field. The maximum decrease in $K_{\theta\theta}$ found was about 40%. There is therefore no doubt that latitudinal variation in diabatic heating accounts for at least half the diabatic dispersion, with a significant contribution from zonal and temporal variations.

In this paper, we have adopted a purely Lagrangian view of the dispersion, and have referred to the dispersion as "diffusive" when the potential temperature variance of an ensemble of parcels increases linearly with time. We have shown that this occurs on a timescale that is longer than the time for parcels to lose the memory of diabatic heating. The loss of memory occurs on a timescale τ_L which is about 10 days in both the northern and southern extratropics. For times longer than this, on the order of one month for the time period studied here, the potential temperature variance increases linearly with time and a diffusion coefficient can be defined. The loss of memory means that the parcel positions become independent of the initial conditions and the dispersion is irreversible.

Given that a diffusion coefficient can be defined for an ensemble of parcels the issue remains as to whether the time evolution of a tracer field can be described by a diffusion equation, or equivalently, whether a flux-gradient relation exists for the tracer transport. While the existence of a flux-gradient relation implies a linear increase in displacement variance (for constant diffusivity), the converse is not necessarily true. Describing the evolution of a tracer by a diffusion equation is appropriate only when the characteristic scale of the inhomogeneity in the tracer is much larger than the equivalent mean free path or step of the random walk (see *Corrsin* [1974]). This step size is $\sqrt{\langle \delta W^2 \rangle}\tau_L$, and in the surf zone is on the order of 5 K ($\sim$ 200m). Thus a diffusion equation for the evolution of tracer structures will be appropriate only for vertical scales $\gg 200m$. In addition, simple diffusion will not be an appropriate dispersion model if the tracer chemical lifetime is much smaller than the correlation time τ_L.

The statistical properties of the diabatic heating and horizontal wind fields relevant to diabatic dispersion are slowly varying in the vertical range 400–600 K . This leads to a single value for $K_{\theta\theta}$ in this range, and can be exploited to estimate the diffusivities from the Eulerian fields. For example, in the northern hemisphere surf zone, we may assume that the vertically diffusing tracer is horizontally well-mixed. In this case, the Lagrangian variance $\langle \delta W^2 \rangle$ in Eq. (6) is given to a good approximation by the spatial variance of the Eulerian diabatic heating field $H(\lambda,\phi,\theta)$. Thus $\langle\delta W^2\rangle \sim \overline{\delta H^2} = \overline{(H-\overline{H})^2}$, where the overbar indicates time averaging and spatial averaging over the surf zone. The correlation time τ_L is on the order of the time for a parcel to traverse the surf zone, thus $\tau_L \sim \ell/V$ where $\ell \sim 40°$ latitude is approximately the size of the surf zone, and $V \sim (\overline{(v-\overline{v})^2})^{1/2}$ is the rms meridional velocity. During the winter period Jan/Feb 93, V=5.5 m s^{-1} and $\overline{\delta H^2} = 0.42$ K ^{2}day^{-2} in the northern hemisphere between 15N and 60N on the 500 K surface. The above estimates then lead to a correlation time τ_L=9.4 day and diffusivity $K_{\theta\theta} \sim \overline{\delta H^2}\tau_L = 3.9$ K ^{2}day^{-1}, which agrees very well with the values found previously from the Lagrangian calculations. Eulerian estimates for $K_{\theta\theta}$ at higher altitudes give $K_{\theta\theta} \sim 20$K^{2}day^{-1} at $\theta = 800$ K and $K_{\theta\theta} \sim 60$K^{2}day^{-1} at $\theta = 1000$ K , although we note that at these higher altitudes the statistical properties of the winds and heating are changing much more rapidly with height.

What are the implications of the results reported in this paper for conventional chemical transport models? Firstly, the fact the dispersion of the global ensemble is advective over the two-month period studied here throws further doubt on the relevance, certainly on a seasonal time scale, of one-dimensional models of the type studied by *Holton* [1986] and *Mahlman et al.* [1986] in which the only transport is vertical diffusion. *Plumb* [1996] has noted the important role of transport barriers in limiting the validity of such 'global diffuser' models. Certainly in our calculations the transport barrier between tropics and surf-zone plays an important role in preventing a transition from advective to diffusive dispersion of the global ensemble.

Turning to two-dimensional zonally-averaged models, the results allow examination of the usual assumption that cross-isentropic advective transport dominates cross-isentropic diffusive transport, [e.g. *Tung*, 1982, see also *Holton et al.*, 1995]. For example, using the estimates for $K_{\theta\theta}$ and the average extratropical heating rate $\langle W\rangle =$ 0.4 K day^{-1}, we find that the ratio of the standard deviation to the mean potential temperature displacement, $\sqrt{K_{\theta\theta}t}/\langle W\rangle t$, is of order unity after two months. At this time, both quantities are equal to about 25K, equivalent to about 1 Km in geometric height. Thus on time scales comparable to or less than 2 months (or vertical length scales comparable to or less than about 1 Km) vertical transport is not well characterized by a mean advective transport. On the other hand, for larger time scales or vertical length scales the advective transport dominates.

Note that while many two-dimensional models omit any explicit cross-isentropic diffusion in their transport parameterizations, that is not to say that the phenomenon of diabatic dispersion is not present in such models. The combined effect of the horizontal transport, e.g. along-isentrope diffusion, and the latitudinally varying cross-isentropic advection will almost certainly give rise to diabatic dispersion. Such an effect will, of course, be augmented by any explicit cross-isentropic diffusion that might in some models be included in order to parameterize the effects of longitudinal variations in the diabatic heating field.

