Chapter 2

The Influence of Dynamical Processes on Ozone Abundance

2.1 Introduction

Ozone abundance is affected by dynamical processes in two important ways: first, through temperature and, second, through transport and mixing. Although the two aspects are related in practice – temperature is affected by the same dynamical processes that control transport and mixing – it is useful to maintain a distinction between them. Accordingly, the first section of this chapter addresses our present understanding of the dynamical structure of the stratosphere, while the second section addresses the global structure of transport and mixing. Subsequent sections focus on specific regions of the stratosphere, in terms of their role in determining ozone abundance.

2.2 The Dynamical Structure of the Stratosphere

The stratosphere is the region of the atmosphere between approximately 10 km and 50 km altitude, containing roughly 20% of the mass of the atmosphere. It is distinguished, as its name suggests, by a strong vertical stratification, or layering, that inhibits vertical motion. The existence of the stratosphere is a consequence of the ozone layer. Although the strongest heating of the atmosphere occurs from the earth's surface, there is a secondary heating maximum throughout the ozone layer. Rapid vertical heat transfer by convection in the lowest part of the atmosphere, referred to as convective adjustment, leads to a layer of weak stratification (the troposphere), capped by a layer of strong stratification (the stratosphere). The boundary between the two is the tropopause. The global mean height of the tropopause is thus determined by a balance between the amount of stratospheric ozone, the surface temperature, and the vertical temperature structure of the troposphere (which involves the distribution of water vapour and clouds). It follows that changes in any of these quantities can be expected to lead to changes in the global mean height of the tropopause.

The height of the tropopause varies significantly with latitude. It is greatest in the tropics, where it is typically around 18 km, and decreases to around 8 km as one moves toward the poles. Part of this variation can again be explained on radiative-convective grounds: since surface heating is strongest in the tropics, the depth of convective adjustment is greater there. However, there are two other contributing effects that act in the same direction. First, there is a persistent mean meridional mass circulation in the stratosphere that draws air upward in the tropics and pushes it downward in the extratropics. Second, although the photochemical production of ozone takes place primarily in the tropics, significant quantities of ozone are transported poleward, thus affecting the latitudinal distribution of radiative heating. Both effects – which turn out to be closely related (see Section 2.3) – act to raise the tropopause in the tropics and to lower it in the extratropics. In addition, there is a sharpening of the tropopause slope in midlatitudes associated with the subtropical jet stream, which is controlled by tropospheric synoptic-scale weather systems. Thus, the latitudinal structure of the tropopause reflects a complex interplay between the mass circulation of the stratosphere, the ozone distribution, upper-tropospheric synoptic-scale disturbances, and the latitudinal temperature gradient of the lower troposphere. Once again, water vapour and clouds play a critical role in the latter. Our understanding of how these different processes interact to determine the structure of the tropopause is qualitative at best.

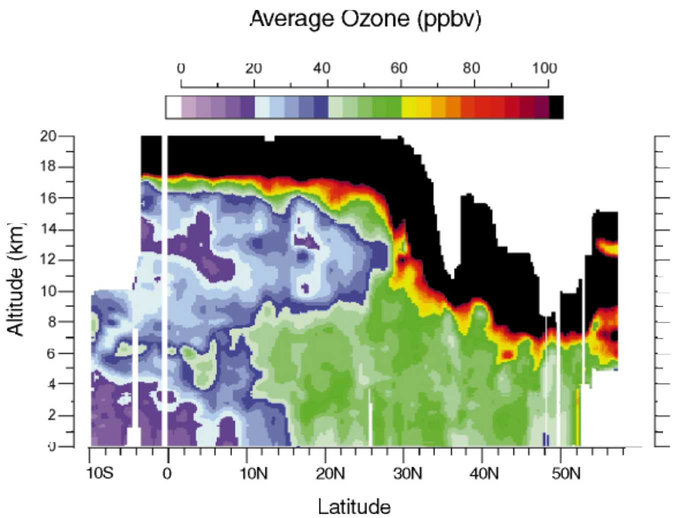


Figure 2.1 Composite latitudinal ozone distribution obtained with an airborne lidar over the western Pacific during the NASA Pacific Exploratory Mission conducted in February-March 1994 (private communication, E.V. Browell, NASA Langley Research Center, Hampton, Virginia, U.S.A.). The strong gradient region (yellow to black) follows the tropopause.

