Stratospheric Transport

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Abstract. Improvements in our understanding of transport processes in the stratosphere have progressed hand in hand with advances in understanding of stratospheric dynamics and with accumulating remote and *in situ* observations of the distributions of, and relationships between, stratospheric tracers. It is convenient to regard the stratosphere as being separated into four regions: the summer hemisphere, the tropics, the wintertime midlatitude "surf zone", and the winter polar vortex. Stratospheric transport is dominated by mean diabatic advection (upwelling in the tropics, downwelling in the surf zone and the vortex) and, especially, by rapid isentropic stirring within the surf zone. These characteristics determine the global-scale distributions of tracers, and their mutual relationships. Despite our much-improved understanding of these processes, many chemical transport models still appear to exhibit significant shortcomings in simulating stratospheric transport, as is evidenced by their tendency to underestimate the age of stratospheric air.

1. Introduction

It has long been recognized that stratospheric trace gases with sufficiently weak chemical sources and sinks have similar global distributions, in the sense that their isopleths (surfaces of constant mixing ratio) have similar spatial shapes. This was evident from some of the earliest global observations of CH_4 and N_2O from the SAMS instrument on the Nimbus 6 satellite (Jones and Pyle, 1984) and has been confirmed (albeit with some reservations, as we shall discuss below) in subsequent observations and modeling studies of many such tracers. Examples of these characteristic shapes are shown in Fig. 1 for monthly mean distributions around the equinoxes. The two tracers shown have opposite large-scale gradients (HF has a stratospheric source and tropospheric sink, CH_4 a tropospheric source and stratospheric sink); nevertheless, the shapes of their isopleths are very similar, with isopleths bulging upward in the tropics and poleward/downward slopes in the extratropics.. Especially in the winter or spring hemisphere, there are strong horizontal gradients in the subtropics, relatively flat isopleths in middle latitudes, and strong gradients again near 60° latitude in the winter hemisphere. The similarity of structure between two chemically unrelated tracers is strong evidence that these characteristics are determined by transport, and thus by stratospheric (and mesospheric) dynamics.

Comprehensive perspectives of the global-scale dynamics of the stratospheric circulation have been given elsewhere (*e.g.*, Holton et al. (1995), to which the reader is referred for details and for original references). For present purposes, we will just summarize the main points that are relevant to trace gas transport.

2. Overview of stratospheric circulation and transport

The stratospheric circulation

The meridional circulation of the stratosphere, known as the "Brewer-Dobson" circulation, after the pioneering deductions of Brewer (1949) and Dobson (1956) from observations of stratospheric water vapor and ozone, respectively, comprises a two-cell structure in the lower stratosphere, with upwelling in the tropics and subsidence in middle and high latitudes, and a single cell from the tropics into the winter hemisphere at higher

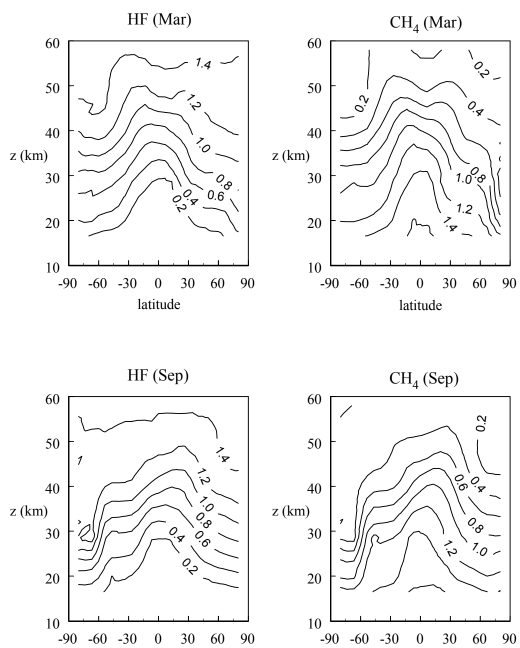

Figure 1. Monthly mean meridional distributions of (left two panels) HF (parts per billion by volume) and of (right two panels) CH_4 (parts per million by volume) during March (top) and September (bottom), from measurements by the HALOE instrument (Russell et al., 1993a) on the Upper Atmosphere Research Satellite. Calculation of the monthly mean distributions for these data is described in Randel et al. (1998).

altitudes. These characteristics have been confirmed from modern radiation calculations based on observed distributions of temperature and radiatively active constituents (Rosenlof, 1995; Eluszkiewicz et al., 1996), provided one interprets "mean circulation" as "residual mean circulation", rather than the straightforward Eulerian mean (it is the residual mean that is relevant to tracer transport [*e.g.* Andrews et al., 1987)). A schematic of the residual circulation of the atmosphere (up to the mesopause) is depicted in Fig. 2. In the

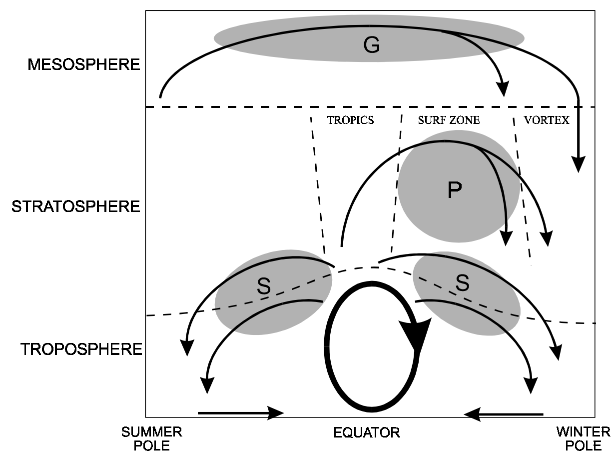

Figure 2. Schematic of the residual mean meridional circulation in the atmosphere. The heavy ellipse denotes the thermally-driven Hadley circulation of the troposphere. The shaded regions (labelled "S", "P", and "G") denote regions of breaking waves (synoptic- and planetary-scale waves, and gravity waves, respectively), responsible for driving branches of the stratospheric and mesospheric circulation.

tropical troposphere, the well-known Hadley circulation can be understood, at least in its simplest form, as a nonlinear circulation driven by latitudinal gradients in thermal forcing (Held and Hou, 1980). Despite the temptation to interpret the apparently thermally-direct circulation of the stratosphere in the same way, it is clear that other processes must be involved, since air following the circulation must lose angular momentum as it moves poleward. The direct, large-scale, effects of friction being utterly negligible in the stratosphere, such loss of angular momentum can only be ascribed to the impact of waves: the presence of wave drag is thus crucial to the stratospheric circulation.

