Transport, stirring and mixing in the atmosphere

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1 Introduction

1.1 Outline

Our understanding of atmospheric transport has increased considerably over the last twenty years or so. There have been important theoretical advances, often prompted by enormous improvements in the observations, both of dynamical quantities and of chemical species, and in numerical models. The combined use of numerical models and observations has been particularly important, especially in the last decade. At the same time, there has been much interest in the application of results of dynamical systems theory to fluid dynamics under the heading of 'chaotic advection'. [See e.g. the textbook by *Ottino* (1989).] Such results are of undoubted qualitative relevance to atmospheric flows, but nonetheless there are relatively few specific examples where insight from chaotic advection has lead to concrete, quantitative insight into atmospheric transport.

This article will review our current understanding of atmospheric transport, stirring and mixing and identify areas where insight from theories of chaotic advection may be quantitatively useful. The article is intended to be of interest to a reader who has some knowledge of dynamical systems and chaotic advection and wishes to learn more about possible applications to the real atmosphere.

The remainder of this section will summarize relevant aspects of our knowledge of the observed state of the atmosphere. Then in §2 some of the ideas underpinning our understanding of large-scale atmospheric dynamics will be reviewed, focussing on those that

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have particular relevance to characterising the transport, stirring and mixing problem in the atmosphere. The two important features that emerge are (i) that the stable stratification tends to inhibit vertical motion, so some important aspects of atmospheric transport may be captured by considering only quasi-horizontal motion along stratification surfaces (in fact, as explained in §2, isentropic surfaces) and (ii) that in many cases the large-scale flow gives the dominant contribution to advection. (i) and (ii) together mean that two-dimensional chaotic advection provides a useful paradigm for certain aspects of atmospheric transport. §3 considers observations and models of quasi-horizontal transport, stirring and mixing along isentropic surfaces, which is the part of the overall transport, stirring and mixing problem to which the application of chaotic advection ideas seems most promising. §4 considers some of the transport, stirring and mixing issues to which vertical transport and vertical structure are central. §5 summarises and identifies some areas where application of the mathematical of chaotic advection and dynamical systems may give greater understanding of the real atmosphere.

1.2 Observational background

We shall be primarily concerned here with transport, stirring and mixing due to the large-scale flow in the troposphere and stratosphere. The troposphere is the part of the atmosphere below about 10km (higher in the tropics, lower at the poles) where the temperature decreases with height. The stratosphere is the part of the atmosphere above the troposphere, and below about 50km, in which the temperature increases with height. (The temperature variation with height has important implications for the stability to vertical displacements - see later.) Books such as *James* (1994), *Holton* (1992), and *Andrews et al.* (1985) are dynamical texts (the last is primarily concerned with the stratosphere) which also give useful summaries of the observed state of the atmosphere.

It is perhaps useful to point out at the beginning that in many respects the atmosphere is relatively well-observed. [See, for example, *James* (1994), §2.2.]. The requirements for reliable weather forecasts have lead to an extensive network of observing stations, giving improved knowledge of the winds, temperatures etc. in the troposphere and lower stratosphere. These observing stations are, of course, concentrated in the continental areas of the Northern Hemisphere and coverage in the Southern Hemisphere in particular is extremely sparse. More uniform geographical coverage has been provided by a number of satellite-based instruments used to measure temperature, water vapour, ozone and other quantities. This satellite data has not had quite the impact on weather forecasts that one might have expected, since in the troposphere the vertical resolution of the data tends to be rather poor and the presence of clouds also causes problems. However satellite data has had an enormous impact on our knowledge of the stratosphere.

Besides 'operational' satellites, which make measurements on a routine basis, there have been a number of important satellites that operate for a restricted period. For example, the Upper Atmosphere Research Satellite (UARS), launched in 1991, has measured a large range of different trace chemicals, such as nitrous oxide (N_2O), nitric acid (HNO_3) and chlorine monoxide (ClO), as well as ozone (O_3), water vapour (H_2O), temperature, and aerosol. See, for example, the recent issues of Journal of Atmospheric Sciences (volume 51, number 20) or Journal of Geophysical Research (volume 101, number D6), devoted to UARS observations and their interpretation.

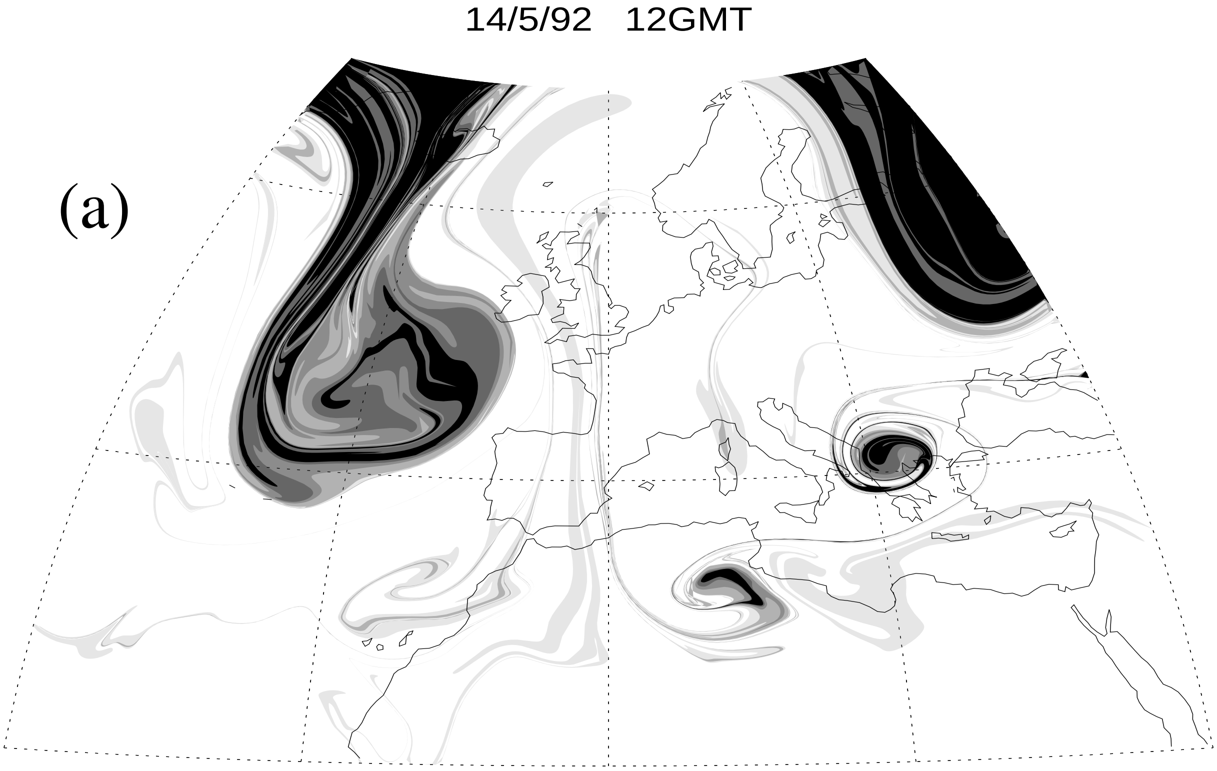
The advantage of satellite data is that the coverage is quasi-uniform over the globe. The disadvantage is that the spatial resolution tends to be rather coarse. At the opposite extreme is data obtained from aircraft or balloons, often during special observational campaigns such as the Antarctic Airborne Ozone Experiment (AAOE), the Arctic Airborne Stratospheric Expedition (AASE), and their successors. This data tends to have very spatial high resolution, but is obtained only in a few locations, leaving a non-trivial problem about how best to interpret it, given that it is a restricted sample of chemical and dynamical fields that vary in three spatial dimensions and in time.

One important problem in numerical weather prediction has been to devise ways of effectively using observational data, obtained on an irregular space and time grid, in a numerical model where the corresponding grid is regular. A number of different techniques have been applied to use observational data to provide initial conditions for such numerical models. One approach has been 'analysis' where the observational data is interpolated in space and time and combined with fields predicted by the model in an integration started at an earlier time. [See e.g. *Daley*, 1991.] This usually has to be followed by another procedure called 'initialisation' before integration of the model can proceed. A different technique is to insert data in a continuous fashion into a model as it is integrated forward in time. This procedure is called assimilation. Development and improvement of assimilation methods is a significant area of current research [see e.g. the literature survey by *Courtier et al.*, (1993)]. Existing methods routinely used for numerical weather prediction models (which normally focus on the troposphere and lower stratosphere) have been successfully applied to models which aim to represent the full troposphere-stratosphere system (*Swinbank and O'Neill* 1994). There is also the prospect that, in future, assimilation techniques may allow much better use of chemical observations (*Fisher and Lary* 1995).

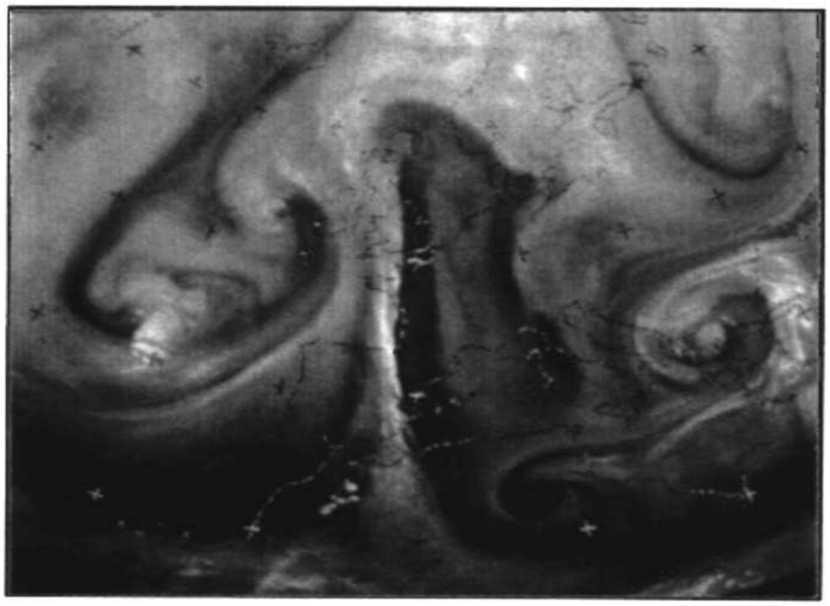
Historically, many observations of the atmosphere or the ocean have provided measured temperature or density, but not velocity. Velocity therefore has to be deduced indirectly. A crude approach is to build up a pressure field via the hydrostatic approximation and then to infer the velocity field from geostrophic balance. (See §2 for precise definitions of these terms.) Both 'analysis' and assimilation provide alternative, more accurate, approaches to this problem. Both give systematic ways of using temperature data (and what ever other data is available, including perhaps some direct velocity observations) together with a dynamical model, to infer velocity fields. Such velocity fields have recently been used with much success to perform various numerical experiments to give greater insight into observed chemical tracers, e.g. associated with one of the airborne expeditions, or with the observations from UARS. (The term 'chemical tracer' is used to mean a chemical species that is conserved following the fluid motion for more that one or two eddy turnaround times, i.e. for more than a few days in the cases of interest here. One might say that, at leading order, the distribution of such a species was determined by transport. Many chemical species in the troposphere and stratosphere have a very short photochemical lifetime and their distributions are determined, at leading order, not by transport, but by local photochemical equilibrium.)

Some aspects of the numerical experiments mentioned above will be discussed in §3, but a first example is given in Figure 1, taken from *Appenzeller et al.* (1996). Figure 1a shows the predicted contours of a tracer on a quasi-horizontal surface, at a level of about 10km above the surface of the Earth. This tracer might represent water vapour and for comparison a direct satellite measurement of water vapour is also shown in Figure 1b. The latter has rather coarse horizontal and, most importantly, vertical resolution and this, plus a number of other factors, makes it difficult to quantify the precise relation between the fields shown in Figures 1a and 1b. Nonetheless, the same gross structures are present in both and it is believed that Figure 1a gives a useful impression of the fields of water vapour and other tracers at scales that are well below the resolution of present day observational techniques.

(a)



(b)



Meteosat water vapour image 14 May 1992 12:00UT

Figure 1: (a) Passive tracer on 320-K isentrope for May 14 1992, at 1200UT, predicted using a high-resolution Lagrangian tracer-advection algorithm, using winds from meteorological data. The tracer contours were initialized five days earlier to coincide with contours of a dynamical tracer, potential vorticity, that was itself calculated from meteorological data. For more details about potential vorticity, see §2.3. (b) Meteosat 5.7 to 7.1 μm water vapour image. The dark regions signify high radiance values and, in effect, low moisture content in the upper troposphere and lower stratosphere. (Both (a) and (b) taken from Appenzeller et al., 1996.)

2 Large-scale atmospheric dynamics

Some readers may wish to skip dynamical details and should proceed directly to §2.5 at this point.

2.1 The equations

The full equations of motion for the atmosphere describe a vast range of different sorts of flow, including sound waves, three-dimensional turbulence, convection, gravity waves, large-scale weather systems in the troposphere and their stratospheric analogues. The emphasis here is on the last two, i.e. on the flow on space scales of a hundred kilometres or more and time scales of a day or more. This sort of flow has a vortex-like character and is sometimes described as 'slow' motion in order to distinguish it from 'fast' sound waves and gravity waves. It is the transport associated with 'slow' motion that largely determines the global scale distributions of quantities such as ozone and water vapour, as well as a whole host of other chemical species that play a leading role in the chemical-climate system.

Understanding of the large-scale 'slow' flow and the transport associated with it is aided by a number of mathematical (and hence conceptual) simplifications. One such simplification, which is particularly useful when considering transport, arises from the fact that the atmosphere is stably stratified. A new vertical coordinate may be chosen that neatly takes account of some of the effects of the stable stratification and is discussed below in §2.2. The stable stratification, plus the fact that the atmosphere is rotating rapidly relative to an inertial frame, also allows the derivation of an approximate set of governing equations that gives a much simplified view of the dynamics.

2.2 Stable stratification and potential temperature

If we are concerned with large-scale motion in the atmosphere, on scales of a few tens of kilometers or more, i.e. safely larger than the scale of clouds, then the atmosphere may be regarded as stably stratified. By this we mean that the density gradient is such that if a fluid parcel is displaced in the vertical, the density of its surroundings and the corresponding pressure forces are such as to return it towards its original position [see e.g. *Holton* (1992), §2.7]. Stable stratification therefore implies inhibition of vertical mixing and vertical transport. [For a recent review see *Fernando*, (1991).]