There is a very direct connection between the spatial distribution of ozone and the spatial structure of the tropopause. Figure 2.1 shows a cross-section of ozone taken from airborne lidar measurements. Comparatively high values of ozone are found in the stratosphere, and comparatively low values in the troposphere. On short (i.e., several day) timescales, the tropopause acts as a deformable material surface. When the tropopause descends, the fraction of stratospheric air above that location increases, thereby increasing total ozone; when the tropopause ascends, there is a corresponding decrease in total ozone. This correlation is well established on short timescales associated with dynamical variability in the tropopause height. On longer timescales, other effects come into play. Nevertheless, this highlights the fact that the structure of the tropopause has a significant impact on ozone distribution and must be accounted for in any assessment of ozone changes.

The elevated tropical tropopause implies that a local minimum in lower stratosphere temperatures will occur in the tropics – the reverse of the situation in the troposphere. This feature is present in all seasons and is associated (via thermal-wind balance) with the subtropical jet stream. At higher altitudes, the latitudinal temperature structure has a dramatic seasonal dependence. Ozone that has been transported poleward provides a major source of radiative heating in the sunlit polar summer, which maintains the relative warmth of polar temperatures relative to tropical values, and is reflected in the easterly zonal winds of the summer hemisphere stratosphere. In the winter hemisphere, in contrast, the lack of radiative heating over the polar night leads to extremely cold temperatures and the formation of an intense westerly polar vortex. Thus, in polar regions, the seasonal cycle of temperature consists of a cooling trend in the fall and a warming trend in the spring.

The cold temperatures of the polar night are mitigated by adiabatic heating arising from the compression of the descending, poleward-moving air. However, there are important contrasts between the Antarctic and Arctic wintertime regimes. In the Antarctic, the wintertime mass circulation is comparatively weak and fairly steady, with little interannual variability, although the effect of the quasi-biennial oscillation (QBO) is discernible. In the Arctic, in contrast, the wintertime mass circulation is much stronger, and far more variable; this leads to warmer temperatures on average than those found in the Antarctic winter, with episodic intense warming events known as stratospheric sudden warmings. These characteristics are demonstrated in Figure 2.2, which shows time series of mid-stratospheric polar temperatures over the Antarctic and Arctic from observations and from the Canadian Middle Atmosphere Model [Shepherd 1995; Beagley et al. 1997].

An understanding of this behaviour requires consideration of what determines the mean meridional mass circulation. To a first approximation, the adiabatic heating (or cooling) associated with sinking (or rising) air is balanced by radiative cooling (or heating). Although one might be tempted to conclude that the radiative cooling or heating is the *cause* of the vertical motion (e.g., air rises where radiation is warming it), it is now well established that, on a global scale and over seasonal timescales, it is rather the opposite: the observed local radiative imbalances are themselves caused by the mean meridional mass circulation [Andrews et al. 1987; see also Shepherd 1995]. This can be seen in several ways. First, the mass circulation of the stratosphere is essentially thermally indirect: air rises where it is relatively cold (the tropics) and sinks where it is relatively warm (the extratropics). Thus, the energetic forcing of the stratosphere must be mechanical rather than thermal. Second, the rotation of the Earth acts to inhibit mean meridional (north-south) motion. In order to maintain a mean meridional mass circulation, a zonal (east-west) mechanical force is required to balance the zonal Coriolis force that acts on meridional motion. (To put this another way, tropical air has more angular momentum than polar air, so to move air from one region to the other a zonal force must be applied to either increase or decrease its angular momentum.) This zonal mechanical force is provided by the drag force associated with the breaking and dissipation of wave motions propagating into the stratosphere from the more massive troposphere. In the stratosphere, most of this drag force is associated with large-scale planetary waves, with