In contrast to the much more quiescent summer hemisphere, the wintertime stratosphere is dominated by large-amplitude, planetary-scale Rossby waves propagating upward from the troposphere. Intermittently, these waves break, stirring air more or less isentropically across large distances of the winter stratosphere within a region that has become known as the "surf zone" (McIntyre and Palmer, 1983), bounded by sharp gradients of potential vorticity (PV) and of tracers in the winter subtropics and at the edge of the polar vortex. Apart from the direct effects of this stirring on tracers, it has an indirect effect through the meridional circulation induced by the stirring of PV. For adiabatic, inviscid flow, and assuming steady state (*i.e.* solstice conditions), the zonal mean momentum budget can be written (*e.g.*, Andrews et al., 1987)

$$-\bar{v}^* \bar{P}^* = \overline{\hat{v}\hat{P}}^* \tag{1}$$

where P is Ertel PV, v is northward velocity, the notation $\bar{v}^*$ denotes the mass-weighted zonal mean of v along isentropic surfaces, and $\hat{v} = v - \bar{v}^*$ the departure from that mean. Eq. (1) simply states the need for zero net flux of PV in a conservative steady mean state: mean and eddy fluxes must cancel. Therefore, a nonzero eddy flux of PV—which will be present whenever waves are breaking, unless there is no background PV gradient—requires a nonzero mean circulation in steady state. In fact, in the usual case of northward mean PV gradient and downgradient eddy flux, $\overline{\hat{v}\hat{P}}^* < 0$, and (1) implies a poleward circulation (since the sign of $\bar{P}^*$ has the same sign as latitude, except perhaps very close to the equator). Thus, the wave drag drives the flow poleward, through a mechanism we will here refer to as the "Rossby wave pump"[1], providing an explanation for the poleward flow in the winter stratosphere (for a thorough discussion, see Haynes et al., 1991; Holton et al., 1995). There are outstanding issues, however, in our understanding of the latitudinal extent of the circulation. In the tropics, the observed circulation clearly extends far beyond the surf zone, deep into the tropics and even across the equator: issues involved in explaining this behavior are discussed in Plumb and Eluszkiewicz (1999). At least a component of the tropical circulation may be a nonlinear, thermally driven circulation, analogous to the tropospheric Hadley cell (Dunkerton, 1989; Semeniuk and Shepherd, 2002a,b). The component driven by wave drag may also extend into the tropics, either via a similar nonlinear mechanism or through the effects of other wave motions within the tropics (Plumb and Eluszkiewicz, 1999; Scott, 2002). At the vortex edge, theoretical arguments lead us to expect little penetration of the steady circulation into the vortex, even under nonadiabatic conditions (Bühler and Haynes, 1999; Sobel and Plumb, 1999), despite the observational evidence there of strong descent of tracer isopleths (such as in Fig. 1).

In the lower stratosphere, the effects of synoptic-scale tropospheric disturbances, present throughout the year, are probably responsible for driving the strong two-cell circulation there and in the upper troposphere, through the same Rossby wave pumping mechanism (the local dynamics of these waves, away from their baroclinic sources, being just Rossby wave dynamics). Near the surface, these same baroclinic disturbances drive an equatorward return flow (Held and Schneider, 2000; Koh, 2001) in the form of cold air outbreaks.

The mesosphere is dominated by a strong global circulation (with $\bar{v}^*$ of several ms^{-1}) from summer pole to winter pole, which manifests itself in the dramatic reversal of pole-to-pole temperature gradient in the upper mesosphere. This circulation is believed to be driven primarily by upward propagating inertia-gravity waves. Unlike Rossby waves, inertia-gravity waves can propagate through both mean easterlies and westerlies, and their effects on the mean zonal flow can be of either sign, depending on their phase velocities. Because of selective dissipation of these waves in the stratospheric winds, it turns out that their effect on the mesosphere—through what we here call the "gravity wave pump"—is such as to drive the flow generally equatorward in summer and poleward in winter, as observed. While the mesosphere itself is not our prime concern here, this mechanism is strong enough to pump significant descent of mesospheric air deep into the stratosphere, as we shall see below.

Rapid stirring in the surf zone

Within the surf zone of the midlatitude winter stratosphere, tracers are subjected to both isentropic stirring and diabatic advection. If the eddy motions are almost adiabatic, then, for a tracer with mixing ratio χ,

$$\frac{\partial \bar{\chi}^*}{\partial t} + \bar{v}^* \frac{\partial \bar{\chi}^*}{\partial y} + \bar{\dot{\theta}}^* \frac{\partial \bar{\chi}^*}{\partial \theta} = -\frac{1}{\bar{\sigma}} \frac{\partial}{\partial y} \left(\bar{\sigma} \overline{\hat{v}\hat{\chi}}^* \right) \tag{2}$$

where θ is potential temperature and $\sigma = -g^{-1}\partial p/\partial\theta$ is the isentropic density[2]. Now, in (1), we saw that the

[1] We prefer to avoid the nomenclature "extratropical pump" proposed by Holton et al. (1985); as (1) implies, for a given PV flux, the mechanism becomes more efficient closer to the equator, where $\bar{P}^*$ is small.

[2] The reader may wonder (as a reviewer did) about the self-consistency of retaining diabatic terms in (2) while neglecting

dynamical balance is such that eddy and mean transport of PV are of the same magnitude: is the same true of tracers? Suppose that the eddy fluxes (of PV and tracers) are such as to destroy the gross mean gradient across the surf zone in a characteristic time scale τ, so that the eddy flux of PV is

$$\overline{\hat{v}\hat{P}}^* \sim \frac{\Delta P}{\tau} L \,,$$

where L is the surf zone width, and ΔP the gross mPV gradient across the surf zone. Then, from (1),

$$\overline{v}^* \sim \frac{\Delta P}{P} \frac{L}{\tau} \,,$$

where P is a typical value of PV in the surf zone. Therefore the time scale T for mean tracer advection horizontally across the surf zone, or vertically across a density scale height H, is

$$T \sim \frac{H}{\bar{w}^*} \sim \frac{L}{\bar{v}^*} \sim \tau \frac{P}{\Delta P} \,.$$

Hence the ratio of eddy to mean tracer transport time scales is

$$\frac{\tau}{T} \sim \frac{\Delta P}{P} \,.$$

Now, suppose the surf zone has width comparable with the Earth radius, but is weak enough that there is little tendency toward homogenization of PV within it; then $\Delta P \sim P$, and the two contributions to the tracer budget are comparable. However, the real stratospheric surf zone has weak PV gradients—a consequence of strong stirring (Juckes and McIntyre, 1987)—such that $\Delta P \ll P$, implying that isentropic stirring dominates the tracer budgets within the surf zone. This fact, which is simply a consequence of the decreasing efficiency of the Rossby wave pump under strong stirring, has important consequences for stratospheric transport.