In an incompressible stratified fluid it is the density that is the appropriate measure of stratification. Fluid parcels conserve their density and if the density decreases with height then the fluid is stably stratified. Often, e.g. in laboratory experiments, density variations are associated with temperature variations and it is temperature increase with height that implies stable stratification. The atmosphere is composed of compressible gas and neither temperature nor density is a relevant measure of stratification. Instead the relevant measure is the density that a fluid parcel would have if it were moved adiabatically (i.e. without irreversible thermodynamic change) to a reference pressure, p_* say. For a perfect gas this measure is simply a function of the entropy, conveniently chosen to be the potential temperature θ, where

$$\theta = T(p/p_*)^{-\kappa}$$

where T is the temperature, p is the pressure and κ is the ratio of specific heat at constant volume to that at constant pressure (equal to 5/7 for a diatomic gas). [The potential temperature is the temperature that a parcel of air would have if it were brought adiabatically, i.e. without change of entropy, to a reference pressure p_*].

If the fluid is stably stratified then the potential temperature must increase monotonically upwards and it may therefore be used to replace geometric height as a vertical coordinate. The corresponding coordinate surfaces are surfaces of constant potential temperature, or equivalently, constant entropy. These surfaces are often referred to as isentropic surfaces and the coordinates as isentropic coordinates. The vertical velocity in such a coordinate system is proportional to the material rate of change of potential temperature and therefore to the rate of change of entropy. Viewing the motion in isentropic coordinates therefore has the advantage of focussing attention on whether or not air parcel motion is adiabatic versus diabatic (i.e. undergoing irreversible thermodynamic changes).

In the atmosphere there are a number of different physical mechanisms by which diabatic changes may occur. The two that are most important from the point of view of this paper, which focusses attention on the upper troposphere and the stratosphere, are (i) radiative transfer and (ii) molecular dissipation occuring in patches of three-dimensional turbulence. In the middle and lower troposphere one would also have to take account of diabatic effects due to moist processes and due to boundary-layer turbulence etc. With regard to (ii) note that there is no strong evidence in the upper troposphere and lower stratosphere for an ubiquitous background field of three-dimensional turbulence, presumably because of the stabilising effect of stratification. However, there is evidence for intermittent patches of turbulence, almost certainly associated with the breakdown of inertio-gravity waves that are excited by flow over topography, frontal systems, convection etc. (e.g. *Dewan* 1981, *Barat* 1982, *Sato and Woodman* 1982 and references therein).

There is a useful division of time scales between an advective time scale along isentropic surfaces, which might be typically three or four days, and the time scale for diabatic effects, which might typically be ten to twenty days. Equivalently one might consider the relevant velocities, which along isentropic surfaces are about 10 $\mathrm{ms}^{-1}$and across isentropic surfaces are less than $10^{-3}\mathrm{ms}^{-1}$, giving a ratio of about 10^{-4}. This should be contrasted with the natural geometric aspect ratio between vertical and horizontal length scales of about 10^{-2} or so. There is a real sense in which diabatic vertical velocities are weak. Thus, viewed in isentropic coordinates, fluid transport can usefully be divided into rapid motion along isentropic surfaces and much slower motion across isentropic surfaces. This leads to considerable conceptual and practical advantages to using isentropic coordinates when considering atmospheric tracer transport. Isentropic coordinates have been exploited formally to good effect in mathematical formulations of the tracer transport problem (see e.g. *Andrews et al.* , 1987, Chapter 9). Here we shall be content to note some of the qualitative insights that arise from their use. See also *Hoskins* (1991) and *Holton et al.* (1995) for further details and discussion. The basic point is that, if we are interested in transport on timescales of a few days, then we may neglect diabatic changes and consider only transport along isentropic surfaces. The full three-dimensional transport problem has therefore been reduced to the two-dimensional problem of transport along isentropic surfaces by the horizontal velocity field. It is this aspect of the transport problem that will be discussed in §3. For reference, Figure 2 shows the configuration of isentropic surfaces, labelled by potential temperature, in the lowest 40 km of the atmosphere. Note that many such surfaces, particularly in lowest 10km, cover quite a broad range of pressures (and of geometric heights).

2.3 Balanced dynamics and the quasi-geostrophic equations

Returning to questions of dynamics, the derivation of a simplified form of the governing equations that applies to the 'slow' motion when the fluid is stably stratified and rapidly rotating is a standard exercise in geophysical fluid dynamics, and covered in textbooks such as those by *Gill* (1982) or *Pedlosky* (1987). The reader who is interested in mathe-

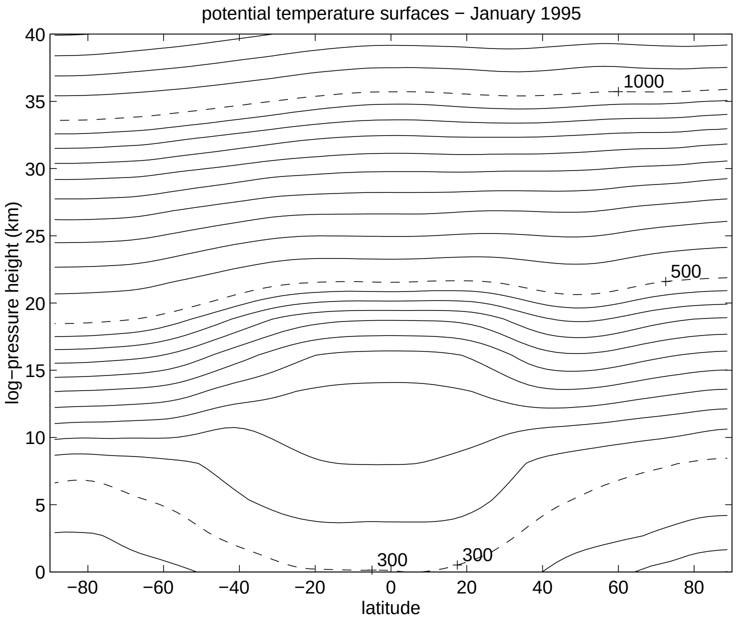


Figure 2: Contours of potential temperature averaged longitudinally and over month of January 1995, as a function of latitude and log-pressure height, calculated using UK Meteorological Office data. The latter is defined to be log (1000mb / pressure) multiplied by 7km and is a reasonable approximation to geometric height. Contours are at intervals of 20K, up to 500K, then 50K above that. The 300K, 500K and 1000K contours are dashed. The contours show the average configuration of potential temperature surfaces, often know as 'isentropic' surfaces.

matical details is recommended to consult such textbooks. Here only a brief motivation is given.

One key to finding a suitable set of equations is to use one of the interesting properties of a stratified fluid, that there is a a quantity called potential vorticity that is materially conserved when the flow is frictionless and adiabatic. The potential vorticity P is generally defined by

$$P = \zeta_a . \nabla\theta / \rho$$

where ζ_a is the absolute vorticity, i.e the vorticity measured with respect to an inertial frame, so that $\zeta_a = \nabla \times \mathbf{u} + 2\mathbf{\Omega}$, where $\mathbf{u}$ is the velocity with respect to the rotating Earth and $\mathbf{\Omega}$ is the rotation rate of the Earth, ρ is the density and θ, as before is the potential temperature. [In fact, θ may be replaced by any quantity χ that (a) is itself materially conserved in adiabatic frictionless motion and (b) such that $\nabla\chi.(\nabla\rho \times \nabla p) = 0$ where p is pressure.]

At first sight the material conservation of potential vorticity may seem no more than a curiosity. It allows the possibility of constructing a materially conserved quantity from dynamical quantities such as velocity and temperature and hence a way of visualising the motion by displaying time sequences of the P field. But, of course, the same could be done using observed fields of any chemical tracer. The special property of potential vorticity is that for the 'slow' motion it satisfies an invertibility principle. That is to say, knowledge of the potential vorticity field, plus certain other quantities, specifically the potential temperature field on boundaries, plus some knowledge of the 'average' stratification, perhaps measured by the total mass distribution between isentropic surfaces, instantaneously determines all other flow quantities such as velocity field, temperature and pressure. The instantaneous relations between potential vorticity and the other flow quantities follow through certain 'balance' conditions, that eliminate the 'fast' gravity-wave and sound-wave motion. This gives a complete description of the 'slow' fluid motion: a single prognostic equation describes the advection of the potential vorticity field by the fluid velocity field (and perhaps the changes in potential vorticity through the effects of friction or diabatic heating) and at each instant the velocity field (and other flow quantities) are determined by the potential vorticity. [For a more detailed account, see e.g. *Hoskins et al.* (1985), *McIntyre* (1992).] Potential vorticity (hereafter PV) is sometimes described as an 'active' tracer (to be contrasted with a 'passive' tracer) since its redistribution by transport affects the structure of the flow.

In general there is no closed-form mathematical representation of the invertibility principle (and hence of a system of equations for the 'slow' motion). However, approximate forms may may be written down in a straighforward mathematical way when at least one of two important dimensionless parameters, the Rossby number (which measures the ratio between a typical rate of change and the rotation rate) or the Froude number (which measures the ratio between typical velocities and the gravity wave speed) are small. One much-used example that is valid at small Rossby number is the quasi-geostrophic system, which describes the time evolution of a flow that is close to geostrophic balance, i.e with the dominant balance in the horizontal momentum equation between the Coriolis force and the pressure gradient. Geostrophic balance therefore gives the necessary instantaneous balance conditions between different flow quantities. Again, see textbooks such as those by Gill and Pedlosky for derivations of the quasi-geostrophic system. Here we simply note the form of the equations and some of the implications. For simplicity we will work in a Cartesian coordinate system where x, y and z are distances in the eastward, northward and vertical directions respectively. For realistic atmospheric applications there are advantages to using pressure p in place of z as a vertical coordinate, but that is not necessary here. The assumption that the flow is close to geostrophic balance means that we can define a leading order 'geostrophic' flow field with horizontal components (u_g, v_g) given by $u_g = -\partial\psi_g/\partial y$ and $v_g = \partial\psi_g/\partial x$, where ψ_g is the quasi-geostrophic streamfunction equal to $\tilde{p}/f_0\rho_0$. Here f_0 is the local value of the Coriolis parameter (twice the local vertical component of the Earth's rotation), and ρ_0 is an average density. The perturbation density $\tilde{\rho}$ may be deduced from the hydrostatic equation as $\tilde{\rho} = -f_0\rho_0 g^{-1}\partial\psi_g/\partial z$, where g is the gravitational acceleration. [The terms 'perturbation pressure' and 'perturbation density' effectively mean the horizontally varying parts of the total pressure and density fields respectively.] There is a single predictive (or ' prognostic') equation for the flow in the interior (away from horizontal boundaries) of the form

$$\frac{D_g q_g}{Dt} = 0 \tag{1}$$

where D_g/Dt denotes the time derivative following the 'geostrophic' flow and q_g is the quasi-geostrophic potential vorticity (hereafter QGPV) defined in terms of ψ_g by

$$q_g = \frac{\partial^2\psi_g}{\partial x^2} + \frac{\partial^2\psi_g}{\partial y^2} + \frac{\partial}{\partial z}\left\{\frac{f_0^2}{N(z)^2}\frac{\partial\psi_g}{\partial z}\right\} + \beta y. \tag{2}$$

$N(z)$ is a ' background' buoyancy frequency associated with the stable stratification, and β is the latitudinal gradient of the Coriolis parameter. The βy term an approximation to the latitudinal variation of the Coriolis parameter.

Note that here we have the structure identified earlier, an equation (1) which represents the material conservation of PV (or an approximation to it) and an invertibility principle (2) which allows calculation of all other flow quantities, given q_g. Recall that knowledge of ψ_g determines the leading-order velocity field and also the density field.

The calculation of ψ_g from the QGPV field q_g requires the solution of an elliptic equation, so that there is a global dependence of other flow quantities on q_g and in addition there is a requirement for boundary conditions on ψ_g. The relevant boundary condition at 'side' boundaries is that of no normal flow. At 'top' and 'bottom boundaries the natural condition is on the vertical velocity, but the leading order flow is horizontal. In fact the leading-order terms in the density equation may be used to deduce the leading-order vertical velocity, w and in terms of the quasi-geostrophic streamfunction it follows that

$$w = \frac{g}{N^2\rho_0}\frac{D_g\tilde{\rho}}{Dt} - \frac{f_0}{N^2}\frac{D_g}{Dt}\frac{\partial\psi}{\partial z} \tag{3}$$

Note that if w is known on the upper and lower boundaries, then (3) represents another prognostic equation, for $\partial\psi/\partial z$.

The form of the quasi-geostrophic equations has many similarities with the more widely known equations for two-dimensional vortex dynamics (hereafter 2DVD), which take the form

$$\frac{Dq}{Dt} = 0, \tag{4}$$

where the velocity $\mathbf{u} = (u, v)$, may be defined in terms of a streamfunction ψ, with $u = -\partial\psi/\partial y$ and $v = \partial\psi/\partial x$. The relation between the vorticity q and the streamfunction ψ is that

$$q = \frac{\partial^2\psi}{\partial x^2} + \frac{\partial^2\psi}{\partial y^2}. \tag{5}$$

Given suitable boundary conditions on ψ, (5) may be regarded as an invertibility principle for the two-dimensional flow in the same way that (2) is an invertibility principle for the quasi-geostrophic flow.

2.4 Quasi-geostrophic motion

What is the nature of quasi-geostrophic motion and, hence, what its stirring and mixing properties? The type of dynamics described by (4) and (5) is familiar from many studies of 2DVD and two-dimensional turbulence. For example, in a flow where vorticity is concentrated into localised coherent vortices (point vortex dynamics is an extreme idealisation of such a flow, but flows with this character are more generally observed in e.g. later stages of 2-D turbulent 'spin-down' experiments), each vortex moves with a velocity that is determined by the strengths and relative positions of the other vortices. This is an indication of the non-local dependence of ψ on q implied by (5). Far vortices are less important than near vortices in this respect; the velocity field associated with a given vortex decays inversely with distance from that vortex.

Given the close similarity between the quasi-geostrophic equations (1) and (2) and those for 2DVD (4) and (5), one might expect quasi-geostrophic flows to evolve in a similar manner. A coherent vortex flow analogous to that described above would in the QG system again have each vortex moving horizontally under the influence of the others, but the vortices might be on different levels and the decay of the velocity field due to a particular vortex would be as the inverse square of the distance.

The similarity between the two systems is one of the reasons why 2DVD and two-dimensional turbulence have received a great deal of attention in the literature, even though two-dimensionality is almost impossible to achieve in a homogeneous fluid, except under conditions where there is a very strong geometric constraint, e.g. in soap films (*Couder and Basdevant*, 1986). There have now been many analytical and numerical studies of quasi-geostrophic flows, e.g. simple point vortex configurations (*Walsh and*

Pratt, 1995), coherent vortex evolution including effects of background shear and strain and interactions between vortices (*Meacham et al.* 1995, *Viera* 1995, *Dritschel and de la Torre*, 1996), and in turbulent spin-down experiments (*McWilliams* 1989, *McWilliams et al.* 1994, *Dritschel and Ambaum* 1997).