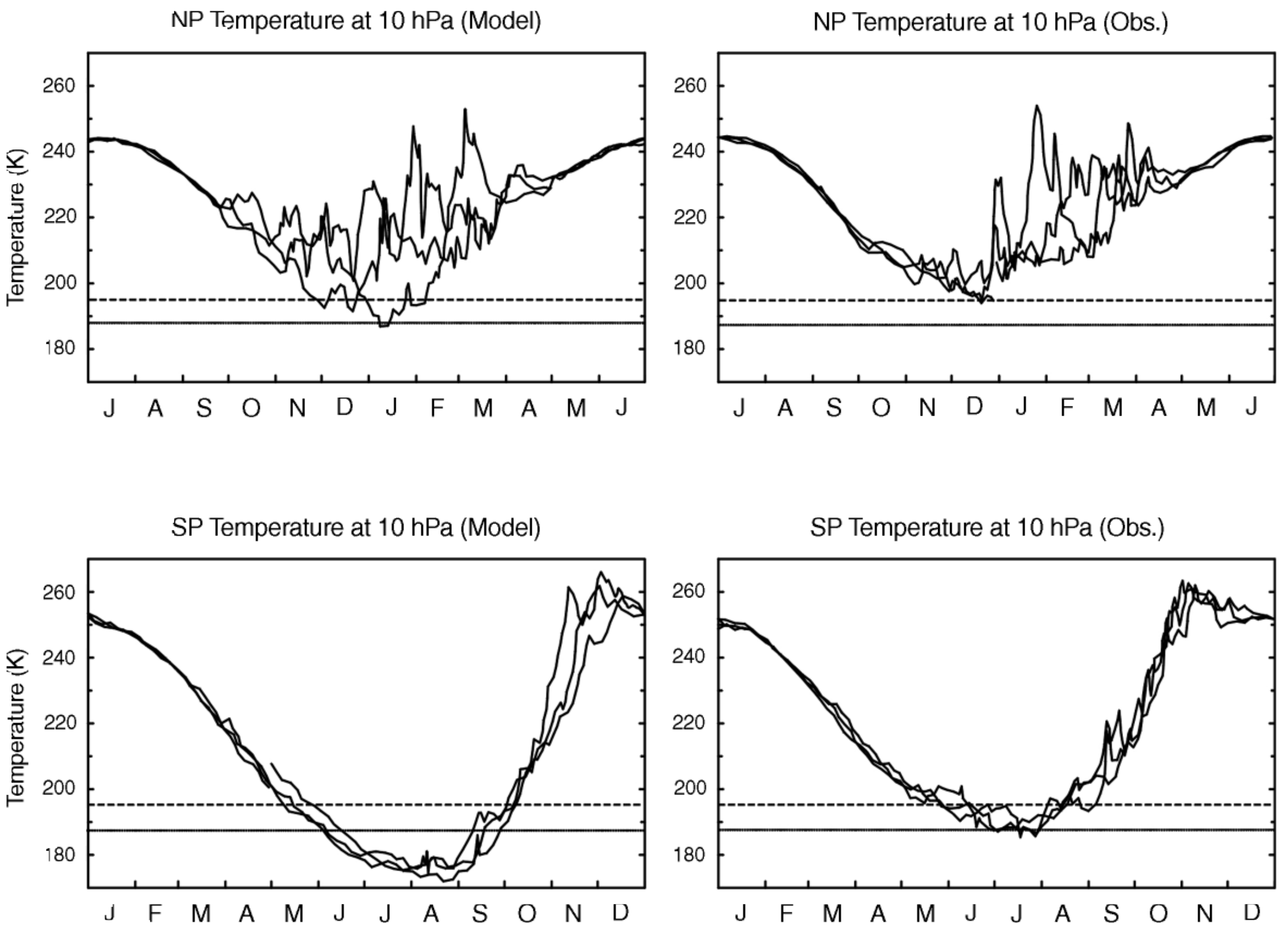


Figure 2.2 Annual variation of temperature at 10 hPa over the North Pole (NP) and South Pole (SP), centred in midwinter, for the Canadian Middle Atmosphere Model and observations. In each case, three different years are shown; the observations correspond to UK Meteorological Office analyses during 1993–1995. The temperature thresholds of 195 K and 188 K for Type 1 and Type 2 polar stratospheric clouds are indicated with dashed and dotted lines respectively. (Figure provided by John Koshyk, University of Toronto.)

a secondary contribution from smaller-scale inertia-gravity waves. It is the difference in the strength and variability of the stratospheric wave drag between the northern and southern hemispheres (resulting from their different surface topography) that accounts for the difference in the strength and variability of the wintertime mass circulation between the Arctic and Antarctic.