3. Tracer-tracer relationships

Equilibrium slopes and compact relationships

The most immediate consequence of the dominance within the surf zone of isentropic stirring is that, for "long-lived" tracers for which the effects of local chemical sources and sinks, and of entrainment from outside the surf zone, are negligible on the time scale τ, horizontal mean tracer gradients are almost, but importantly not quite, eliminated. This implies that tracer isopleths (surfaces of constant mixing ratio) have shallow slopes, and in this regime the slope is determined by a balance between the flattening effect of isentropic stirring and mixing, and the steepening effect of diabatic advection (Plumb and Ko, 1992; Plumb, 1996, 2001; Sparling et al., 1997; Thuburn and McIntyre, 1997), implying that the isopleths are in the "slope equilibrium" regime described by Holton (1985) and Mahlman et al. (1985). In this regime, the shapes of the tracer isopleths are determined solely by this kinematic balance, and so the isopleths of all sufficiently long-lived tracers exhibit the same shape, in agreement with the observed characteristics discussed in Section 1. Moreover, Plumb and Ko (1992) argued that this in turn implies that a plot of one such tracer versus another would form a "compact" relationship, as illustrated in Fig. 3. This deduction remains true whether variations in mixing ratio are from temporal or spatial (vertical or horizontal) sampling, in agreement with observational findings (*e.g.*, Ehhalt, 1983; Fahey et al., 1990). From a theoretical viewpoint, the tracer-tracer relationship can never be precisely compact in the presence of mixing unless the relationship is linear (Plumb, 2001) but, in practice, scatter is often weak, unless (as we shall see) data are included from outside the surf zone.

The practice of presenting measurements of long-lived tracers by plotting against a reference tracer (frequently N_2O or CH_4) has become widespread. Amongst other things, the reproducibility of tracer-tracer plots makes for a more concise and useful form of data presentation than, say, time series or spatial profiles of tracer mixing ratios, which will usually differ from one profile to another simply because of air movement between samples. More importantly, the robustness of tracer-tracer relationships allows for the empirical determination of a "canonical" relationship for each long-lived tracer pair that can be used as a reference to isolate anomalous or otherwise noteworthy characteristics of tracer measurements. Interpretation of such relationships is not always easy, as most of our understanding of tracer characteristics is based on the spatial structure of transport, and hence of the tracer structures themselves. In order to utilize tracer-tracer plots to the full, there is a need to be able to think in tracer-tracer space. As yet, the body of theory that would allow us to do that is incomplete, but some important results are known, of which some will be discussed in the following.

them in (1). The neglected terms in the latter represent diabatic advection of momentum, which is small under reasonable assumptions (such as near-geostrophic conditions) because of the modest vertical gradient of the zonal flow. In the tracer budget (2), however, diabatic advection remains important, even when the diabatic flow is weak, since then the mean vertical gradient $\partial\bar{\chi}^*/\partial\theta$ becomes very large.

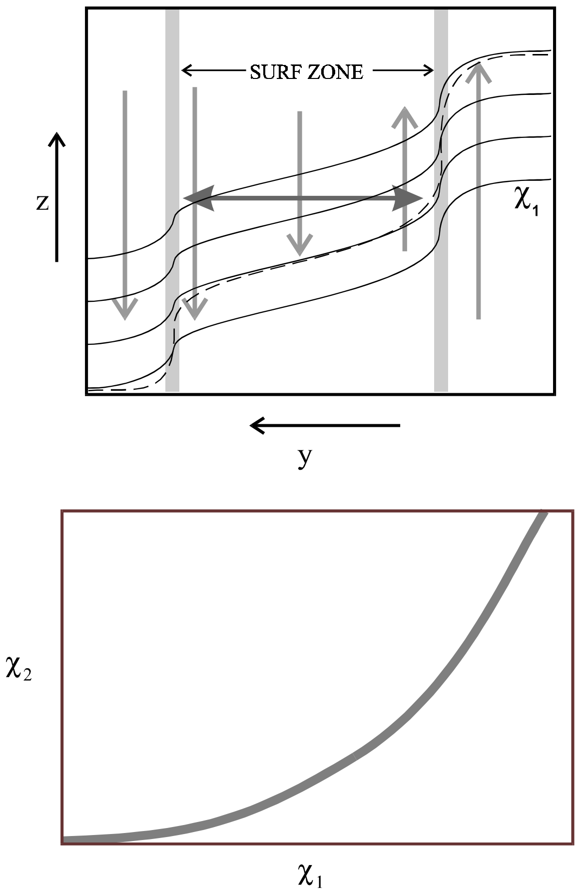

Figure 3. Illustrating compact relationships within the surf zone (schematic). The upper figure shows isopleths of mixing ratio χ_1 for one tracer in the winter hemisphere; the isopleth slope is weak within the surf zone (the middle region) but may be steep at the surf zone edges. The dashed contour is an isopleth mixing ratio χ_2 of a second tracer: within the surf zone, the isopleth is almost the same shape as one of χ_1. This implies that whenever χ_1 has a certain value within the surf zone, χ_2 always has a fixed value, so that a plot of χ_2 *vs.* χ_1 is compact, as shown in the lower curve.

Tropical isolation (and non-isolation)

The subtropical edge to the winter surf zone is evident, like the vortex edge, as a region of strong local gradient in tracer distributions, as seen in Fig. 1, for example, on the global scale. Its sharpness also shows up clearly in *in situ* aircraft data, as a near-discontinuity in local mixing ratios (*e.g.*, Murphy et al., 1993). In fact, as Fig. 1 illustrates, there are regions of strong subtropical gradient on both sides of the equator, which persist throughout the year. Fig 4 shows their seasonal evolution at several stratospheric altitudes, as determined from six years of HALOE observations of CH_4 (Neu, 2000). The edge locations are remarkably consistent from year to year. The "tropical region" (*i.e.*, the region bounded by the subtropical edges) migrates, most markedly at higher altitudes, into the summer hemisphere, qualitatively similar to the migration of maximum upwelling (Rosenlof, 1995; Eluszkiewicz et al., 1996). However, the location of the winter edge in fact corresponds more closely with the zero wind line than with the transition from upwelling to downwelling (Neu, 2000; Neu et al., 2002). This is consistent with calculations of "effective diffusivity" (Nakamura, 1996) which show high values within the surf zone, decreasing sharply at the vortex and subtropical edges, with