It seems clear that almost all phenomena encountered in 2DVD have analogues in the QG system. Nonetheless there are differences between the two systems and these may be particularly important when one is considering specific applications to the atmosphere. The extra space dimension in the QG system allows some new phenomena. For example, interactions between coherent vortices at different levels but similar horizontal positions are possible in the QG system but have no analogue in 2DVD.

One important question that is of current research interest concerns spatial isotropy. From (2) one can infer a useful rough guide to length scales for quasigeostrophic flows, that the ratio of horizontal to vertical scales is about N/f_0, often referred to as Prandtl's ratio. However that does not mean that there is necessarily exact three-dimensional isotropy if the z coordinate is suitably scaled. For one thing, it is only the horizontal components of the gradient of the streamfunction that determine the velocity field. The recent simulations of *McWilliams et al.* (1994) suggest that the manifestation of breakdown of isotropy in a spin-down experiment may be the formation of tall vortex structures. However, detailed understanding of the mechanisms that lead to the formation of these structures is still lacking. (And rather contradictory results have been obtained by *Dritschel and Ambaum* (1997)).

The quasi-geostrophic system is no longer regarded as suitable for detailed numerical calculation of atmospheric and oceanic flows, e.g. in numerical weather prediction, even for flows that are dominated by 'slow' motion. However, it remains an extremely useful conceptual model on which to base understanding of 'slow' atmospheric and oceanic flows and with which to interpret observed flows and realistic numerical simulations, in particular because it includes an explicit and easily understood invertibility principle.

In what follows we shall concentrate on transport, stirring and mixing of passive tracers. But it should be plausible from what has been said already that considering the transport of PV itself can give important insights into the dynamics. See e.g. *Hoskins* (1991), *McIntyre* (1992), *Rhines* (1986), the latter focussing on the oceanic case.

2.5 Implications for the transport and mixing problem

The close relation between stratified, rotating flows and 2DVD has important implications for how stirring and mixing occurs. One might say that we have two extreme paradigms for stirring and mixing. One is chaotic advection, where flow that varies smoothly in space and time, and in particular has a well-defined length scale, leads to small-scale structure in advected tracer fields. Since the advection is dominated by the large scales, the eddy turnover time remains the same at all scales. The other extreme is that of three-dimensional turbulence where eddy turnover time decreases as scale decreases. It follows that, at least above the Kolmogorov scale where viscous dissipation becomes important, the eddies of a given length scale make a substantial contribution to stirring and mixing at that scale.

2DVD, and, by implication, rotating stratified flow in the atmosphere and in the ocean appear to be closer to the first paradigm, where large-scale advection dominates. Exactly why this is the case is a subtle issue. For example, as noted by *Pierrehumbert et al.* (1995), spectral arguments for quasigeostrophic dynamics and 2DVD suggest that all scales of the flow contribute equally to local strain rates. *Pierrehumbert et al.* (1995) gain some insight into why this is not the case in practice by constructing analogues to 2DVD systems in which there is a different relation between the materially conserved quantity and the streamfunction. One interesting example that can also be motivated by certain atmospheric flows (e.g. *Juckes* 1994, *Held et al.* 1996), is where the materially conserved quantity corresponds to a first spatial derivative, rather than a second spatial derivative of the streamfunction. It turns out that this is relevant to quasi-geostrophic flows in which there is constant interior QGPV, but where there are lower (or upper) boundary density gradients and it is the boundary density anomalies that control the flow, through (3) with $w = 0$. In this case when a lower boundary density anomaly reduces in scale the induced vorticity increases. Such flows are characterised by the formation of filamentary structures in the boundary temparature field that frequently roll up into small vortices. Indeed it is useful to constrast the canonical example of the roll-up of a shear layer in the two cases. In conventional 2DVD the shear layer (equivalent to a filament of anomalous vorticity) is unstable and rolls up into vortices that, in the absence of dissipation, are connected by thin filaments of anomalous vorticity. These filaments do not themselves roll up into vortices. The reason appears to be that the filaments are being stretched out in the strain field due to the vortices, which has the effect of inhibiting vortex roll-up

(*Dritschel et al.* 1991). In surface temperature dynamics, on the other hand, the filaments connecting the primary vortices themselves roll up into vortices. The reason is that as the surface temperature filaments are stretched out the vorticity that they induce increases and this means that the strain field due to the primary vortices is ineffective in inhibiting the roll-up of such filaments. Nice examples contrasting the two cases are shown by *Held et al.* (1996).

It therefore appears that the phenomenon of stabilization of vortex filaments by the strain field due to the large-scale flow means that in practice small-scale vorticity features are usually, although not always, passive and it is large scales that dominate the velocity field. The chaotic-advection paradigm for transport, stirring and mixing is therefore potentially relevant to 2DVD and to quasigeostrophic dynamics.

2.6 Rossby waves and instabilities

The close analogy between 2DVD and quasigeostrophic flows implies the possibility that some atmospheric and oceanic flows will have the character of interactions between small numbers of coherent vortices. In such cases the sort of chaotic advection studies pioneered by *Aref* (1984) might be relevant. However, more often atmospheric flows are dominated by strong background shear flow in the longitudinal direction. Disturbances of such flow away from longitudinal symmetry, either by waves due to external forcing or by the spontaneous growth of disturbances through hydrodynamic instability, are crucical for effective transport, stirring and mixing.

The relevant wave propagation mechanism for these flows is associated with gradients of vorticity or potential vorticity. When fluid parcels are moved in a direction perpendicular to such gradients, the vorticity field is changed and a pattern of circulations is set up that tends to restore the fluid parcels to their original position. This mechanism is often referred to as the 'Rossby restoration mechanism' and gives rise to the propagation of 'Rossby waves'. The mechanism is analogous to, but different in important details from, that which operates in a stably stratified fluid and gives rises to the propagation of internal gravity waves. Figure 3 shows in a schematic way how the mechanism works in a simple case. Rossby waves can propagate in the horizontal and in the vertical and therefore provide an important mechanism for communicating the effects of forcing to parts of the flow that are far from the forcing location. For example, it has been suggested that the effects of tropical heating are communicated to midlatitudes through Rossby wave propagation and thereby affect the month-to-month variability of mid-latitude weather systems. Similarly, Rossby waves that are generated in the troposphere, e.g. by geographical variations in topography and heating, propagate up into the stratosphere, where they have important implications for transport, stirring and mixing.

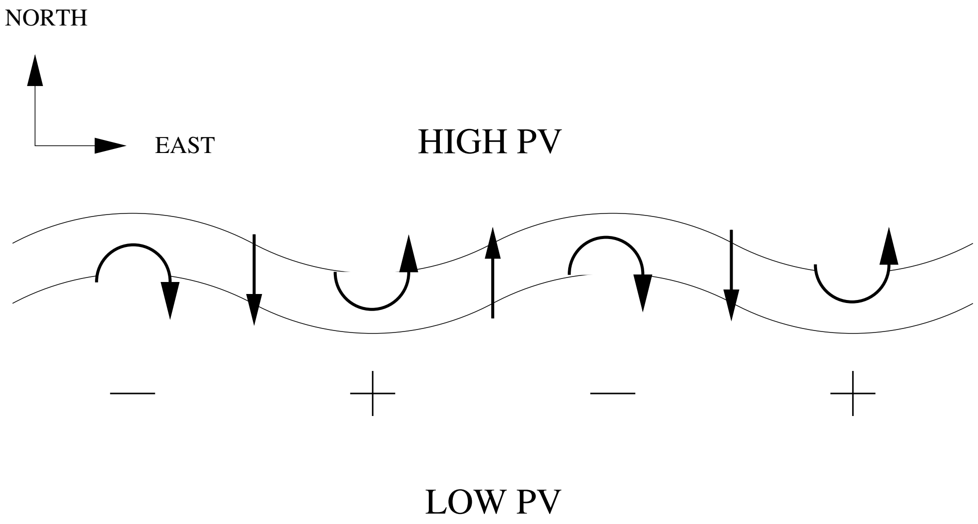


Figure 3: The propagation mechanism for Rossby waves: In the background flow there is a north-south gradient of potential vorticity, so that potential vorticity contours generally run east-west. Here a wave pattern of north-south displacements has been applied so that the potential vorticity contours, shown as wavy lines, are disturbed from their background configuration. This implies a pattern of relative vorticity anomalies of alternating positive and negative sign (shown by the +s and −s). This relative vorticity field has a corresponding velocity field shown by the bold arrows. Note that the positive northward velocities are displaced one quarter of a wavelength to the west of the positive northward displacements (and correspondingly for negative velocities and negative displacements). This implies that the pattern of displacements will propagate to the west, as a Rossby wave.

The alternative idea, that flow asymmetries arise through instability, is most relevant in the troposphere. The longitudinally symmetric flow that might be expected to arise through the action of differential heating may be argued to be unstable, through a mechanism known as baroclinic instability, so that what is achieved in practice is a flow with strong longitudinal asymmetries that can act to transport tracers in the latitudinal direction. In the stratosphere baroclinic instability is much less effective and the main mechanism causing flow asymmetry is the upward propagation of large-scale Rossby waves mentioned earlier. Both baroclinic instability, and another relevant hydrodynamic instability, barotropic instability, which is more closely analogous to the shear instability familiar from 2D vortex dynamics, may be understood in terms of Rossby wave dynamics. Both instabilities involve the disturbance of the PV field by the flow and the resulting change in the flow field due to the changes in PV. Again *James* (1994) and *Andrews et al.* (1985) provide useful background material on Rossby wave dynamics in the atmosphere and *James* (1994) discusses tropospheric baroclinic instability More details of the physical mechanisms for Rossby waves and for barotropic and baroclinic instabilities are provided by *Hoskins et al.* (1985).

Of course, longitudinal asymmetry of the flow is not enough, by itself, to ensure effective transport. The latitudinal particle excursions associated with an asymmetric flow may be entirely reversible and have no permanent effect on tracer fields. There has been considerable emphasis on this fact in the literature on stratospheric transport, partly for the historical reasons that the simplest mathematical models often assumed that disturbances to the flow were steady linear waves and that attention was often focussed on deriving parametrizations of transport for use in two-dimensional (height-latitude) models. When disturbances have non-trivial time dependence they are usually effective in transporting tracers and we shall ignore this issue from now on. A careful discussion is given in *Andrews et al.* (1987, Chapter 9).

3 Quasi-two-dimensional transport

3.1 Introduction

Restricting interest to advection along isentropic surfaces means that the transport problem is two-dimensional. This is a considerable simplification and, of course, allows comparison with studies of chaotic advection based on two-dimensional velocity fields. In this section we review the structure of two-dimensional isentropic transport in the stratosphere that has been revealed over the decade or so by new tracer observations. We also review some of the techniques using wind fields constructed from observations that are now used to gain further insight into transport. We then consider how this picture of transport is consistent with that predicted by chaotic advection studies and how ideas from chaotic advection can be used to give quantitative insight into observed flows.

3.2 Transport by breaking Rossby waves – the surf zone

One approach to recognising the spatial structure of transport, stirring and mixing along isentropic surfaces is to consider the distribution of PV, which, as noted earlier, is itself a tracer. PV fields calculated for large-scale atmospheric flows typically show two very different sorts of behaviour. The difference is particularly clear in the winter stratosphere, where the flow is dominated by a strong westerly (eastward) vortex around the high-latitude 'polar night'. This vortex is disturbed by Rossby waves that are generated in the troposphere and propagate up into the stratosphere.

In one part of the flow, generally at higher latitudes and corresponding to the main vortex, the PV contours are displaced from latitude circles and are distorted, but remain quasi-circular. This behaviour is that which is implicit in linear wave theory and can be regarded as a manifestation of the upward Rossby-wave propagation. At lower latitudes, on the other hand, PV contours are stretched out by the flow and strongly deformed away from a quasi-circular shape. This behaviour is outside the scope of linear wave theory and it has been argued by *McIntyre and Palmer* (1983, 1984) that it is a manifestation of the breaking of Rossby waves. Indeed McIntyre and Palmer named the region outside the main vortex – where PV contours are strongly stretched and deformed – the 'stratospheric surf zone', in order to emphasise the analogy with surface gravity waves breaking on an ocean beach. It is clear that there will be effective stirring and mixing of chemical tracers, as well as PV, in the surf zone.

The flow studied by McIntyre and Palmer was in the middle stratosphere, at around 30km. But we now realise that that the stirring properties of the flow are highly inhomogeneous in latitude at almost all levels in the stratosphere, with the location of the regions of strongest and weakest stirring varying substantially with height.

The distinctive surf-zone, main vortex structure is illustrated in Figure 4, which shows particles that are advected by the flow in a high-resolution, single-layer, numerical simulation of the type pioneered by *Juckes and McIntyre* (1987), [McIntyre and Pinhey, personal communication; see also *McIntyre and Pinhey* (1998)]. In such simulations the disturbances are forced in-situ by a large-scale vorticity forcing, but the effect is rather similar to the arrival in a given layer between two isentropic surfaces, say in the middle stratosphere, of Rossby waves that have propagated up from below. High-resolution three-dimensional simulations (e.g. *Mahlman and Umschied* 1987, *Haynes* 1990) have confirmed this similarity.