Although there is a consensus on the essential validity of the above picture, our quantitative understanding of it and predictive ability remain poor. For example, the most sophisticated atmospheric general circulation models are plagued by systematic errors in their polar temperatures, most notably the so-called “cold pole” problem in the Antarctic winter, which can be seen in Figure 2.2. The origin of this discrepancy is not known, though it is believed to be associated with missing gravity-wave drag [Garcia and Boville 1994]. If true, this implies a serious fundamental uncertainty in our quantitative understanding and predictive capability, since gravity-wave drag is extremely poorly constrained from either observations or theory. The magnitude of the temperature discrepancy in the Antarctic, 10 K or more, is enough to compromise quantitative modelling of ozone depletion; in practice, it is corrected through forced adjustment to observed winds and temperatures, but such “relaxational” corrections have been shown to be problematical in the context of climate-change modelling [Shepherd et al. 1996].

In our present climate, the difference between Antarctic and Arctic mid-stratospheric wintertime temperatures has enormous effects on the respective amounts of ozone loss, because the difference range happens to straddle the critical temperatures required for the formation of polar stratospheric clouds (PSCs; see Chapter 3). If, for example, the mean temperatures were 10 K higher in the Antarctic or 10 K lower in the Arctic, then the difference in ozone loss between the hemispheres would presumably be much less. (Of course, polar temperatures are only part of what matters for polar ozone loss; confinement by the polar vortex is also critical, as will be discussed in Section 2.5.) The high degree of variability of Arctic wintertime polar temperatures, not only within each season but also from one year to another, is responsible for the considerable interannual variability of Arctic ozone loss. As noted above, the variability of Arctic wintertime polar temperatures is directly attributable to dynamical variability. Since dynamical variability is poorly understood, and difficult to predict for any given year, this places a major limitation on our quantitative understanding of present and future Arctic ozone loss.

2.3 The Global Structure of Transport and Mixing

The ozone distribution provided one of the earliest pieces of observational evidence for the transport of trace species in the stratosphere [Dobson 1956]. Since most ozone production occurs in the tropics but the largest values of total ozone are found in the extratropics, there must, therefore, be systematic poleward transport out of the tropics. This inference is consistent with the observed low values of stratospheric water vapour, from which Brewer [1949] concluded that air must enter the stratosphere through the region of extremely low temperature at the tropical tropopause: the so-called "cold trap". The stratospheric transport circulation implied by these observations – the "Brewer-Dobson" circulation – rises in the tropics, moves poleward, and (by mass conservation) descends back into the troposphere in the extratropics. Confirmation of this picture by global observations is comparatively recent, as it is dependent on satellite measurements – most notably those from the Upper Atmosphere Research Satellite (UARS) launched by NASA in 1991. An example of such measurements is shown in Figure 2.3.

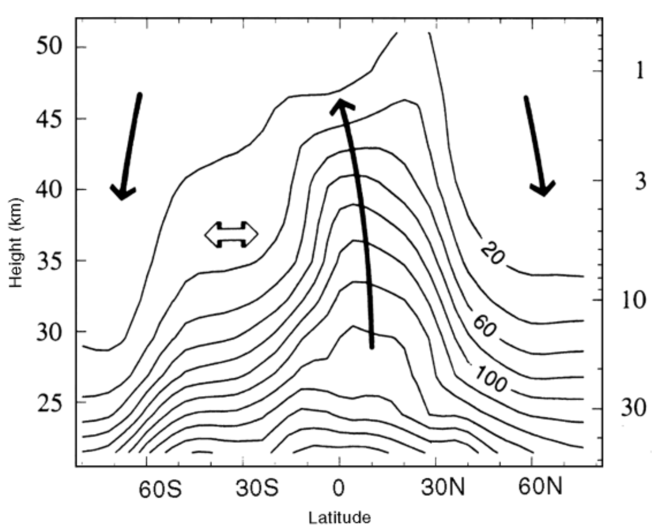


Figure 2.3 Longitudinally averaged structure of nitrous oxide (N_2O) mixing ratio (in parts per billion by volume) during 1–20 September 1992 as measured by the CLAES instrument on UARS. Heavy solid lines denote the mean stratospheric circulation in the latitude-height plane, and the horizontal arrows denote the location of quasi-horizontal mixing by planetary waves. (From Randel et al. [1993].)