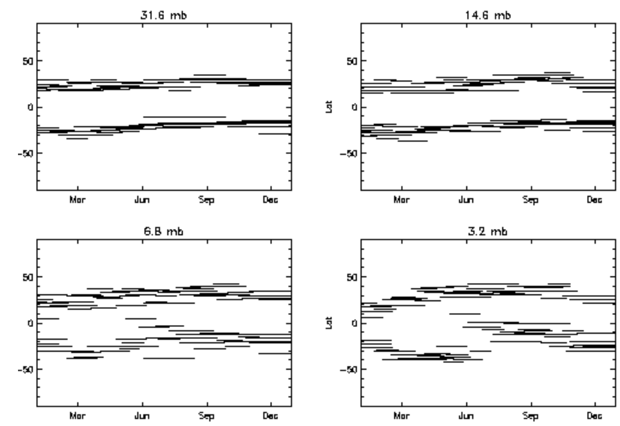

Figure 4. Seasonal evolution of the subtropical edges, as determined from HALOE observations of CH_4 during the period 1993 through 1998. Each line shows the location of the edge through a HALOE observing period. (After Neu, 2000.)

much smaller values throughout the tropical easterlies (Haynes and Shuckburgh, 2000; Allen and Nakamura, 2001). Taken together with the observed equatorward migration of the subtropical edge in early winter, such evidence suggests that the wintertime subtropical edge, like the vortex edge, is produced and sharpened by entrainment of edge air into the surf zone, associated with surf zone stirring. On the summer side of the equator, where such processes are at best weak (except at low altitudes), the situation is not so clear. However, it appears that the summer subtropical edge is not merely a survivor of wintertime formation, but is actively sustained by diabatic processes, especially at higher altitudes (Neu, 2000).

Evidence that the subtropical edges act in some sense as transport barriers, at least partly isolating the tropics from the vigorous stirring of the surf zone, comes from the observation that tracer-tracer relationships in the tropics are distinct from those of the surf zone (*e.g.*, Plumb, 1996; Volk et al, 1996). Among several other lines of evidence (such as the year-long persistence of successive phases of the quasi-biennial oscillation), perhaps the most visual is the now-famous "tape recorder" signature in the tropical lower stratosphere (Mote et al., 1996): the annual cycle of temperature at the tropical tropopause imprints a similar cycle on water vapor which, in turn, is converted by mean upwelling into a cyclic vertical structure. About one complete wavelength is visible: since there is little annual cycle in extratropical water vapor, this implies that the time scale for injection of extratropical air into the tropics is of the order of a year. While this is long enough to ensure that the tropical region is distinct from the surf zone, with its much shorter mixing time scales, it is not so short as to be unimportant in tracer budgets within the tropics, where the competing transport process (mean vertical advection) is very slow (tropical upwelling velocities in the lower stratosphere are typically around 1 km per month). Thus, it is not accurate to regard the tropical region as being completely isolated. Indeed, many studies have deduced similar time scales (most in the range 1 - 1.5 yr) for detrainment of surf zone air into the tropical lower stratosphere, on the basis of the observed budgets of chemical tracers (*e.g.,* Avallone and Prather, 1996; Volk et al., 1996; Hermann et al., 1998) or of age (Hall and Waugh, 1997; Neu and Plumb, 1999). Indeed, Hall and Waugh (1997) used simultaneous analysis of observations of age and of the tape recorder to make independent determinations of all components of transport in the region, which amongst other things allowed them to confirm that the effects of vertical diffusion are negligible (as had previously been assumed).

The formation of tracer relationships in the surf zone

As a first step toward understanding what makes surf zone tracer relationships they way they are, consider a simple, hypothetical stratosphere comprising two regions: a tropical region in which there is upwelling, and a surf zone with rapid isentropic stirring and net mean downwelling, as illustrated in Fig. 5. (This case is considered more explicitly in Plumb, 2002.) The net mass flux associated with the upwelling and downwelling decreases with height, so there must be entrainment of air into the surf zone across the subtropical edge separating the two regions. As we have seen, there may also be de-

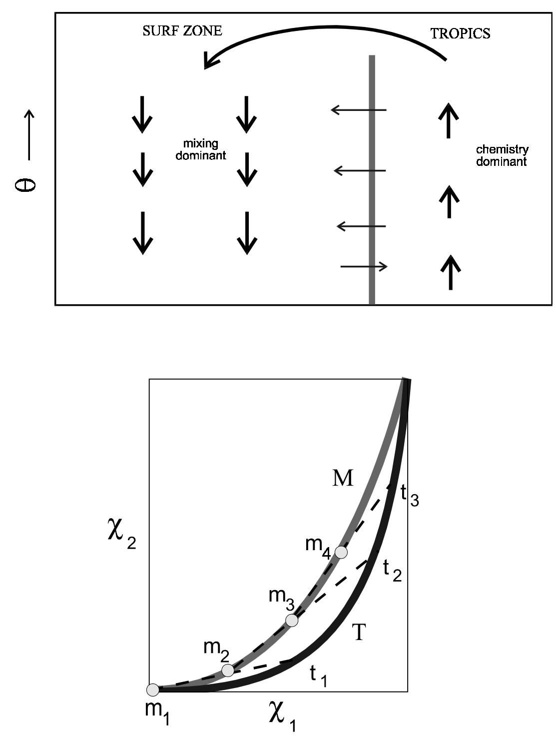

Figure 5. Schematic showing the deveopment of tracer-tracer relationships in the stratospheric "surf zone". Upper figure shows a schematic latitude/potential temperature cross section, with upwelling in the tropics and downwelling in the surf zone. Lower figure shows the relationship between the mixing ratios of two tracers in the tropics (curve "T") and in the surf zone (curve "S"). See text for discussion and for explanation of other symbols. (After Plumb, 2002.)

trainment into the tropics; we will consider its influence later.