The particles shown in Figure 4 are shaded to mark their initial latitude, with darker shades corresponding to higher latitudes. The flow is on the surface of a sphere and is viewed from above the North pole. The initial flow takes the form of a strong anti-clockwise vortex at high latitudes, with weaker clockwise flow at low latitudes. This longitudinally symmetric flow is subsequently disturbed by a large-scale forcing, centred at midlatitudes. The particles are stirred up by the flow, but the pictures clearly show the strong inhomogeneity of the stirring. The largest stirring region, the 'surf zone', is

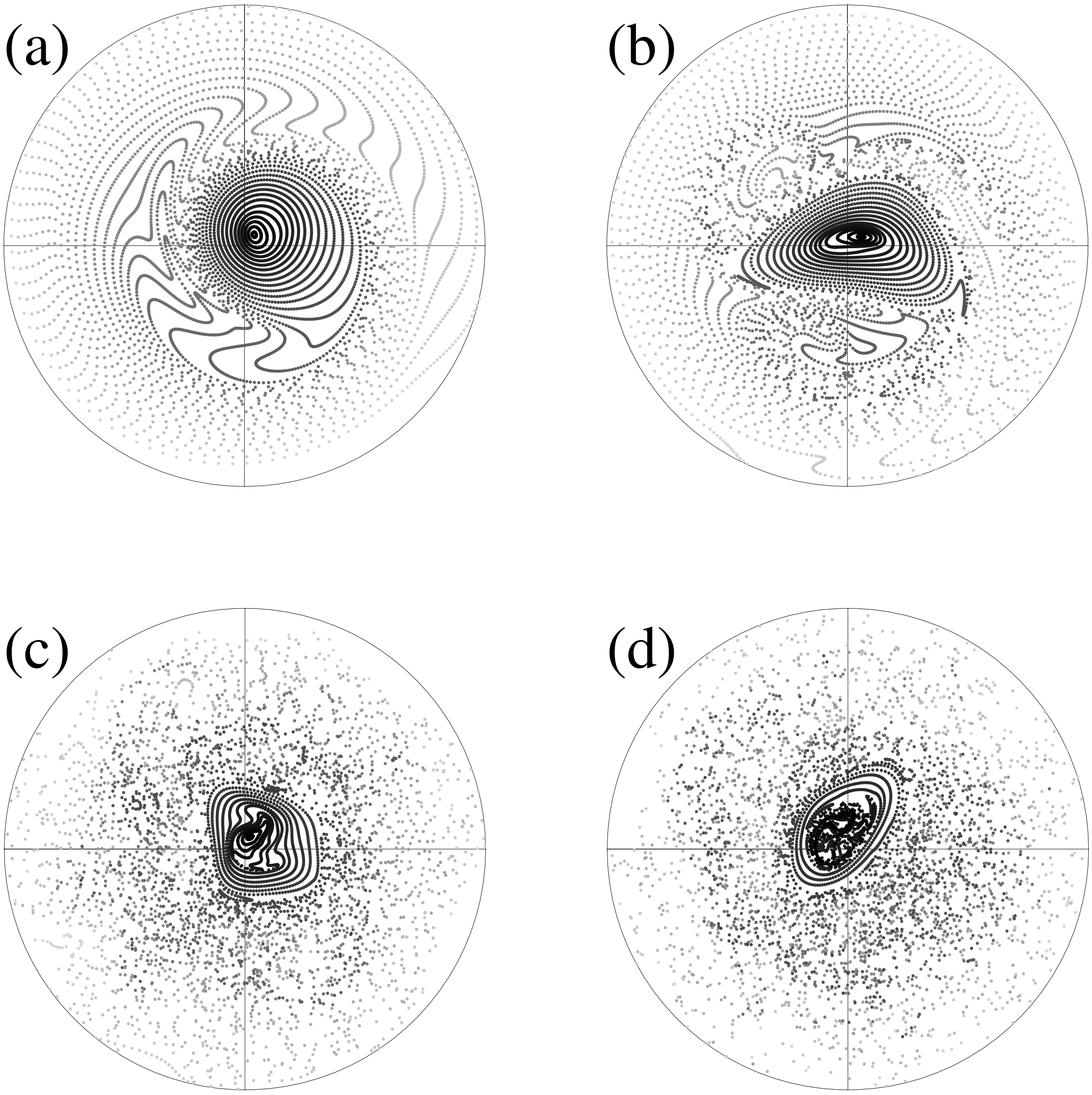


Figure 4: Tracer particles advected in a dynamically consistent numerical simulation of single-layer flow on a sphere. The flow is viewed from above the North pole. The particles are shaded according to their initial latitude, with darker shading corresponding to higher latitude. The flow consists of a strong cyclonic (anti-clockwise) vortex at high latitudes, disturbed by a large-scale forcing centred at mid latitudes. The amplitude of the forcing is smoothly increased from zero at the beginning of the simulation and is smoothly decreased and then increased again, with a period of 20 days, as the simulation progresses. Particles are shown at (a) 10 days, (b) 20 days, (c) 40 days, and (d) 80 days. It may be seen that there is a systematic adjustment of the particles to a statistically quasi-steady state where they are apparently well-mixed outside the vortex and inside the vortex, but there is no exchange across the vortex boundary.

at mid and low latitudes. The high-latitude vortex persists as a coherent material entity. There is evidence of stirring within the vortex, but there appears to be a strong barrier to transport associated with the vortex edge. In this particular simulation the barrier is perfect and no particles leak from the interior of the vortex to the surf zone or vice versa.

3.3 Tracer edges

We have seen how in the flow depicted in Figure 4 the strong stirring is localised to the surf zone. Simple consideration of the effect of localised stirring on a tracer field with an initially smooth distribution shows that, across the edges of the stirring region, a sharp jump in the value of the tracer will tend to form. Figure 5 gives a schematic illustration of how this occurs. If the pattern of stirring in the real atmosphere is similarly localised, then we might expect to see evidence for sharp jumps in the values of tracers, both chemical tracers and PV at the vortex edge, as the poleward boundary of the stirring region.

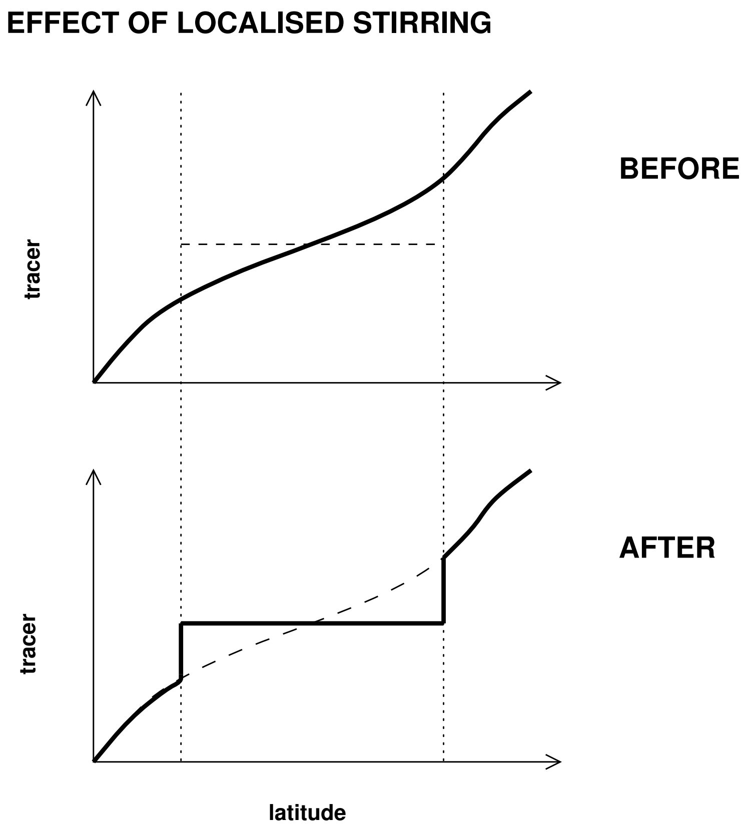


Figure 5: Effect of localized stirring on a tracer distribution. The variation with latitude of initial tracer distribution is shown by the solid curve. The tracer is assumed subsequently to be stirred (and perfectly mixed) over a limited range of latitudes, bounded by the finely dashed lines. The effect of the localised stirring and the resultant mixing is not only to make the tracer constant in this region, but also to produce sharp tracer gradients at the edges of the region.

Figure 6 [which is a reconstructed version of Plate 5 in Tuck (1989)] shows tracer data from the AAOE (Antarctic Airborne Ozone Expedition), measured on the ER-2 aircraft. The resolution of the data is limited by the response time of the various instruments and the aircraft speed, but is certainly better than 10km or so. The tracers shown are N_2O , which generally decreases poleward in the stratosphere, NO_y , which generally increases poleward, and ozone, which also increases poleward, at least in early and midwinter and in midlatitudes. (By NO_y we mean a group of reactive nitrogen compounds. The total of the members of this group is relatively long-lived in the lower stratosphere, but the individual members are not, since there is rapid photochemical conversion from one species to another within the group.)

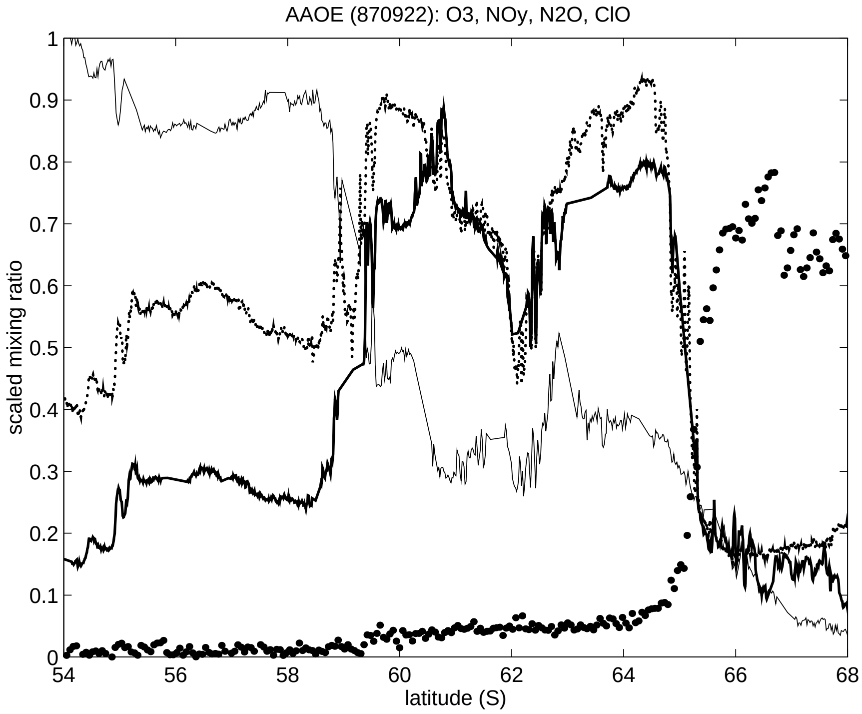


Figure 6: [After Tuck (1989), Plate 5]. Variation of mixing ratios of ozone (O_3) (dotted curve), nitrous oxide (N_2O) (fine solid curve), 'odd nitrogen' (NO_y) (heavy solid curve) and chlorine monoxide (ClO) (heavy dots), measured along flight track of ER-2 aircraft on 22 September 1987 during AAOE. The mixing ratios have had a constant value subtracted and then multiplied by a suitable factor to allow all to be shown together. What is plotted is, for O_3, 0.0005 times (mixing ratio (ppbv) - 300), for N_2O, 0.008 times (mixing ratio (ppbv) - 100) for NO_y, 0.1 times (mixing ratio (ppbv) - 2), and for ClO, mixing ratio (ppbv). See text for further details and explanation.

There is a clear jump in the value of these tracers at about 59 deg S. Unfortunately it is not possible to measure PV directly, so there are no corresponding measurements to verify that there is a jump in PV colocated with that in the chemical tracers. It is possible to construct PV fields from large-scale meteorological analyses, but such field will have very coarse resolution compared to the aircraft measurements. However, simple mathematical models of a dynamical structure incorporating a PV edge suggest that there will be a jump in the wind shear across the PV edge. The aircraft measurements indeed show that there the shear changes sign from north of 58 deg S to south of 60 deg S. This is at least an indication that there is a PV edge at this location.

There are a number of other features seen in Figure 6 that deserve explanation. Firstly there is another striking jump in the values of some tracers, particularly NO_y and O_3 , at around 65 deg S. Note that here there is apparently no jump in N_2O , nor is there any striking change in the wind shear. The accepted explanation for this feature is that it marks the edge of the chemically perturbed region, defined not by any change in transport regime, but by whether air parcels have encountered polar stratospheric clouds. The latter form when temperatures are sufficiently low and there is no reason why the boundary of this region should coincide with the PV edge. The polar stratospheric clouds allow chemical reactions that decrease NO_y and increase ClO , but do not affect N_2O . The ClO can, under appropriate conditions, catalyse the destruction of ozone. There is another interesting feature at around 62 deg S. This is interpreted by *Tuck* (1989) as being associated with a filament of low ozone air originally from inside the chemically perturbed region, that is partially mixed with its surroundings so that, in particular, the high NO_y introduced by mixing has destroyed all the ClO . This is a reminder that in the stratosphere (and elsewhere in the atmosphere) stirring and mixing may have important dynamical consequences.

The schematic picture of the effect of localised stirring shown in Figure 5 suggests that, if the strong stirring associated with Rossby wave breaking is confined to midlatitudes, there should be a sharp edge in tracers not only on the poleward side, but also on the equatorward side. There is indeed evidence for strong latitudinal tracer gradients between the tropics and subtropics in the stratosphere from aerosol observations e.g. *Trepte and Hitchman* (1992) and from observations of chemical tracers e.g. *Murphy et al.* (1993). Figure 7 has been constructed following Figure 5 in the latter paper and shows a composite of lower stratospheric aircraft measurements of NO_y and ozone (O_3) . Both NO_y and O_3 may be regarded as conserved tracers in the lower stratosphere. The fact that the aircraft measurements are from a number of different heights, and that the concentrations of both NO_y and O_3 have strong vertical gradients, means that latitudinal contrasts are difficult to distinguish when the concentrations are plotted individually. However, the ratio of the two concentrations (which may itself be regarded as the concentration of a conserved tracer) has a much weaker vertical gradient and when, as in Figure 7, it is plotted against latitude it shows clearly that there are strong latitudinal gradients centred at about 18 deg S and 15 deg N.

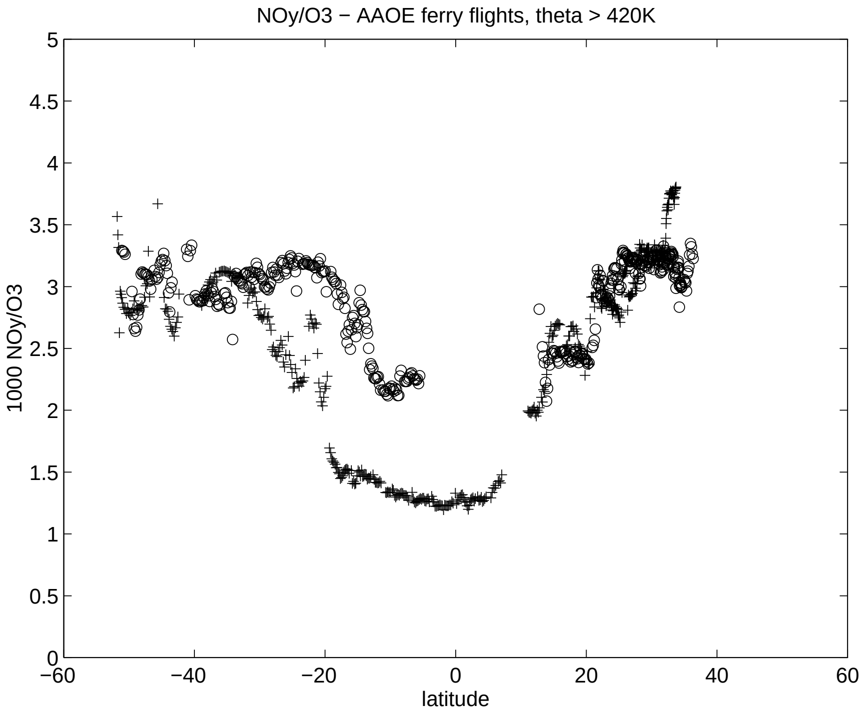


Figure 7: [After *Murphy et al. 1993*, Figure 5.] Ratio of mixing ratios of NO_y and O_3 versus latitude for AAOE ferry flights from NASA Ames to Punta Arenas. Points marked $\circ$ are for the southward flights (12, 14, 15 August 1987) and points marked + for the northward flights (29 September, 1 and 3 October 1987). Only values corresponding to potential temperature greater than 420K have been plotted.