The inferred structure of the Brewer-Dobson transport circulation is broadly consistent with the mean meridional mass circulation inferred from purely dynamical considerations. In one sense this is no surprise: the mass circulation describes the mean movement of air across (quasi-horizontal) isentropic surfaces (i.e., surfaces, of constant potential temperature, which determine stratification; air from everywhere on such a surface would, if compressed to the same pressure, acquire the same temperature and density). Thus, the mass circulation is linked in a fundamental way to the bulk vertical motion of air parcels. However, it must be emphasized that the mass circulation is an Eulerian diagnostic (the mean is taken at a fixed latitude); it is therefore distinct, and can be quite different, from the mean circulation defined by the movement of trace species, which is a Lagrangian diagnostic (the mean is taken following air parcels). An example in which the difference between the two circulations is particularly important is described in Section 2.5.

A central feature of the Brewer-Dobson circulation is its pronounced seasonal cycle; although there is tropical upwelling all year long, the poleward motion (and polar downwelling) occurs predominantly in the winter hemisphere. This behaviour is exhibited in Figure 2.4, which shows the seasonal cycle of the mean meridional mass circulation in the Canadian Middle Atmosphere Model [Beagley et al. 1997]. (Reliable estimates of the mass circulation from observations are difficult to obtain. As for a tracer-based transport circulation, even its definition is problematical.) It must be emphasized that the tropical upwelling rates are quite slow, so that an air parcel confined to the tropics takes more than a year to ascend through the ozone production region into the upper stratosphere; thus tracer distributions such as that shown in Figure 2.3 for nitrous oxide reflect the annual mean of the circulation shown in Figure 2.4, modified by more rapid quasi-horizontal mixing.

At a certain level, it may be said that we understand the origin of the Brewer-Dobson circulation and its seasonal cycle [Holton et al. 1995]. Planetary waves propagate westward relative to the mean wind, and thus exert a westward (i.e., negative) zonal force where they break. This westward force drives poleward motion in both hemispheres, which induces upwelling in the tropics and downwelling in the extratropics. The seasonal cycle in the Brewer-Dobson circulation is likewise a consequence of the seasonal cycle in stratospheric planetary-wave drag [Rosenlof 1995]; most planetary-wave