Now suppose, also for simplicity, that photochemical sources and sinks are confined to the tropics; the tracers of interest are conserved within the surf zone. To be specific, we consider two tracers with tropospheric sources, and whose tropospheric mixing ratios can be considered fixed, with stratospheric sinks. Tracer 1 is supposed to have a longer lifetime than tracer 2 (so the pair could be, *e.g.*, N_2O and CFC-11). Within the tropics, their mixing ratios χ_1 and χ_2 decrease with height, χ_2 the faster, as it has the shorter lifetime. Therefore the $\chi_2 : \chi_1$ relationship will look something like that shown in the curve marked "T" on Fig 5; moving up in altitude corresponds to sliding down the curve T from upper right to lower left. In the simplest case, this curve will be purely photochemically controlled. In general, however, the curve may not be compact—we know little about mixing processes within the tropics—but that does not matter greatly for our present arguments. At sufficiently high altitudes the mixing ratios of both tracers will be close to zero, corresponding to the point labeled "m_1" on the curve. Let us now follow this air into the surf zone, where it will begin to descend to some lower altitude. In the absence of any local entrainment of air from the tropics, the mixing ratios will be preserved and so, in tracer-tracer space, the air is still at point m_1. However, at the same potential temperature, the tropical mixing ratios are located at point t_1, where both are greater than at m_1. If there is now entrainment of tropical air, which is then assimilated into the surf zone at the same θ, the resulting air mass will now lie at point m_2, somewhere along the line joining m_1 and t_1 (Waugh et al. (1997)), exactly where depending on the relative masses of air being mixed together. Subsequently, as shown on Fig. 5, one can imagine a sequence of similar events of descent followed by entrainment and mixing, such that m_2 mixes with t_2, to produce m_3, which mixes with t_3 to produce m_4, and so on. If we imagine this sequence of discrete events being replaced by a continuous interplay of descent and mixing, the resulting surf zone tracer-tracer relationship will be the curve "M" on Fig. 5, which is distinct from the photochemically-determined tropical relationship. Note that the surf zone relationship lies on the concave side of the tropical one; we can now see that this simply results from the fact that surf zone air at any altitude is a mixture of tropical air from all higher altitudes. Note also that the separation of the two relationships depends on nonlinearity of the tropical curve: if it were linear, a mixture would always lie on the same line, and M would overlie T.

If the tracer-tracer curve within the tropics were not compact (but still exhibited the general tendency of curve T), one would expect little difference in the predicted curve M. Surf zone mixing would still guarantee the compactness of M, and the precise shape of M would depend on the particular mix of tropical air that is entrained into it.

A complete separation between M and T, manifested by the tracer-tracer scatterplot comprising these two curves only, presumes that assimilation of tropical air into the surf zone occurs completely and instantaneously. During the time mixing actually occurs, however, intermediate mixing ratios will be formed and so, at any instant in time, or averaged over time, the scatterplot will include a smaller population of air intermediate between M and T (including discrete mixing lines if individual entrainment/mixing events are captured). (This is discussed in more detail in Plumb, 2002.)

We saw earlier that there is evidence for a significant amount of detrainment of surf zone air into the tropics. Thus the assumption we made earlier in this Section, that the tropical tracer-tracer curve is purely photochemically determined, is likely to be invalid. Inclusion of surf zone air is likely to reduce the gross curvature of T. The nature of the tracer-tracer relationships in this case can be revealed by iterating the above arguments, allowing for modification of the tropical tracer profiles, but the end result is qualitatively unchanged: the surf zone curve lies on the concave side of the tropical curve, because the central fact that surf zone air comprises a mixture of tropical air is unaffected by the existence of detrainment into the tropics.

Tracers in the vortex

It is clear from evidence such as that shown in Fig. 1 that tracer isopleths are depressed substantially within the polar vortex. While this is frequently interpreted as being indicative of greater mean diabatic descent within the vortex than outside, this is not necessarily so. Determinations of mean residual or diabatic velocity (Rosenfield et al., 1994; Rosenlof, 1995; Eluszkiewicz et al., 1996) frequently show broad regions of descent in midwinter throughout high and middle latitudes, not necessarily concentrated within the vortex. Rather, the difference lies in the differing climatologies of stirring in the vortex and surf zone. Within the surf zone, rapid stirring ensures that a typical trace particle, while it may be subjected to strong diabatic cooling when it is near the vortex edge, does not remain there for long enough to undergo a substantial downward displacement. Over seasonal time scales, as noted earlier, air is stirred throughout the surf zone and thus responds to the net diabatic descent (averaged over the surf zone), rather than to the much stronger local descent in high latitudes. By contrast, vortex air remains within the vortex for long periods and is thus subjected to sustained diabatic cooling. Thus, vortex air is characterized by "unmixed descent", in the words of Russell et al. (1993b), although, as we shall see below, this should be qualified to "relatively unmixed descent". All this is just another way of saying that the strong tracer gradients at the vortex edge are produced by differential isentropic stirring, rather than by differential diabatic descent.

That said, there is some degree of mystery as to the dynamical origins of the high latitude descent. There are theoretical grounds to expect Rossby wave pumping in the surf zone to produce most descent at the vortex edge, rather than deep within, and the sustained vortex descent through the winter, evident in calculations based on radiation codes and in tracer measurements, is difficult to explain on the basis of the transient circulation produced by fall cooling and the concomitant setting up of the polar night jet. Rather, there is growing evidence of a substantial contribution to vortex descent, all the way down to the lower stratosphere, from the "gravity wave pump" of the mesosphere. Estimates of the mean poleward velocities within the mesosphere are sufficient to flush the entire mesosphere over the course of a single winter season, much of that air reaching the lower stratosphere by late winter. Direct evidence of this degree of descent has come from observations made during the SOLVE campaign; in March 2000, markers of mesospheric air (anomalous apparent SF_6-based "age" (Moore et al., 2001), and CO, of upper mesospheric origin) were observed and modeled in the lower stratospheric vortex (Plumb et al., 2002).

Just as we have discovered for the tropics, it is now recognized that the vortex edge, though clearly impermeable to a first approximation, is not a perfect barrier to transport. The most compelling evidence of this comes from observations of anomalous vortex relationships between certain pairs of long-lived tracers (Michelson et al., 1998; Kondo, 1999; Plumb et al., 2000; Rex et al, 2000), whose only apparent explanation is that surf zone air is being mixed into the vortex. Indeed, it now appears that distinct compact relationships were established within the Arctic vortex by late winter of 2000 (Ray et al., 2001), implying also the presence of rapid mixing within (in addition to possible weak mixing into) the vortex. In some ways, the development of compact vortex relationships, through the mixing of surf zone air, parallels the discussion of Section 6, and we might replace Fig. 5 with Fig. 6, in which we add a vortex to the system, and in which