3.4 Use of observed velocity fields for transport studies

As noted earlier, one of the important developments that has complemented the improved observations of chemical tracers has been the construction of velocity datasets for the stratosphere, using assimilation techniques. These velocity datasets have been exploited in a number of different techniques for tracer studies.

One technique is simply to use the velocity fields to follow trajectories of air parcels. Trajectory studies have a long history in dynamical meteorology but the improved velocity datasets together with increases in computing power have allowed a very large number of trajectories to be followed simultaneously. The so-called 'domain filling trajectory' approach has been used by *Fisher et al.* (1993) and *Sutton* (1995) to follow the three-dimensional trajectories of air parcels on seasonal time scales, but has also been used by *Bowman* (1993a) to follow particle trajectories restricted to isentropic surfaces, to gain insight into transport on shorter timescales.

A second technique is to use the velocity fields in a tracer model, in which the tracer equations are integrated using finite-difference, finite-element or spectral methods. Most such studies to date have restricted the problem to two dimensions by considering the evolution of the tracer on an isentropic surface. For example, *Chen et al.* (1994) have used this approach to study transport into and out of the tropics on different isentropic surfaces.

The third technique in common use is to use the velocity fields to follow the evolution of a material contour. The contour is defined by a finite set of particles, but the algorithm used allows redistribution of the particles around the contour, or the introduction of new particles on the contour, as the integration proceeds, so as best to represent the contour. Once again the problem is usually restricted to two dimensions by using winds on a particular isentropic surface. This approach is generally referred to as 'contour advection' or in its more sophisticated versions 'contour advection with surgery' (hereafter CAS) (*Norton* 1994, *Waugh and Plumb* 1994). The 'surgery' refers to the fact that as the contour becomes more and more contorted, as it inevitably does in a flow that is stirring effectively, it may be necessary to remove parts of the contour, e.g. associated with thin filaments, in order to keep the number of particles needed to define the contour to within the limits necessary for practical computation. Algorithms for effective representation of contours and effective application of surgery were first developed in the context of solving the two-dimensional vorticity equation for flows in which the vorticity field is piecewise constant (and therefore defined entirely by the contours dividing vorticity of different values) (see e.g. *Dritschel* 1989).

All of these techniques have limitations. They are usually applied to an initial-value problem, and the picture of the tracer field obtained for a particular date and time will depend both on the initial condition and the time that has elapsed since the initial condition was applied. For example, CAS calculations are typically initialised by placing contours to coincide with contours of a tracer for which large-scale observations are available. This is usually PV, calculated using wind and temperature from meteorological datasets, but chemical tracers such as ozone, for which there are satellite data, have also been used (A. Mariotti, personal communication 1997). The large-scale observations typically have useful resolution only at scales of a few hundred kilometers or more. Integrating forward in time from the initial condition, the contour is deformed by the flow and typically develops filamentary structure at increasingly small scales. At the end of the integration, typically ten to twenty days, the resulting contour is taken, in some sense, to give a fine-resolution view of a tracer field for which only coarse-resolution data was available initially. The precise relation between the predicted contour and contours of a real tracer is a subtle one. Any small-scale filaments that are represented are only those that have been produced from large-scale tracer features over the integration period. Small-scale filaments that persisted for the period of the integration would not be captured (assuming that they were below the useful resolution of the observations used for the initialisation). Small errors in the observed velocity fields will lead to corresponding errors in the positions of filaments that will often be as large, or larger than, the width of the filaments themselves. It is therefore clear that any simple quantitative measure of the correlation between CAS predictions and high-resolution tracer measurements is unlikely to show strong agreement.

Nonetheless CAS, and the other techniques mentioned above, have significantly enhanced useful interpretation of tracer observations. For example, CAS has been used to follow the evolution of filaments of vortex air that have peeled off into the surf zone and has shown convincing qualitative agreement with filamentary structures seen in aircraft data (see, for example, Figures 1 and 2 of Waugh et al. (1994). Such studies have been important in supporting the idea that the structures seen in the aircraft data can be explained as due to stirring by the large-scale flow on isentropic surfaces. It seems that the success of CAS here is due to the fact that the strongest contrasts in tracer are associated with filaments of vortex air that are drawn away into the surf zone by relatively infrequent erosion events. These are exactly the sorts of structures, arising over a period of a few days from the contrast in tracer across the vortex edge (that is, of course, captured in the large-scale observations), that CAS is best able to capture. It is also the case that techiques such as CAS have given a qualitative sense of the tracer structures that would be observed if the high resolution aircraft measurements gave global coverage, or if satellite measurements were at much higher spatial resolutions. But once these qualitative insights have been gained, one might legitimately look for more quantitative insights from such techniques.

A more quantitative application of CAS has been to quantify transport, e.g. across the boundary of the vortex. *Waugh et al.* (1994) have performed calculations in which a contour is initialised to coincide with a PV contour in the neighbourhood of the vortex edge. The subsequent advection leads to the formation of filaments on this contour, which are stretched out into the surf zone. The area associated with such filaments is interpreted as a measure of the transport out of the vortex into the surf zone and is quantified by applying the surgery algorithm with a rather coarse cut-off scale. This leads to the removal of filaments and a reduction in the area enclosed by the contour. A similar approach has been used by *Waugh* (1996) to quantify transport across the subtropical barrier.

3.5 Relevance of chaotic advection

It is perhaps appropriate at this stage to note the predictions of theories of chaotic advection and their possible relevance to atmospheric flows. The first requirement for the usefulness of the chaotic advection paradigm is that small-scale structure arises in tracer fields through the action of velocity fields that have simple large-scale spatial structure. It has already been argued from basic dynamical considerations in §2 that this is the case in the atmosphere, at least on large enough scales. There is also useful evidence from CAS calcuations that, at least in the stratosphere, it is indeed the large-scale part of the velocity field that dominates. For example, *Waugh and Plumb* (1994) compare results of contour advection calculations with velocity fields at different spatial resolutions and show that there is rather good agreement between results with velocity fields that are resolved at say, 3 deg latitude $\times$ 3.6 deg longitude and 10 deg latitude $\times$ 12 deg longitude.

Much of the work on chaotic advection has concerned two-dimensional, non-divergent flows, i.e. Hamiltonian dynamical systems (*Aref* 1984), and this may be argued to be relevant to the real atmosphere on the following basis. The potential relevance of two-dimensional flows has already been justified by arguing that restriction to isentropic surfaces is relevant, perhaps for twenty days or so in the lower stratosphere, less in the middle and upper stratosphere. Whilst the horizontal flow on isentropic surfaces is not exactly non-divergent, it is non-divergent to some reasonable approximation. (For example, for flows in which the Rossby number Ro is small, the divergence is $O(\mathrm{Ro}\ )$ smaller than a naive estimate of velocity gradients.) There have, of course, been a great many studies of chaotic behaviour in Hamiltonian dynamical systems, and much of the structure found, chaotic regions, in which there is mixing, bounded by regions of invariant tori, i.e. surfaces made up of trajectories, which act as transport barriers, is generic. Two studies that are particularly motivated by atmospheric and oceanic flows are those of *Pierrehumbert* (1991b) and *del Castillo-Negrete and Morrison* (1993). Given the structure of the wintertime stratospheric flow, as revealed by numerical simulation and observations, it is not surprising that it has been suggested that the surf-zone should be regarded as a chaotic mixing region, with the vortex edge as a set of invariant tori (e.g. *Pierrehumbert* 1991a, *Pierce and Fairlie* 1993, *Pierrehumbert and Yang* 1993).

Much of the study of two-dimensional Hamiltonian systems has been based on time-periodic flows, since this allows, through the use of Poincaré sections, application of the theory of area-preserving maps. As noted by Wiggins (1988), flows that are not time-periodic are ubiquitous in practice and one of the continuing challenges in this subject is to develop methods for analysing transport in non-periodic flows (such as those in the atmosphere). One possible approach to quantifying atmospheric transport, that does not depend on time-peridodicity is to use finite-time Liapunov exponents as a diagnostic of stretching (*Pierrehumbert*, 1991a). The finite-time Liapunov exponent $\lambda(t)$ is defined for a fluid particle by assigning an infinitesimal fluid line element $\delta\mathbf{x}$ to that particle and following the stretching of that line element. Then

$$\lambda(t) = \frac{1}{t}\log\{\frac{|\delta\mathbf{x}(t)|}{|\delta\mathbf{x}(0)|}\}. \tag{6}$$

Note that $\lambda(t)$ is not independent of the initial orientation of the line element, but in a stretching flow might be expected to become independent of the initial orientation after a few stretching time scales. For an ensemble of fluid particles in a mixing region of a simple chaotic advection flow, the distribution of the $\lambda(t)$ will, with increasing time, narrow about the value of the Liapunov exponent (the latter defined in the usual way for chaotic dynamical systems). However, at any finite time there will be a spread of values of the $\lambda(t)$.

Pierrehumbert's idea is that transport barriers (or tori) will be manifested as regions where the $\lambda(t)$ are anomalously low. In *Pierrehumbert* (1991a) examples are shown where low values of the $\lambda(t)$ for a simple chaotic map reveal the presence of invariant tori embedded within a mixing region. The advantage over Poincaré sections is that only a relatively short time window is needed to identify such features (and, in particular, there is scope for application to non-periodic flows). Diagnostics based on finite-time Liapunov exponents have been applied to the atmosphere using wind fields from large-scale observations (*Pierrehumbert* 1991a) and also from a general circulation model (*Pierrehumbert and Yang* 1993) to advect particles and stretch line elements.

Another diagnostic, which is closely related to finite-time Liapunov exponents, calculates the change in length of a finite material contour rather than the changes in lengths of an ensemble of material line elements. The evolution of the contour length may be followed simply by tracking large numbers of particles or using the CAS technique. The stretching of the contour is clearly equivalent to a weight average of finite-time Liapunov exponents over a set of particles distributed along the contour. If a set of contours is considered, anomalously low stretching of a subset may indicate that the subset is embedded in a transport barrier.

In performing such calculations the clearest results are obtained if the contours are initialised in a configuration that is aligned with the flow. The reason for this is that, if a material contour is placed in a flow, its length initially increases rapidly with time whether or not the flow is 'stretching' in the sense of a chaotic mixing flow. Such stretching is just that which would be seen if the contour were placed in a simple linear steady shear. During this initial adjustment material lines tend to align with the flow and eventually, e.g. for a material line in a chaotic mixing region, the initial linear growth is swamped by the exponential growth associated with chaotic stretching. The practical approach that has often been taken to minimize the effects of this initial linear growth is to choose the initial configuration of the contours with PV contours (determined from observed data or from a numerical model as appropriate). The idea is that, since PV is approximately materially conserved, the potential vorticity contours are aligned with the flow in the required manner. A chemical tracer could be used instead of PV. Initialisation along instantaneous streamlines is another possibility.

Such contour-lengthening calculations were first performed by *Bowman* (1993b) and *Norton* (1994), both using single-layer dynamical models of the stratospheric circulation, and *Pierce and Fairlie* (1993) using velocity fields from a coarse resolution three-dimensional model. All these investigations showed clear differences in stretching rates between contours embedded in the vortex and those in the 'surf-zone'. Important applications to observed atmospheric flows have made by *Chen* (1995), for example, who used to contour stretching rates to identify in an objective manner the presence of a transport barrier at the edge of the Antarctic polar vortex and *Waugh et al.* (1994) who showed that large stretching rates sometimes occur inside the Arctic polar vortex, indicating that there may be effective mixing within the vortex, even if there is a strong transport barrier at its edge.

Another important transport barrier is believed to exist on isentropic surfaces that intersect the tropopause, i.e. the notional, quasi-material boundary separating stratosphere and troposphere. On such isentropic surfaces the tropopause is marked by a sharp jump in PV, just as is the case for the stratospheric vortex edge. Indeed it has been argued that the tropopause may form through a similar mechanism to the vortex edge, i.e. through inhomogeneous eddy stirring (*McIntyre and Palmer* 1984, *Ambaum* 1997, *Haynes and Scinocca* 1998). The eddies thought to be important in this case are the 'synoptic' eddies, with scales of a few thousand kilometers that are responsible for day-to-day and week-to-week variations in the weather at midlatitudes. The presence of a transport barrier in this region is required by the strong chemical contrasts between troposphere and stratosphere. If there were no barrier then these strong contrasts could not persist. However it has been remarkably difficult to distinguish such a barrier using the tracer-advection techniques outlined above. *Bithell and Gray* (1997) report that contour advection calculations do not show a clear minimum in stretching rates at the expected location of the tropopause. *Pierrehumbert and Yang* (1993) calculated finite-time Liapunov exponents for a large ensemble of particles on a relevant isentropic surface, but no clear minima in stretching rates are seen in the expected locations. One problem may be that both such diagnostics are, by their very nature, Lagrangian. For this reason they may fail when the transport barrier being considered is rather leaky. For example, if a material contour is placed in the location of a partial barrier, but then subsequently portions of that contour are transported rather rapidly into mixing regions, then the stretching rate will be substantial. Furthermore, there is unlikely to be a strong minimum in stretching as the initial position of the contour is varied. Quantifying the effectiveness of the hypothesised transport barrier at the tropopause remains an important problem if we are to understand fully the processes that control chemical distributions in this part of the atmosphere.

4 Three-dimensional effects

4.1 Beyond isentropic transport

Focussing on transport, stirring and mixing along isentropic surfaces has proved a productive approach and, as noted above, potentially allows exploitation of the large body of theory concerning chaotic advection by two-dimensional time-dependent flows. However, there are many aspects of the atmosphere that cannot be explained by considering isentropic transport alone. Vertical transport across isentropic surfaces or the presence of vertical structure must be included in models if the important physical mechanisms are to be captured.

Here we consider two important ways in which these extra effects play a crucial role. The first concerns the way that cross-isentrope and along-isentrope transport interact to set up the observed variation of chemical tracers in height, latitude and longitude. The second concerns the importance of the formation of small vertical scales in tracer fields as a route to dissipation and mixing.