Vertical diffusivities of the same order of magnitude ($\sim 0.1\,\mathrm{m^2s^{-1}}$) as those predicted by our diabatic dispersion calculations have been inferred from the dispersion of volcanic aerosols [*Shibata et al.*, 1984]. Preliminary calculations for a northern hemisphere summer period indicate a dispersion that is several times smaller than that found during northern hemisphere winter, consistent with the seasonal dependence found by *Shibata et al.* [1984]. However, as noted in the Introduction, diabatic dispersion is one of two processes that can lead to cross-isentropic dispersion of a tracer such as aerosol. The other is small scale vertical mixing associated with thin turbulent layers which are thought to arise from breaking inertio-gravity waves [*Dewan*, 1981; *Woodman and Rastogi*, 1984; *Fukao et al.*, 1994]. Observations of cross-isentropic tracer dispersion cannot by themselves be used to estimate the magnitude of the two processes independently. [1]

Dewan (1981) has argued that cross-isentropic transport through many encounters with turbulent layers, which are localized and highly intermittent, both spatially and temporally, may be represented as a diffusive process. The studies of *Woodman*

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[1] There is an interesting contrast here between atmosphere and ocean. There is no radiative transfer in the ocean and dispersion across isopycnal surfaces is accomplished entirely by turbulence associated with gravity wave breaking events. The associated diffusivity has recently been estimated by *Ledwell et al.* [1994] using the observed dispersion of an inert tracer over a period of months.

and Rastogi [1984] and *Fukao et al.* [1994] estimate corresponding effective vertical diffusivities of 0.1–1 $\mathrm{m^2 s^{-1}}$. If these estimates are correct then this process would be at least as important as diabatic dispersion for the cross-isentropic dispersion of tracers. There is an important distinction between the two processes, however, that it is only the encounters with turbulent layers that give rise directly to mixing on the molecular scale [*Lilly et al.*, 1974]. Two chemically active tracers at initially different potential temperatures cannot be brought rapidly into reactive contact by diabatic dispersion alone. This distinction gives the possibility of estimating the effective vertical diffusivity due to encounters with turbulent layers, excluding the effects of diabatic dispersion, by considering evidence for mixing, or the lack of it, in tracer microstructure. Such an estimate has recently been obtained from an analysis of midwinter aircraft data by [*Balluch and Haynes*, 1997], and upper bounds on the vertical diffusivity were found to be in the range $0.01 - 0.02\,\mathrm{m^2 s^{-1}}$. It is possible that the tracer structures studied in this analysis had encountered an anomalously small number of turbulent events, so that the diffusivity estimates are anomalously small. Certainly [*Bacmeister et al.*, 1996] find little evidence of breaking gravity waves in tracer spectra derived from a fairly extensive set of aircraft observations, suggesting that turbulent events are highly intermittent and therefore that the time between encounters of a given air parcel with such events may be rather long. But if the above estimate stands up to scrutiny then it suggests that diabatic dispersion makes the dominant contribution to cross-isentropic dispersion, at least in the winter season.

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Figure 1. Zonal mean diabatic heating rate on 1 Feb 93, on the 500 K surface. Heating rate is expressed as a rate of potential temperature change (K/day). (a) Calculated using GSFC radiation scheme and GSFC temperatures; (b) MIDRAD radiation scheme and UKMO temperatures. The heating rate at each zonal gridpoint is overplotted.

Figure 2. Parcel positions after 2 months projected onto the θ–ϕ plane, (a) GSFC, (b) UKMO. The dotted lines at 25S and 15N in (a) and 30S and 20N in (b) are (approximate) tropical boundaries.

Figure 3. Sample parcel trajectories $(\phi(t), \theta(t))$ over the 2 month period Jan/Feb 1993, (a) GSFC and (b) UKMO calculations. In both (a) and (b), 100 trajectories were chosen at random from the total ensemble of over 10,000. The dashed lines are as in Fig. 2.

Figure 4. Lagrangian mean trajectories for each GSFC ensemble. The length of the horizontal lines intersecting each trajectory is twice the standard deviation of the latitude distribution and is shown at times 7, 25, and 52 days; for clarity, the lines for the poleward branches are indicated by dotted lines. The vertical dotted lines delineate the tropics; the fractional areas of the tropics and extratropics are indicated near the bottom of the figure. The fraction of parcels contributing to each ensemble is also shown.

Figure 5. The time evolution of the potential temperature variance for the global ensemble and each subensemble in the GSFC calculation. Various powers of t are indicated on the graph for comparison.

Figure 6. The effect of the location of the tropical boundaries ϕ_S and ϕ_N on the time evolution of the extratropical dispersion. (a) $\langle \delta\theta(t)^2 \rangle$/2t for parcels remaining south of ϕ_S, for various ϕ_S; (b) $\langle \delta\theta(t)^2 \rangle$/2t for parcels remaining north of ϕ_N, for various ϕ_N. The diffusivity $K_{\theta\theta}$is defined by $K_{\theta\theta} = \lim_{t\to\infty} \frac{\langle \delta\theta(t)^2 \rangle}{2t}$.

Figure 7. Diabatic heating LAF $\mathcal{C}_W(\tau)$ for surf zone, SH and global ensembles. The dotted line is the exponential function exp(-t/10) which is an excellent approximation to $\mathcal{C}_W(\tau)$ in the SH.