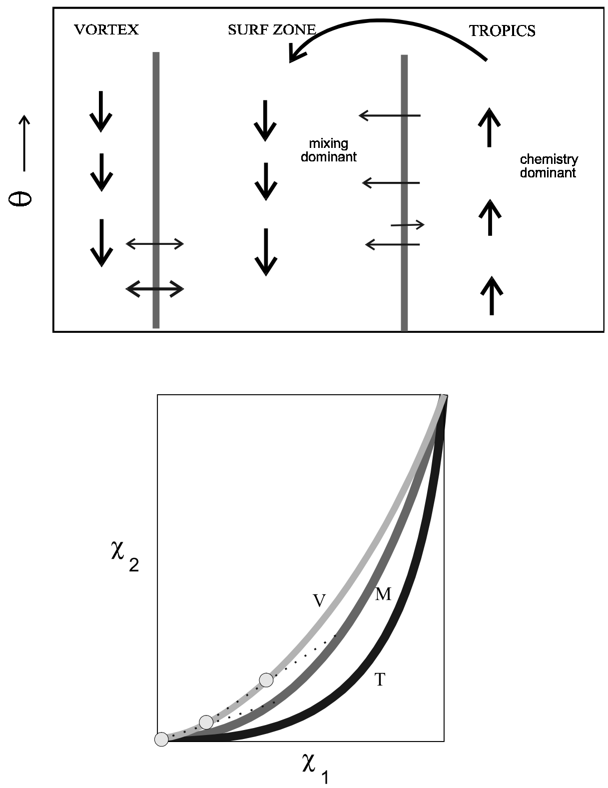

Figure 6. As Figure 5, but adding the polar vortex. Upper figure is a schematic showing a latitude/potential temperature depiction of transport in the three regions. The lower figure shows the relationship between tracer 1 and 2, the curves "T", "M", and "V" representing the relationships in the tropics, surf zone, and vortex, respectively. (After Plumb, 2002).

a compact relationship is established within the vortex through vortex descent and assimilation of surf zone air, in just the same way as we discussed in Section 6 for development of the surf zone relationship. One key difference, however, is that the vortex edge is a transient phenomenon, and it is not clear to what extent the surf zone air that is assimilated into the vortex relationship gets there by being transported across the vortex edge during winter, rather than simply being caught up in the vortex at the time of its formation in fall (*cf.*, the remarks of Ray et al., 2001).

Such development of distinct vortex relationships undermines attempts to use anomalous tracer-tracer relationships as a means of quantifying chemical losses of ozone (*e.g.,* Proffitt et al., 1990; Müller et al., 1996) or sedimentation of NO_y (*e.g.* Fahey et al., 1990). Use of mixing line arguments (Rex et al., 2000) to remove transport effects are unlikely to be reliable in general, since it is necessary to assume a single mixing event (Plumb et al., 2000) and the production of a compact vortex relationship requires many such events. One promising approach, proposed by Esler and Waugh (2001), is to use a linear combination of independent tracers that exhibit a near-linear canonical relationship with a reference tracer; since this linear relationship is unaffected by mixing, anomalies can arise only though anomalous sources and sinks.

4. Quantification of transport: age

Definition of age

All long-lived tracers carry information about transport in their distributions. As we have seen, their surfaces of constant mixing ratio are determined entirely by a balance between competing effects of transport. However, their gradients normal to such surfaces are dependent on chemical sources and sinks, and so they cannot be used to make unambiguous deductions about transport. It is difficult, for example, to determine whether errors in model simulations of tracer distributions result from errors in transport or in the specification of sources and sinks. Identification of transport effects are more easily made on the basis of the distributions of temporal tracers, those which have known, time-dependent, mixing ratio $\chi^T(t)$ in the troposphere, and negligible internal stratospheric sinks (or sources). A special case of such tracers (or, rather, a derivative of them) is "age". (See Waugh and Hall (2002) for an up-to-date comprehensive review of the theory and applications of the concept of "age" in the stratosphere.) In it simplest form, one might be tempted to define age $\Gamma(\mathbf{x})$ at location $\mathbf{x}$ as the time lag between local and tropospheric concentrations, *i.e.*,

$$\chi(\mathbf{x}, t) = \chi^T[t - \Gamma(\mathbf{x})] \ . \qquad (3)$$

However, because of the dominance of stirring and mixing in stratospheric transport, one must think of age not as a simple time lag, but as a statistical distribution of lag times (the "age spectrum"), as emphasized by Hall and Plumb (1994). Eq. (3) is valid, for linearly growing tracers only, if Γ is equated with the "mean age"—the first moment of the age spectrum. Fortunately, we have at least two stratospheric tracers, SF_6 and CO_2, which approximately meet the criteria of negligible sinks and of linear growth, though neither does so exactly: in the lower stratosphere, the secular growth of CO_2 is contaminated by the annual cycle transported from the troposphere (Boering et al., 1996), and SF_6 has a mesospheric sink, which corrupts determinations of age in air (such as air in the polar vortices, as we noted in Section 7) that has been in the mesosphere (Hall and Waugh, 1998; Moore et al., 2001). Nevertheless, ages can be determined from SF_6 in "young" lower stratospheric air (where CO_2 is influenced by the annual cycle), from CO_2 in "old" air within the polar vortices (where SF_6 is influenced by its mesospheric sink), and from either in the considerable region of overlap where both species are good tracers of age [see Waugh and Hall (2002) and references therein]. Thus, between the two species, we can obtain reliable determinations of mean age from observations throughout the stratosphere.

Modeling of stratospheric mean age

Observations of age provide us with a reference against which the transport characteristics of stratospheric chemical transport models (CTMs) can be assessed. In the intercomparison project "Models and Measurements II" (Park et al., 1999), many CTMs (both 2-D and 3-D) were used to simulate age. The results of this exercise are discussed in detail in Hall et al. (1999). Nearly all of the models seriously underestimated age throughout the stratosphere, including in the tropical lower stratosphere where the air has only recently entered from the troposphere. Moreover, as a demonstration of the relevance of the simulation of age to the proper simulation of stratospheric chemistry, the modeled abundances of real tracers, including the crucial chemical families of total inorganic chlorine and total reactive nitrogen, were shown to correlate strongly with age simulations. It appears that the current generation of stratospheric CTMs exaggerate stratospheric transport, which raises concerns about their ability to generate an accurate simulation of stratospheric chemistry. Recent experiments (Mahowald et al., 2002; W. A. Norton, private communication) have shown that use of isentropic coordinates greatly improves the simulation of stratospheric age (and therefore of transport). Use of these coordinates guarantees the crucial connection between diabatic processes and vertical transport. Mahowald et al. (2002) showed such improvement, using the same input winds to off-line pressure-coordinate and isentropic-coordinate models, and ascribed much of the improvement to a better simulation of slow vertical transport in the equatorial lower stratosphere.

Flux of age

Age is itself a tracer; if we write the general tracer transport equation as

$$\rho^{-1}\left[\frac{\partial}{\partial t}(\rho\chi) + \nabla \cdot \mathbf{F}_\chi\right] = S \ ,$$

where ρ is air density and $\mathbf{F}_\chi$ the advective-diffusive flux of χ, then given (3) for a linearly growing tracer in steady transport, it is easy to show that Γ satisfies the equation

$$\rho^{-1}\nabla \cdot \mathbf{F}_\Gamma = 1 \ , \tag{4}$$

subject to the tropospheric boundary condition $\Gamma^T = 0$. Thus, age is the steady state solution to a tracer budget with unit source (air ages by one year per year). Waugh and Hall (2002) call age defined by (4) "ideal age". Therefore, age can be treated like other long-lived tracers (provided it is borne in mind that age has a source everywhere) and, for example, it exhibits compact relationships with other tracers such as N_2O (Boering et al., 1994, 1996).