4.2 The structure of transport in the height-latitude plane

We can now put together a simple picture of the structure of the atmospheric transport in height and latitude. This is depicted in the schematic shown in Figure 8. The inhomogeneity of the transport along isentropic surfaces noted in the previous section suggests that the winter stratosphere should, on each isentropic surface, be divided into 'vortex', 'surf-zone' and 'tropical' regions, separated by (imperfect) barriers to transport. There is evidence from e.g. contour advection studies (*Chen* 1994) that below about 16km (corresponding roughly to the 400K isentropic surface) the transport barrier associated the polar vortex itself weakens and there is free stirring of air parcels between mid and high latitudes. *McIntyre* (1995) calls this the 'sub-vortex' region. In the summer hemisphere of the stratosphere there is no analogue of the vortex, surf-zone structure, since the characteristics of Rossby waves are such that they cannot propagate up into the easterly winds that are present there. However, there is some stirring in the lower stratosphere due to the upward penetration of the effect of the tropospheric synoptic-scale eddies. Finally, as noted earlier, in each hemisphere there is a transport barrier associated with the tropopause.

Superimposed on this picture of transport along isentropes there must be cross-isentropic transport, associated with diabatic heating or cooling. Such heating or cooling is in reality longitudinally varying, but it turns out that we can gain a reasonable qualitative sense of transport if we simply consider the longitudinally averaged cross-isentropic transport, corresponding to the longitudinally averaged diabatic heating or cooling. There is not space here to explain why this is the case (see e.g. *WMO* 1985 [Chapter 6], *Andrews et al.* 1987 [Chapter 9]). Nor is there space to explain how this 'diabatic circulation' arises, except to note that much of it arises not from latitudinal variations in e.g. solar heating, but from momentum transport associated with dissipating Rossby waves and gravity waves. (See e.g. *Andrews et al.* 1987 [Chapter 7], *Haynes et al.* 1991, *Holton et al.* 1995.) In the troposphere and in the lower stratosphere the diabatic vertical velocities are generally upward at low latitudes and downward at high latitudes. A number of different quantitative estimates of the diabatic circulation have been made over the past fifteen years or so. For a recent calculation of the diabatic circulation in the stratosphere see *Rosenlof* (1995), in the troposphere see *Karoly et al.* (1997). [In the troposphere the upward motion at low latitudes is associated with the well-known Hadley circulation. The corresponding upward motion in the stratosphere is sometimes referred to as part of the

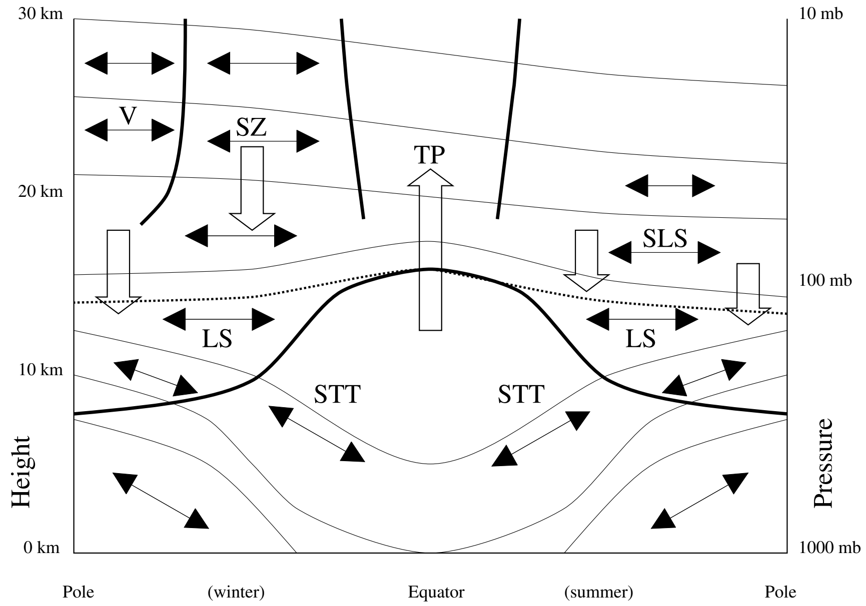


Figure 8: Schematic of the dominant pattern, under solstice conditions of large-scale transport in the troposphere and lower stratosphere . The solid quasi-horizontal lines are isentropic surfaces. Stirring along isentropic surfaces is indicated by quasi-horizontal solid arrows. Diabatic transport across isentropic surfaces is indicated by vertical unfilled arrows. The important transport barriers, namely the vortex edge, the two tropical edges and the tropopause, are denoted by thick solid lines. The lower stratosphere may be divided into six regions, the winter polar vortex (V), the surf zone (SZ), the tropical pipe (TP), the summer lower stratosphere (SLS) and, in each hemisphere, the lowermost stratosphere (LS), meaning that region which lies above the tropopause but is accessible from the troposphere along isentropic surfaces. Each region is shown on the schematic picture. Also marked in each hemisphere is the subtropical upper troposphere (STT), that part of the troposphere which is well away from the ground and accessible from the stratosphere along isentropic surfaces. The upper boundary of the lowermost stratosphere is the isentropic surface that just grazes the tropopause in the tropics and is indicated by the dotted quasi-horizontal line.

'stratospheric Hadley circulation'. But this is potentially misleading. The stratospheric transport is much weaker and the mechanisms for the two circulations are believed to be very different.]

The combination of upwelling, plus relative isolation from mid-latitudes, in the tropics in the lower stratosphere means that it is now common to talk about a 'tropical pipe' (*Plumb* 1996). Chemical tracers with sources in the troposphere, such as N_2O , are taken up through the lower stratosphere in the tropical pipe and gradually leak out into the extratropics. [This is part of the reason why, as noted in §3, N_2O tends to decrease poleward on each isentropic surface. In addition, the higher latitude downwelling tends to bring low values of N_2O down from above, since N_2O has a photochemical sink in the upper stratosphere. The same holds for other tracers with similar distributions of sources and sinks.]

Dividing up the atmosphere in the manner shown in Figure 8, and ideas such as the ‘tropical pipe’ might seem simplistic to an outsider, but they are providing a very useful conceptual framework on which to base studies of transport and within which to interpret chemical measurements. For example, a number of recent papers have used different combinations of observational data and simple transport models to estimate the timescales for air parcels to leak into or out of the tropics (e.g. *Mote et al.* 1996, *Minschwaner et al.* 1996, *Volk et al.* 1996, *Hall and Waugh* 1997).

4.3 Vertical dispersion

Many of the earliest models that included transport in an attempt to explain chemical observations were one-dimensional in space, representing vertical but not horizontal variation. In part this was because many of the early chemical observations were of the form of vertical profiles and therefore contained information only about vertical variation. The transport parametrization in such models was diffusive. Of course, in the light of the picture of the circulation described above, the shortcomings of such a transport parametrization are clear. The next generation of chemical models typically resolved both height and latitude variations and included a more physically plausible representation of transport. Again some simplification was sought and it was argued that along-isentrope transport is dominated by the effects of eddies (i.e. circulation features with a strong longitudinal variation) which, in a longitudinally averaged, view lead to dispersion along isentropes and might be represented by an eddy diffusion. The cross-isentrope transport, on the other hand, was argued to be dominated by advection by the longitudinally averaged circulation and might therefore be represented by an advective transport. (see e.g. *Tung* 1982, *Mahlman et al.* 1984, WMO 1985, *Plumb and Mahlman* 1987). This is close to the picture outlined above, except that the strong spatial inhomogeneity of the stirring along isentropic surfaces observed in reality suggests that approximating it by eddy diffusion may be seriously flawed.

The two ingredients of latitudinal diffusion (on a latitudinally confined domain) and vertical advection by a latitudinally varying velocity field correspond exactly to those in the classical fluid-dynamical problem of shear dispersion in a pipe or channel. As shown by G.I. Taylor, the net effect is diffusive dispersion, now referred to as Taylor dispersion, along the pipe. It is similarly the case that, under certain conditions, the vertical transport of globally averaged quantities can be described by a downgradient diffusion (*Holton* 1986, *Mahlman et al.* 1986, *Plumb and Ko* 1992). This result gives some basis for interpreting the results from the early one-dimensional chemical models. The vertical diffusion in those models may be argued to capture the effect of the large-scale circulation, including both the eddies and the longitudinal mean circulation, on global average values of tracers. *Plumb* (1996) refers to these as 'global-diffuser' models. But, of course, it is not at all clear whether such models can be usefully used to interpret chemical profiles taken at individual horizontal locations.

The exact mathematical correspondence between the Taylor problem and that of vertical advection plus latitudinal diffusion in a longitudinally averaged chemical transport model makes it inevitable that in the corresponding globally averaged problem the vertical transport is diffusive. What is in some ways more interesting is to consider whether the diffusive vertical transport persists in more general circumstances, for example, when the latitudinal transport is not diffusive, that might be relevant to the real atmosphere rather than just to idealised models. In the classical pipe problem the cross-pipe diffusion means, in effect, that molecules of tracer follow a random walk in the cross-pipe direction. As they do so, they sample what is, in effect, a random time series of along-pipe velocity and therefore also follow a random walk in the along-pipe direction. On time scales longer than the decorrelation time for the along-pipe velocity experienced by each molecule, equivalent to the cross-pipe diffusion time, this random walk in the along-pipe direction is diffusive.

What is required for diffusive vertical dispersion of an ensemble of particles is thus that each particle experiences a random time series of vertical velocity and that the statistical properties of the time series are identical over all particles. It is not necessarily required that horizontal transport be diffusive. Recent calculations by *Sparling et al.* (1997) have considered the dispersion of particles across isentropic surfaces and have shown when such dispersion is likely to be diffusive.

Figure 9 is taken from *Sparling et al.* (1997) and shows the final positions in the height-latitude plane (with height being defined by potential temperature) of a large set of particles. The particles were all started on the 500K surface and were advected for a period of 60 days. Horizonal velocities were taken directly from large-scale meteorological datasets. Vertical velocities (relative to isentropic surfaces and hence proportional to diabatic heating rates) were calculated using relatively sophisticated radiation schemes (of the type used in atmospheric general circulation models), taking temperatures from the meteorological datasets. The period covered was 1 January 1993 to 1st March 1993 (i.e. in Northern Hemisphere winter). Since such calculations necessarily depend on the quality of the meteorological dataset and the radiative scheme, results from two independent calculations are displayed, each using different meteorological datasets and radiative schemes.

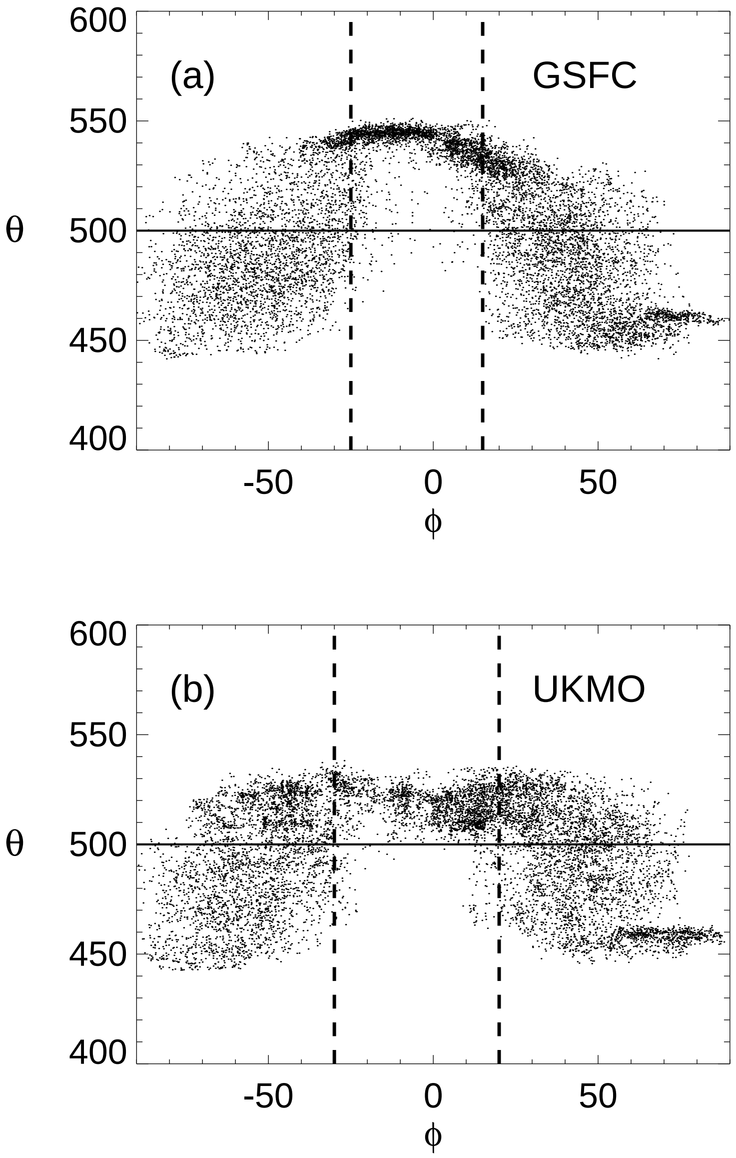


Figure 9: Particle positions after 2 months projected onto the θ (potential temperature) - ϕ (latitude) plane. Particles were initialised on the 500K potential surface and then advected by winds calculated from meteorological data for the period 1 Jan 1993 to 28 Feb 1993. (Vertical winds relative to isentropic surfaces, which are proportional to heating rates, were calculated using a radiative code.) Results from two independent calculations based on different datasets, Goddard Space Flight Center (GSFC) and UK Meteorological Office (UKMO), and radiative codes are displayed. The dashed lines show the approximate boundaries of the tropical subensemble of particles. [Figure reproduced from *Sparling et al.* (1997).]

The first thing that is clear from these pictures is the large-scale latitudinal pattern of upwelling and downwelling. Particles in the tropics have ascended and particles in mid and high latitudes have, on average, descended. However, particularly for the particles in Northern Hemisphere mid-latitudes and also for those in the Southern Hemisphere extratropics, there has been considerable vertical dispersion. This dispersion results because different particles experience different time series of diabatic heating. *Sparling et al.* (1997) show that this is primarily because of particles moving latitudinally across a strong latitudinal gradient in diabatic heating, though there is also a contribution from longitudinal and temporal variations. The largest latitudinal displacements are in the mid-latitude winter hemisphere, i.e. in the surf zone, and in the summer hemisphere, where as noted earlier, there is stirring in the lower stratosphere. The small vertical dispersion at high latitudes in the winter hemisphere is a manifestation of the trapping of particles, which therefore undergo only limited latitudinal displacement, in the polar vortex.