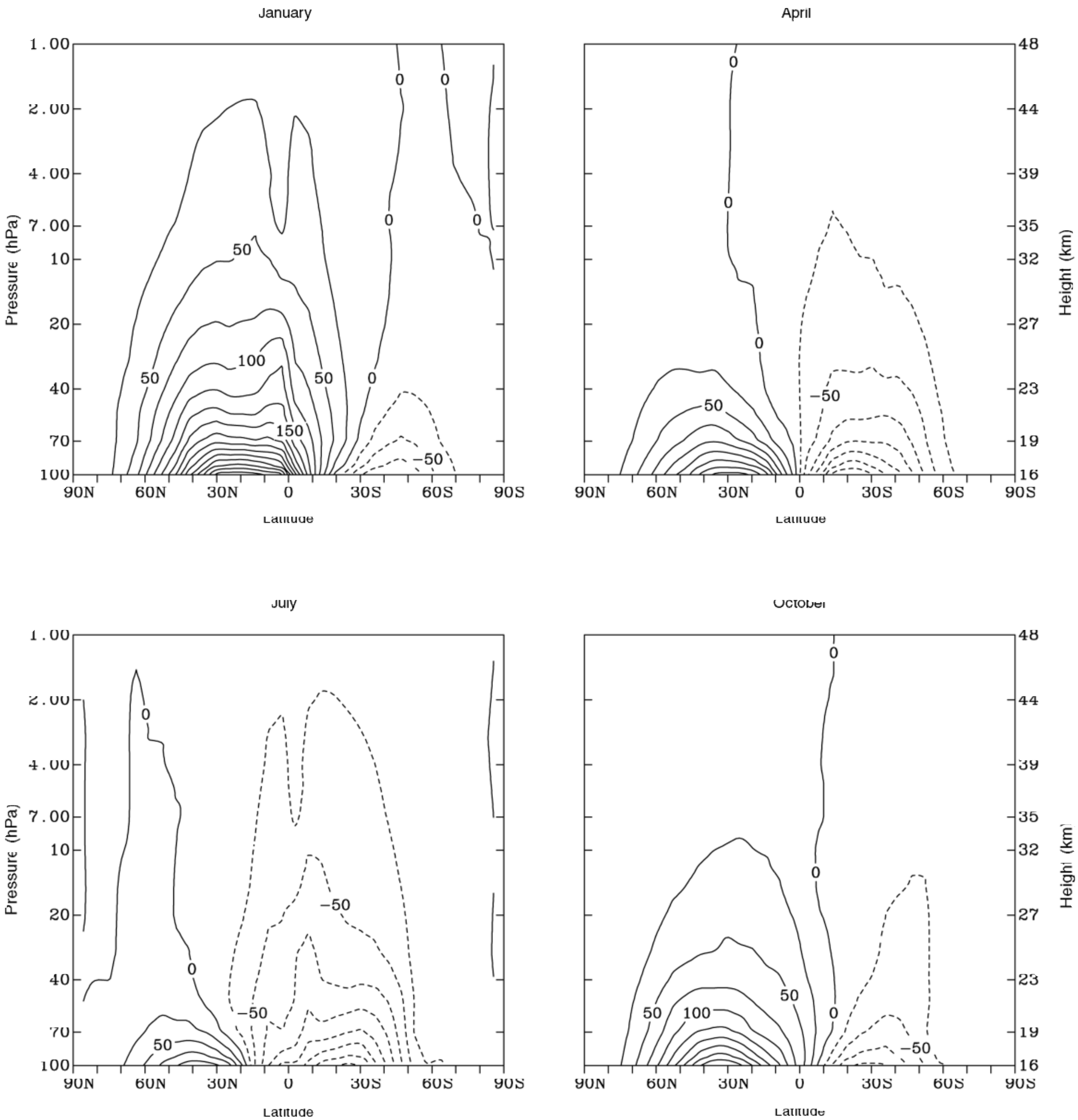


Figure 2.4 The seasonal cycle of the mean meridional mass circulation of the Canadian Middle Atmosphere Model. Each panel represents the average over three different simulated years of the monthly mean for January, April, July, or October. Contours denote mass streamlines, and correspond to rising motion in the tropics and sinking motion in the extratropics. The contour interval is 25 kg m^{-1} s^{-1}. (Figure provided by John Koshyk, University of Toronto.)

drag occurs in the winter hemisphere. A similar relationship must also hold on longer timescales. It follows that the interannual variability in northern hemisphere planetary-wave drag reflected in Arctic wintertime temperatures (see Section 2.2), and the potential for its systematic change over decadal timescales, has concomitant implications for variability and change in the Brewer-Dobson circulation and hence in the ozone distribution. But even if dynamical variability could be predicted, there remain significant gaps in our quantitative understanding of how this relates to ozone transport. Most notably, the argument outlined above relies on the tacit equivalence of the Brewer-Dobson, mean mass (i.e., radiatively balanced), and wave-drag-driven circulations. Although these circulations are certainly related, they are far from identical, and their differences are not yet understood on a quantitative level. This issue is discussed in more detail later in the context of tropical upwelling (Section 2.4) and polar downwelling (Section 2.5).