One consequence of (4) that proves to be useful is that the flux of age through any surface can be easily determined (Neu and Plumb, 1999). If we integrate (4) with respect to mass over the entire atmospheric region R above a global surface S, then

$$\int_S \mathbf{F}_\Gamma.\mathbf{n}\ dA = -\int_R dm = -M_R, \tag{5}$$

where $\mathbf{n}$ is the upward unit normal at S, and M_R is just the mass of the atmosphere in R: the net downward flux of age through S simply equals the air mass above S. If we take S to be the tropopause, the flux of age into the troposphere equals the mass of the stratosphere, which is of course known (to the accuracy with which we know the mean pressure on the tropopause).

5. Stratosphere-troposphere exchange

Net tracer fluxes

On the basis of an analysis in which a single surf zone was assumed to encompass the entire globe, Plumb and Ko (1992) argued that the net global fluxes of two long-lived tracers across a surface of constant mixing ratio are in a ratio equal to the slope of the compact curve relating the two tracers. This result follows directly from the fact that the net flux through such a surface is, under assumptions of rapid mixing, diffusive, so that the flux of each species $F_n \propto -\partial\chi_n/\partial\theta$. Since $\chi_2 = \chi_2(\chi_1)$, it then follows that

$$\frac{F_2}{F_1} = \frac{d\chi_2}{d\chi_1} \ . \tag{6}$$

One can, alternatively, frame (6) as a ratio of atmospheric lifetimes. Plumb (1996) showed that this result remains valid for fluxes across the tropopause even when allowance is made for different tracer-tracer relationships between surf zone and tropics, provided the surf zone relationship is used to determine the slope. We can understand this result using a simpler, but more general, argument which also illustrates some possible limitations. Consider Fig. 7. We separate exchange of

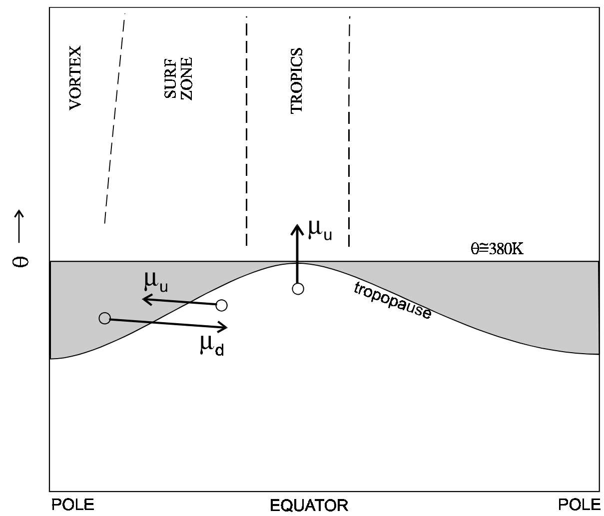

Figure 7. Schematic showing transport across the tropopause. See text for discussion.

air across the tropopause into upward events, with net mass flux μ_u, and downward events, with net mass flux μ_d. Adopting the tropopause as the reference surface allows a powerful simplification, namely that for tracers that are long-lived in the troposphere as well as in the stratosphere, tropospheric gradients are very weak and so it is a good approximation to assume that the tropospheric mixing ratio, χ^T, is uniform. Then the net upward tracer flux is just

$$F = \mu_u\chi^T - \mu_d\chi^S \ ,$$

where χ^S is the mass-weighted average mixing ratio of air entering the troposphere. Note that χ^S may be quite different from χ^T in vigorous cross-tropopause stirring events in which air is taken from deep in the stratosphere. However, since such events are likely to be largely adiabatic, the air will come from within the "lowermost stratosphere"—the shaded region on Fig. 7—lying below the isentrope (about 380K) that skims the highest point on the tropopause. Now, if we average over a year and neglect interannual variability, the net flux of mass into the stratospheric must vanish, so $\mu_u = \mu_d$. Then

$$F = \mu_d \left(\chi^T - \chi^S \right) \ ,$$

from which we obtain, for two species, the flux ratio

$$\frac{F_2}{F_1} = \frac{\chi_2^T - \chi_2^S}{\chi_1^T - \chi_1^S} \ . \tag{7}$$

This relation is obviously similar to (6), but the local slope in the latter is replaced by a ratio of finite differences. In fact, the mutual relationship between pairs of tracers that have no local sources or sinks within the lowermost stratosphere should be linear there (Plumb, 2002), as observations seem to confirm; for such pairs of tracers, we recover (6). We can also see why it is the surf zone relationship that is relevant here: it is the region from which air entering the troposphere originates that matters. Air cannot cross the tropopause adiabatically from the tropical stratosphere, even if it enters the tropical upper troposphere, nor from the polar vortex, which disappears as a distinct entity below about 400K (McIntyre 1995). Air does, of course, enter the tropopause in the summer hemisphere: it is assumed here that the surf zone relationships established in the previous winter are preserved in the midlatitude summertime lower stratosphere, as observations appear to suggest.

The result (6) has been exploited to determine net ozone fluxes through the tropopause (Murphy and Fahey, 1994) and to calculate lifetimes of a range of species (Volk et al., 1997). In order to utilize the result to determine the flux or lifetime of a particular species, it is necessary to know that of a second species. However, as we saw in Section 8, we do know the flux of age, at least to the accuracy with which we know the mass of the stratosphere, and this can be used as the reference tracer, as done by Volk et al (1997). (They actually used the budget of SF_6, rather than of age itself, but the approach is equivalent to using age directly.) However, unlike many tracers, age does have an effective source everywhere, including the lowermost stratosphere, and so its relationship with other long-lived tracers will not usually be linear, as Volk et al. found, a fact that presents some difficulties in applying (7) directly.