The time variation of the vertical dispersion within different ensembles of particles, measured by the variance of potential temperature within such ensembles, is shown in Figure 10, also taken from *Sparling et al.* (1997). As is well-known from classicial work on turbulent dispersion, under many conditions the variance of displacement over an ensemble of particles increases quadratically with time at early times (the so-called 'advective' regime) and then at later times increases linearly with time (the so-called 'diffusive' regime). The transition from one regime to another occurs on a time scale roughly equal to the Lagrangian deorrelation time. The results in Figure 10 show that ensembles of particles that stay in the surf zone, or in the extratropical summer hemisphere, display this transition within the 60-day time period of the integration. However, the global ensemble, of all parcels, shows advective dispersion for the whole time period. This is because, as could be inferred from Figure 9, some of the particles remain trapped in the tropics, others remain trapped in the vortex, etc. In the language used earlier, the decorrelation time for the vertical velocity for the global ensemble, i.e. the time taken for particles to 'forget' the vertical velocity with which they started is certainly larger than 60 days. Hence the one-dimensional 'global diffuser' models, described earlier, for the vertical variation of trace species, can be valid only on timescales substantially longer than this. On the other hand, models that distinguish between say the surf-zone and tropics, have the prospect of being valid on shorter time scales, since the decorrelation time scales within these individual regions are less than the global decorrelation time.

Plumb (1996) has generalised the one-dimensional models to represent both a 'tropical pipe' where chemical species are upwelled from the troposphere and an extratropical region where the transport is assumed to have the same character as in the 'global-diffuser' models. The only difference is that there can be a net vertical mass flux in the extratropics (balanced by an opposing flux in the tropical pipe). This means that the vertical transport of the horizontally averaged chemical concentrations appears as a combinatin of

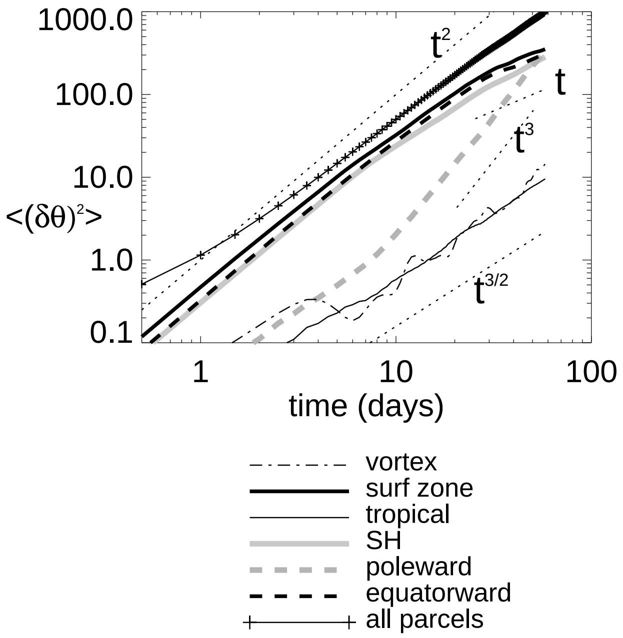


Figure 10: Time evolution of the potential temperature variance of different subensenembles, according to the GSFC calculation. Various powers of time t are indicated on the graph to allow comparison. [Figure reproduced from *Sparling et al.* (1997).

advection and shear-diffusion. This model has the advantage that may predict different chemical distributions in tropics and extratropics, reflecting the strong differences between these regions that are observed. The validity of the diffusive approximation for vertical transport, particularly the lack of dependence of diffusive transport in the horizontal, is further discussed in Thuburn and McIntyre (1997), who derive general conditions under which the approximation is valid.

4.4 Three-dimensional routes to mixing

The second topic of this section concerns routes to mixing in the real atmosphere. We define mixing to mean the physical processes by which two distinct fluid elements, e.g. with distinct chemical characteristics, mix at the molecular level, i.e. essentially become one homogeneous fluid element. This definition is a little different to that which is sometimes used in the dynamical systems context (e.g. see *Aref* 1991). In particular, under this definition mixing cannot arise purely through advection, but requires molecular diffusion to homogenize tracer concentrations at the molecular level. Of course, the stirring action of advection plays a vital catalytic role in mixing, by stretching and distorting fluid elements so that spatial gradients and hence the effect of diffusion are systematically enhanced.

The techniques, such as contour advection, described in §2 for simulating the evolution of tracer fields on isentropic surfaces have played an important role in atmospheric science by allowing visualisation of the formation, through the stirring processes, of fine-scale horizontal structure in tracer fields. However, in order to appreciate the precise significance of these fine-scale structures for mixing, it is necessary to consider the mixing processes in a little more detail.

Processes that lead to dissipation and mixing are generally quasi-isotropic in the sense that, for a structure of a given length scale they do not differentiate by direction. For example, two neighbouring temperature anomalies, one warm, one cold, separated by 100 meters, say, might be expected to affect each other through radiative transfer, but it will not matter whether the separation is in the vertical or the horizontal. Other similarly isotropic physical processes are molecular diffusion in a laminar flow and mixing due to enhanced molecular diffusion in three-dimensionally turbulent flow. In principle scale reduction in the horizontal will lead eventually to dissipation or mixing. However it is clear from observational experience that structures in the atmosphere and the ocean tend to have much smaller length scales in the vertical than in the horizontal. This suggests that mixing and dissipation are most likely to result from small vertical scales rather than small horizontal scales. Is there a systematic reduction in vertical scales, as well as in horizontal scales, due the large-scale flow? This question has recently been investigated by Haynes and Anglade (1997) and some of the key arguments in that paper are repeated again below.

Certainly the large-scale flow is not isotropic. As has been noted earlier, cross-isentrope velocities are much smaller than along-isentrope velocities. This suggests that a suitable model for analysing the scale-cascade process might take a velocity field that is purely horizontal (representing the flow along isentropic surfaces), but which varies in both the horizontal and the vertical. As has already been noted in §3, the effect of chaotic advection along isentropic surfaces is to reduce horizontal scales at an exponential rate. The extra effect that needs to be included is that of vertical variation of the horizontal flow, i.e. vertical shear.

It turns out that the relevant effects are captured by the simple linear flow with components $(\Gamma x, -\Gamma y + \Lambda z, 0)$ in the x, y and z directions respectively, where (x, y, z) are Cartesian coordinates, with z vertical. For simplicity we assume the flow to be steady and therefore take the horizontal strain Γ and the vertical shear Λ to be constants. Exponential reduction in scale in the horizontal at rate Γ is therefore accomplished by the steady horizontal strain. The equation for the evolution in this flow of a tracer with concentration $\chi(x,y,z,t)$ is

$$\frac{\partial\chi}{\partial t}+\Gamma x\frac{\partial\chi}{\partial x}+(\Lambda z-\Gamma y)\frac{\partial\chi}{\partial y}=\kappa(\frac{\partial^2\chi}{\partial x^2}+\frac{\partial^2\chi}{\partial y^2}+\frac{\partial^2\chi}{\partial z^2}), \tag{7}$$

where κ is the diffusivity, assumed constant. This equation has solutions of the form $\chi(x,y,z,t) = \mathrm{Re}\{\hat{\chi}(t)\exp(i[kx+ly+mz])\}$ where k, l and m are functions of time satisfying the ordinary differential equations $\dot{k}+\Gamma k=0$, $\dot{l}-\Gamma l=0$ and $\dot{m}+\Lambda l=0$. It follows that if $(k,l,m)=(k_0,l_0,m_0)$ at $t=0$ then

$$(k,l,m)=(k_0e^{-\Gamma t},l_0e^{\Gamma t},m_0-\frac{\Lambda l_0}{\Gamma}(e^{\Gamma t}-1)). \tag{8}$$

Reduction in the scale of variation of a tracer in this flow may be deduced by considering the time variation of the wavenumber vector (k,l,m). Note that at large times the size of the horizontal wavenumber $(k^2+l^2)^{1/2}$ increase exponentially with time at rate Γ, indicating the expected reduction in horizontal scale associated with the steady horizontal strain flow. However, the inclusion of vertical shear means that vertical wavenumber m also increases exponentially at the same rate, implying an accompanying reduction in vertical scale cascade. The aspect ratio α, say, of horizontal to vertical scales defined by $\alpha=|m|/(k^2+l^2)^{1/2}$ is equal, at large times, to Λ/Γ, i.e. the ratio of vertical shear to horizontal strain. The reason for this is quite clear if one considers the geometry of the flow, as depicted in Figure 11. The effect of adding the vertical shear to the horizontal strain is to cause the axis of strain to slope with height. The angle between the axis and the horizontal is the inverse tangent of Γ/Λ. Convergence of the flow at each level naturally leads to structures in tracer fields that slope at the same angle. These structures thin exponentially fast in the horizontal and therefore also in the vertical.

The implications for diffusive mixing are manifested in the time evolution of the coefficient $\hat{\chi}$ which satisfies the equation

$$\frac{d\hat{\chi}}{dt}=-\kappa(k^2+l^2+m^2)\hat{\chi}. \tag{9}$$

Substituting from (8) and integrating, it follows that

$$\log(\hat{\chi}(t))-\log(\hat{\chi}(0))=$$

$$-\kappa\{\frac{l_0^2}{2\Gamma}(1+\frac{\Lambda^2}{\Gamma^2})(e^{2\Gamma t}-1)+\frac{2l_0\Lambda}{\Gamma^2}(e^{\Gamma t}-1)(m_0+\frac{\Lambda l_0}{\Gamma})+(m_0+\frac{\Lambda l_0}{\Gamma})^2t+\frac{k_0^2}{2\Gamma}(1-e^{-2\Gamma t})\}. \tag{10}$$

The term in brackets on the right-hand side increases with time and diffusive decay becomes important when this term $\sim\kappa^{-1}$. If κ is sufficiently small then the time at

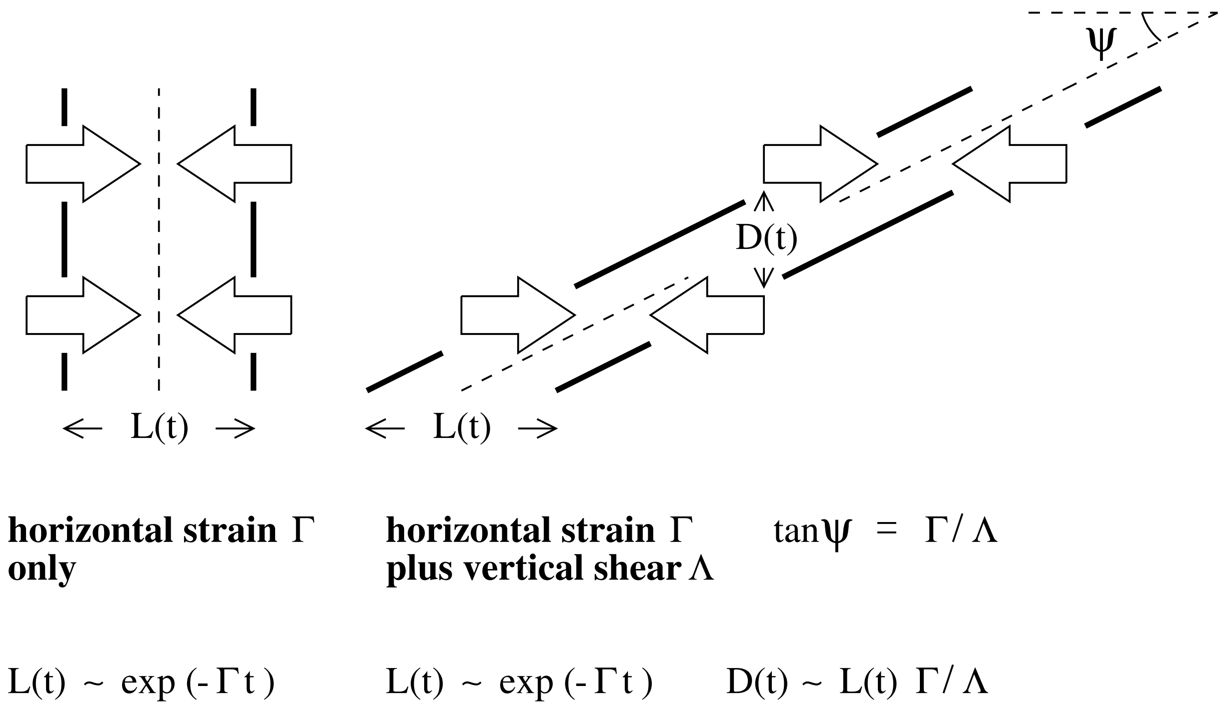


Figure 11: Illustration of the basic mechanism by which vertical scales are reduced in a flow containing a combination of horizontal strain and vertical shear. The effect is shown of such a flow on a tracer sheet, with tracer contours represented by the bold lines. The flow could be that considered in §4.4, in which case the view is along the x-axis, which is the axis of extension for this flow. The left-hand picture shows the effect of a flow including horizontal strain alone. The horizontal thickness L of the tracer sheet is reduced in time as $e^{-\Gamma t}$, where Γ is the horizontal strain rate. The right-hand picture shows the effect when a flow in the y direction with shear Λ in z is added. The axis of the strain field is now tilted to the horizontal at angle $\tan^{-1}(\Gamma/\Lambda)$, as is the resulting tracer sheet. The horizontal thickness L and the vertical thickness D therefore both decrease as $e^{-\Gamma t}$ and stay in the aspect ratio Λ/Γ. [Figure reproduced from *Haynes and Anglade* (1997). Further calculations in that paper show how this effect persists when the horizontal strain and vertical shear fields are time dependent.]

which this occurs will be determined by the first term inside the bracket, which is most rapidly growing for large time. Note first that this term is independent of m_0 since, for sufficiently small diffusivity, it is not the initial vertical structure, but the small vertical structure that arises from tilting of initial horizontal structure, that controls diffusive effects. Note also that this term is increased by a factor $1 + \Lambda^2/\Gamma^2$ over the form it would have in the absence of vertical shear. The diffusive decay, then, is almost exactly as it would be in a problem with no vertical structure, with the effects of horizontal diffusion enhanced by the scale reduction associated with horizontal strain. However, the effect of the vertical shear is to augment the horizontal diffusivity κ by an amount $\kappa\Lambda^2/\Gamma^2 = \kappa\alpha^2$ due to the diffusivity acting on structure of aspect ratio $\alpha = \Lambda/\Gamma$. This augmentation of the horizontal diffusivity is closely analogous to that found by Young et al. (1982) due to the effects of oscillatory vertical shear (argued to result from near-inertial waves). In the simple model reported above the vertical shear should be regarded as that associated with the large-scale flow.

The assumption of steady velocity fields in the above model is, of course, a drastic one. However, Haynes and Anglade (1997) show how similar behaviour is found in more general flows, by integrating the equation

$$\frac{d\mathbf{k}}{dt} = -(\nabla\mathbf{u}).\mathbf{k} \tag{11}$$

for the gradient $\mathbf{k}$ of a conserved tracer, measured following a fluid particle, given the velocity field $\mathbf{u}$. One approach is to impose the components of the velocity gradient tensor $\nabla\mathbf{u}$, i.e. the horizontal strain and vertical shear, as random functions of time. This has been used classically to study tracer evolution in turbulent flows, but might also be considered as a simple representation of the time varying stretching and shearing encountered by a fluid element as it moves in a chaotic advection flow. A second approach is to calculate $\mathbf{u}$ from observations (from the wintertime lower stratosphere). In both cases both horizontal and vertical tracer gradients are found, on average, to increase exponentially at the same rate and to attain a characteristic aspect ratio. For observed wintertime lower stratospheric flows the aspect ratio α is about 250.