Dehydration and transport through the tropical tropopause

Stratospheric dryness was explained in the classic study of Brewer (1949) as a consequence of the passage of air through the cold tropical tropopause. While important details, such as the question of whether the large-scale tropical tropopause temperatures are quite cold enough to explain the observed dryness, remain a matter of active debate, the key element of Brewer's suggestion—the "freeze drying" of air as it passes through the tropical "cold trap"—remains at the center of conventional wisdom. The details of the drying mechanisms lie beyond the scope of this review; however, we take the opportunity to note something that until very recently appears to have been overlooked, namely the likely partial analogy between transport through the tropical tropopause and in the winter surf zone. Mean upwelling, as noted above, is no more than about 1 km per month in the tropopause region and so vertical advection is much slower than probable horizontal (isentropic) transport. Thus, just as in the surf zone, a typical trace particle will act as an integrator of diabatic motion across a wide horizontal domain, responding to the mean upwelling across the domain rather than to local values. Thus, for example, the mean motion of Lagrangian tracers may be even slower than 1 km per month. These considerations are likely to be important, given that the cold trap is geographically local, both in latitude and longitude. Moreover, such a scenario implies that all air will sample a wide horizontal domain during ascent, and may pass through the cold trap even if local diabatic motion is downward there. (The same point has been made by Holton and Gettleman, 2001.)

To illustrate this consider the following simple illustration. We assume a two-dimensional (y, z) system (so that y represents either the zonal or latitudinal direction, or both) of width L, in which the temperature $T(y, z)$ is specified to be 220K everywhere, except in a region $-L_{CT}/2 < y < L_{CT}/2$, and $-H_{CT}/2 < z < H_{CT}/2$ within which

$$T(y, z) = 220 - 30 \left[1 - \left(\frac{2y}{L_{CT}} \right)^2 \right] \sin \left[\frac{\pi}{H_{CT}} \left(z + \frac{1}{2} H_{CT} \right) \right] \ , \tag{8}$$

so the minimum temperature at the center of the "cold trap" is 190K. Air moves vertically through this entire region with specified velocity

$$w(y) = W_0 \left[1 + \alpha \cos \left(2\pi \frac{y}{L} \right) \right] \tag{9}$$

where $W_0 > 0$—mean motion is upward—but the motion is not upward everywhere if $|\alpha| > 1$. In y, horizontal stirring is represented by having air parcels perform a random walk $y(t) \rightarrow y(t + \delta t)$ at each time step δt, where

$$y(t + \delta t) = y(t) + \rho \sqrt{2K \delta t} \ , \tag{10}$$

where ρ is a random number between 0 and 1, and K is the diffusivity corresponding to the random walk. The key parameters of the problem are $\beta = L_{CT}/L$, the fractional area occupied by the cold trap, and $\omega = W_0 L^2/(KH_{CT})$, the ratio of horizontal dispersion time (across the entire region) to vertical advection time (through the cold trap).

We consider a case with $\alpha = -2$ and $\beta = 0.2$; in this case local vertical velocities are downward everywhere within the cold trap, as shown in Fig. 8. Trace particles were initialized every time step at a random horizontal position, a distance H_{CT} below the bottom of the cold trap, and the minimum temperature, T_{min}, they experience during upwelling was recorded. Eventually, the probability distribution of T_{min} in air that has moved to altitudes above the cold trap reaches equilibrium: these equilibrium values of T_{min} are shown in Fig. 9, for different values of ω. Even though the cold trap occupies only 20 percent of the horizontal area, for $\omega < 0.4$ all air parcels pass through the cold trap, and for $\omega < 0.1$ more than 90 percent of parcels experience temperatures within 1K of the minimum temperature. As expected, therefore, in the limit of small ω (rapid horizontal stirring) parcels sample the entire horizontal domain during their mean ascent. Only when $\omega \geq 0.5$ does a significant fraction of particles avoid passing through the cold trap, even though all upwelling takes place outside the cold trap.

Of course, this calculation is highly simplified and, amongst many other things, it is not clear what constitutes the "entire horizontal domain" in practice for air upwelling through the tropical tropopause and we have paid no attention to the microphysical details of the relationship between parcel temperature histories and dehydration. The point being made is that vertical transport, in the tropical tropopause region as in the wintertime surf zone, is horizontally nonlocal, and that it is important not to be misled by the "one-dimensional thinking" into which discussions of troposphere-stratosphere exchange have sometimes lapsed.

6. Final remarks

Many stones have been left unturned in this brief overview, ranging from interannual variability and the impact of the quasi-biennial circulation in low latitude tracer structures to the small-scale mechanisms that allow different air masses, stirred together by the large-scale flow, finally to mix. The summer hemisphere has been largely ignored; while this reflects a long-standing

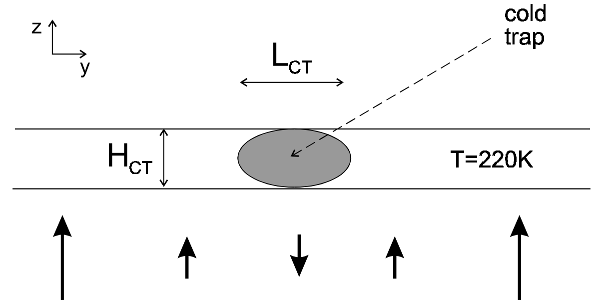

Figure 8. Schematic of the cold trap calculation. See text for discussion.

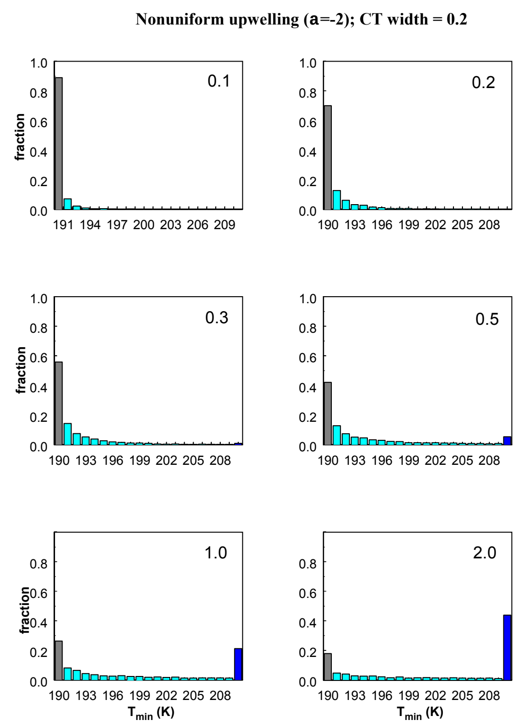

Figure 9. $P(T_{min})$, the equilibrium fraction of parcels that have experienced a minimum temperature of T_{min} (in 1K bins, except that the last bin contains all $T_{min} >$ 210K) during passage through the altitudes of the cold trap. Each frame is labeled at its top right with the value of ω.

tendency to regard the summer stratosphere as dynamically dull, the behavior of tracer structures through the summer is something that must be understood if the full seasonal picture is to be clarified. We also need to understand, and to correct, the shortcomings of stratospheric CTMs: are the problems numerical, or is there something missing in their representation of transport? Along with many other issues, these stand as challenges for the future.

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