4.5 Implications for stratospheric mixing

Assessment of the strength of mixing processes in the stratosphere (and elsewhere in the atmosphere) is important not only because of fundamental interest, but also because of implications for chemical evolution. Recent investigations have suggested that there may be sensitivity of stratospheric chemical evolution in a gross sense (e.g. in the total amount of reaction product or the total amount of a given species destroyed) to the strength of mixing processes (e.g. *Edouard et al.* 1996, *Tan et al.* 1998). In the *Edouard et al.* (1996) case this sensitivity is expressed in terms of sensitivity to numerical resolution. However, any numerical model with finite resolution (however cleverly its transport scheme has been designed) is based on the implicit assumption that chemical species are well-mixed on the scale of the grid. If the grid size is larger than the scale on which mixing actually occurs then the effect is equivalent to artificially enhancing the strength of mixing processes.

The molecular mixing required for chemical reaction, for example, depends ultimately on molecular diffusion. As is well known, the effects of molecular diffusion can be enhanced by fluid-dynamical stirring. Up to now we have considered the stirring effects of chaotic-advection flows. However, as noted earlier, the lower stratospheric flow is not entirely laminar but is believed, on the basis of balloon, aircraft and radar observations, to contain patches of three-dimensional turbulence. The effect of these patches may spectacularly enhance mixing on small scales. *Dewan* (1981) has argued that the effects of the turbulent patches will be to give a diffusive transport in the vertical, associated with a diffusivity D say, essentially because fluid parcels encountering a succession of such patches will undergo a random walk with small vertical step size. We might further hypothesize that representing this transport by downgradient diffusion also captures the mixing effects of the patches – this seems plausible if the turbulent patches become well mixed within their lifetime. (It is useful to contrast the case of the 'diabatic dispersion' discussed earlier, where the dispersive transport might under some conditions be parameterizable by downgradient diffusion, but is not likely, by itself, to lead to an equivalent molecular mixing. In the language sometimes used in other areas of fluid dynamics, diabatic dispersion leads to 'macromixing,' but not to 'micromixing'.)

One way to estimate the strength of the mixing processes and to understand their effects, therefore, is to estimate the vertical diffusivity D and to consider the interaction of the vertical diffusion with the large-scale 'chaotic-advection' flow. The arguments of *Haynes and Anglade* (1997) suggest that the latter interation will lead to an effective horizontal diffusivity $\alpha^2 D$, where α is the aspect ratio. Thus tracer evolution on a quasi-horizontal surface might be explained at leading order, by two-dimensional chaotic advection with diffusivity $\alpha^2 D$. If a typical stretching rate is S, then the minimum horizontal scales seen in such flows, i.e. the horizontal scales at which mixing occurs, are expected to be of order $\alpha(D/S)^{1/2}$.

A number of estimates of the vertical diffusivity D are available from calculations based on radar data. Some of these (e.g. *Fukao et al.* 1994) are based on hypothesised relations between turbulent energy dissipation rates and vertical eddy diffusivities. That of *Woodman and Rastogi* (1984) is based on the model posed in *Dewan* (1981). However most of these calculations seem to give a value of around 0.2 $\mathrm{m^2 s^{-1}}$. A lower bound on D is certainly the molecular diffusivity, which is about $1.5 \times 10^{-4} \mathrm{m^2 s^{-1}}$. These two values for D, together with the *Haynes and Anglade* (1997) estimate of 250 for the aspect ratio α and a stretching rate S of about 0.3 $\mathrm{days^{-1}}$, imply minimum horizontal scales of about 50km and 2km respectively. As noted earlier, there is considerable evidence from aircraft data of small-scale horizontal variation of chemical tracers and it does not seem to be the case that this variation is strongly suppressed below scales of 50km (recall Figure 6). The aircraft data is, indeed, itself a potential source of information about mixing processes and two recent pieces of work, by *Waugh et al.* (1997) and *Balluch and Haynes* (1997), have attempted to estimate D from this data. Both conclude, in broad terms, that D is about $10^{-2}\mathrm{m}^2\mathrm{s}^{-1}$, i.e. considerably less than the radar estimates. This is a troubling discrepancy that it seems important to resolve. Progress is likely to be complicated by the fact that the mixing due to patches of turbulence is almost certainly highly variable in time and space. The value for D of $10^{-2}\mathrm{m}^2\mathrm{s}^{-1}$, which would imply a minimum horizontal scale of about 12km, might represent the 'average' effect of mixing processes. *Balluch and Haynes* (1997) show there is 'believeable' variation in the chemical tracers down to scales of order 1km and suggest that these might be associated with air parcels that have encountered relatively few turbulent patches and hence relatively weak mixing.

5 Discussion

5.1 Summary

It has been argued in the above that transport in the atmosphere is dominated by advection by the large-scale flow and that, at least for time scales of a few days, the transport is, quasi-horizontal, along isentropic surfaces. Thus two-dimensional chaotic advection is an appropriate paradigm in the sense that complicated spatial structure arises through the action of flow that has a rather simple space and time variation. One might describe the flow as layerwise two-dimensional chaotic advection. The structure of the transport on each isentropic surface shows the same sort of strong spatial inhomogeneity, with mixing regions and transport barriers, that is now seen as characteristic of 2-D chaotic advection.

If the description of transport is to be any better than semi-quantitative, then the third space dimension must be taken into account. In particular, if transport on longer time scales is of interest then it is essential to take account of vertical advection across isentropic surfaces. As is well known from the work of *Dombre et al.* (1986) and others, in three dimensions there may be chaotic advection without any time-dependence in the flow. However, it is not very clear studying three-dimensional steady flows will give much insight into the real atmosphere. A more interesting possibility is to add weak vertical advection to otherwise two-dimensional chaotic advection flows. Work on three-dimensional time-dependent flows (e.g. *Feingold et al.* 1988, see also *Cartwright et al.* 1996) suggests that there may be interesting effects in such flows. Whether these effects are relevant to transport in realistic atmospheric flows has yet to be determined. If realistic mixing processes are of interest, then the vertical variation of the flow must be taken into account, since this results in a reduction in vertical scale, as well as in horizontal scale, of tracer fields and it is the vertical-scale reduction that is most significant for mixing.

The remainder of this section discusses some examples of topics in atmospheric transport, stirring and mixing that seem particularly interesting to the author, both in their importance for understanding the atmosphere and in their relation to problems addressed by theoretical work on chaotic advection.

5.2 Dynamical consistency

One aspect of chaotic advection that many working on atmospheric (or indeed oceanic) flows find troubling is the fact that almost all problems are posed kinematically, i.e. the velocity field is taken as a given function of space and time, without any requirement for dynamical consistency. As has been noted in §2, the evolution of large-scale atmospheric and oceanic flows may be understood in terms of an advected scalar (PV) whose distribution at any instant determines the flow at that instant. If a flow is to be time-periodic, for example, then clearly the PV field also has to be time-periodic. It is not clear that this is consistent with the existence of chaotic advection regions in the flow unless the PV (and hence the flow) satisfies certain strong constraints. For example, *Brown and Samelson* (1994) have suggested that it is necessary for the potential vorticity to be constant in the chaotic advection regions. One way round this problem is to consider special flows where the potential vorticity (perhaps in a certain localised region) happens to be a passive tracer, e.g. the nonlinear Rossby-wave critical layer flow (*Ngan and Shepherd* 1997a). While such special cases are useful in justifying the results of kinematic studies as, in some sense, dynamically realisable, they do not, of course, provide any insight into what actually happens in the much larger class of problems where PV is active, rather than passive.

More generally, the properties of the 'inversion' operator appearing in e.g. equations (2) and (5) mean that small-scale features in vorticity or potential vorticity have a relatively weak influence on the flow. Thus the stirring and mixing of vorticity or potential vorticity at small scales is likely to be much as predicted for stirring and mixing of passive tracer by chaotic advection studies, provided that the structure of the large-scale flow is similar. This is nicely discussed and illustrated by *Pierrehumbert* (1991b) who shows results from an initial-value dynamical simulation and notes how the structures arising in the small-scale vorticity field correspond in some detail to those predicted by a kinematic model.

One way to assess which parts of the large body of results for two-dimensional time-periodic flow carry over to the dynamically consistent case is to consider the effects of more general time-dependence (the idea being that the requirement of dynamical consistency will almost always disrupt time-periodicity). There has already been some progress in analysing transport, for example, in the quasi-periodic case (i.e. two or more periods in irrational ratios) (e.g. *Duan and Wiggins* 1996) and even with the aperiodic case (*Malhotra and Wiggins* 1997).

Take, for example, the interesting issue of the 'stickiness' of the regular trajectories that border a chaotic region (e.g. *Meiss* 1992 sections II.D.4 and IX.C.3). These have implications both for transport and for stirring and mixing. For the former, the 'stickiness' implies long-term correlations in velocities that may lead, e.g. to algebraic tails in the probability distributions for time of escape from certain regions, perhaps in turn leading to the possibility of anomalous diffusive effects. For the latter, it implies that the distribution of the cumulative stretching experienced by fluid elements has a larger than expected tail on the low stretching side [see *Pierrehumbert* 1991a and references). This might, for example, have implications for the diffusive decay of tracer variance. If 'stickiness' is a property that only holds for periodic velocity fields, or some other relatively small class of velocity fields, then it and the associated transport, stirring and mixing behaviour, might not be observed for dynamically consistent flows.

5.3 Microstructure

Mathematical models for the tracer microstructure that results from chaotic advection are important for interpretation of small-scale tracer measurements. They are also a useful preliminary step towards understanding chemical evolution or the effects of other processes that depend nonlinearly on tracer concentrations.

One rather crude measure of microstructure is the tracer spectrum. Recent calculations have shown that the horizontal spectra of tracers measured during the various aircraft ozone campaigns has a slope somewhere between -2 and -1.5 (*Strahan and Mahlman* 1994, *Bacmeister et al.* 1996). Theories of tracer behaviour in a large-scale flow fields suggest that in the forced-dissipative case the slope should be -1 (the 'Batchelor' spectrum) [e.g. *Pierrehumbert* (1995) and references] and indeed this has been verified in numerical simulations by *Antonsen et al.* (1996). *Bacmeister et al.* (1996) have suggested that in order to explain the steeper slope of the observed spectra it may be necessary to take account of both advection by the large-scale flow and the advective effect of gravity waves on small-vertical-scale tracer features produced by the large-scale flow. *Ngan and Shepherd* (1997b), on the other hand, have argued that the form of the spectrum may be due to the fact that the dominant tracer features are near discontinuities associated with the vortex edge (and filaments drawn away from it) and that these near discontinuities remain spatially isolated, because dissipative processes are strong enough to remove filaments before they become space-filling. The *Saffman* (1971) model, predicting a -2 slope for the specrum is therefore relevant. Of course, the important scientific question here is what physical processes need to be invoked to explain the spectrum? The *Bacmeister et al.* (1996) suggestion requires more than large-scale advection and simple dissipation, the *Ngan and Shepherd* (1997b) suggestion does not.

Different approaches to quantifying microstructure have, of course, been suggested. *Pierrehumbert* (1995) discusses a number of relevant issues, including the possibility, argued by *Varosi et al.* (1991), that chaotic advection flows may lead to multifractal scaling of tracer gradients. Pierrehumbert does not find such scaling in his numerical simulations and notes the need for further examination of this issue. He does find scaling consistent with tracer fields that are filamentary (i.e., locally one-dimensional) with appreciable spatial correlations across filaments. Certainly filamentary structure is one of the striking features of visualizations of tracers in chaotic-advection flows (and is in strong contrast to the appearance of tracers in fully three-dimensional turbulent flow.) *Balluch and Haynes* (1997) give a heuristic argument for this locally one-dimensional structure, based on the fact that the direction (but not the magnitude) of the tracer gradient at some spatial location and time is determined only over the recent history (a few stretching times) and therefore varies only on the large scale. They note that the analogous structure for 'layerwise two-dimensional chaotic advection' consists of tracer sheets rather than tracer filaments and exploit this in their assessment of the effects of vertical mixing. It might be that this approach could be incorporated into a more sophisticated representation of microstructure.

5.4 Transport barriers

The partial barriers to transport between tropics and surf zone in the lower stratosphere, or at the tropopause, play an important role in determining the distribution of chemical tracers. Partial barriers to transport play similarly important roles in the ocean. For example, it has been noted by *Bower et al.* (1985) that at upper levels there are strong water mass contrasts across the Gulf stream suggesting that it acts as a barrier to transport. At lower levels, on the other hand, there is little contrast, presumably because the eddy motion associated with the Gulf Stream acts as an efficient 'blender'.

Both atmospheric and oceanic problems have already stimulated kinematic studies of transport associated with disturbed jets (*Samelson* 1992, *Duan and Wiggins* 1994, *del-Castillo-Negrette and Morrison* 1996). How can such studies be exploited or extended? What one might seek is some insight into what controls the leakiness of barriers in realistic flows. Consider, for example, a two-dimensional time-periodic flow where there may be perfect barriers to transport. What ingredient of the real atmospheric flow would it be most relevant to add to such a model in order to give realistic leakiness? Adding weak vertical advection is one possibility. Another might be more complex time-dependence. But in the latter case is it weak random fluctuations that are most important or occasional large-amplitude disturbances? One practical approach to addressing these questions for the Southern Hemisphere polar vortex has been made by *Bowman* (1996), who uses observed winds on isentropic surfaces to study the effect on the transport barrier at the vortex edge of applying different time and space filters to the wind fields.

If, in model simulations, such barriers are too 'leaky', tracer distributions are likely to be incorrectly predicted. In the case of tracers such as ozone and water vapour, which play a crucial role in radiative transfer, such errors might feed back on the simulation of the circulation itself.

5.5 Conclusion

There is no doubt that the development of chaotic advection as a sub-discipline of fluid dynamics has given important stimulus to understanding transport, stirring and mixing in atmospheric flows. It is also the case that there is an appealing analogy between the structures typically generated in chaotic advection models and those that have been inferred from new atmospheric observations and numerical simulations over the last decade or so. However, it still seems arguable that the insight provided by the chaotic advection theories is only qualitative. The challenge in the future is to determine to what extent the mathematical apparatus that has been constructed to study nonlinear dynamical systems, in particular chaotic advection, can be used to give hard, quantitative insight into transport, stirring and mixing in realistic atmospheric flows. The examples given above are some suggestions of where progress would certainly be significant and might also be feasible.

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