There are a number of different circulation timescales that are relevant to the determination of ozone abundance. For ozone-depleting substances with tropospheric sources and stratospheric sinks, one may define a residence time based on the decay timescale required to account for observed stratospheric concentrations. Such residence times are clearly different for different chemical species; they reflect both the decay processes themselves and the time it takes for the species to reach the altitudes where the decay occurs. Many substances have stratospheric residence times of decades or longer. A stratospheric age timescale may also be defined, based on the time lag between tropospheric values of long-lived species and their value at a certain location in the stratosphere. For a tracer with linearly increasing tropospheric values, this provides a well-defined age that is a tracer-independent quantity [Hall and Plumb 1994]; it is estimated to be between one and two years in the lower extratropical stratosphere, and greater than four years in the upper stratosphere. (The contours of age look rather like the nitrous oxide contours in Figure 2.3.) These timescales are of the same order of magnitude as those obtained on the basis of the mean meridional mass circulation alone, without mixing [Rosenlof 1995], but they have a very different spatial distribution. There is, however, no straightforward connection between this definition of age and the time lags of species with nonlinear time dependence, especially those with a strong seasonal cycle. In principle, all transport information (for long-lived species) is contained in the "age spectrum" [Hall and Plumb 1994], but the age spectrum is only well-defined for stationary transport and mixing statistics; it is not clear how robust the concept is in the presence of annual and interannual variability in the dynamics. For species affected by stratospheric sources and sinks, the age spectrum provides incomplete information, because one needs to know the distribution of transport pathways.

Traditionally, transport and mixing in the stratosphere has been approached as an advective-diffusive problem [Andrews et al. 1987], with tracer distributions such as those of Figure 2.3 regarded as a balance between the slope-steepening effects of the mean meridional mass circulation and the slope-flattening effects of quasi-horizontal mixing. While this is probably a reasonable description in midlatitudes, like all mixing-length approaches it depends on a spatial scale separation between (large) tracer scales and (small) eddy scales. Thus it is problematical in regions where the size of the eddies is greater than the tracer-gradient length scale, as occurs in strong gradient regions such as the edge of the polar vortex, the edge of the tropics, and the tropopause. The traditional mixing-length approach also fails to account for the robust transport barriers that provide such strong tracer gradients in the first place, as will be discussed in more detail below. (Although these are not perfect barriers, but rather quasi-barriers, the term "transport barrier" will be used hereafter for conciseness.) The role of the wintertime polar vortex in isolating vortex air, thereby allowing heterogeneous chemical processing to occur, was recognized early on as an essential ingredient in polar ozone loss [Schoeberl and Hartmann 1991]. More recently, with the availability of high-quality tropical measurements, analogous transport barriers have been identified at the edge of the tropics [Trepte and Hitchman

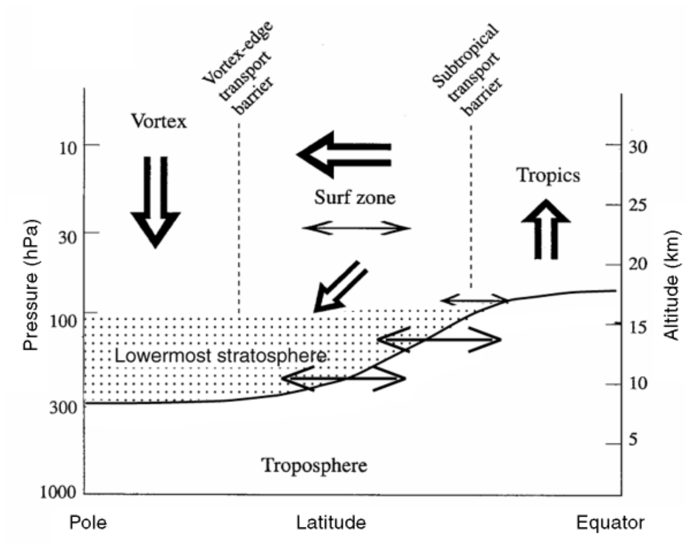


Figure 2.5 Schematic of transport and mixing in the winter hemisphere of the lower stratosphere. Heavy single-headed arrows indicate the mean mass circulation, and thin double-headed arrows indicate strong mixing. Vertical dashed lines represent transport barriers. The solid curve denotes the tropopause, while the stippled region denotes the lowermost stratosphere, in which isentropic surfaces intersect the troposphere.

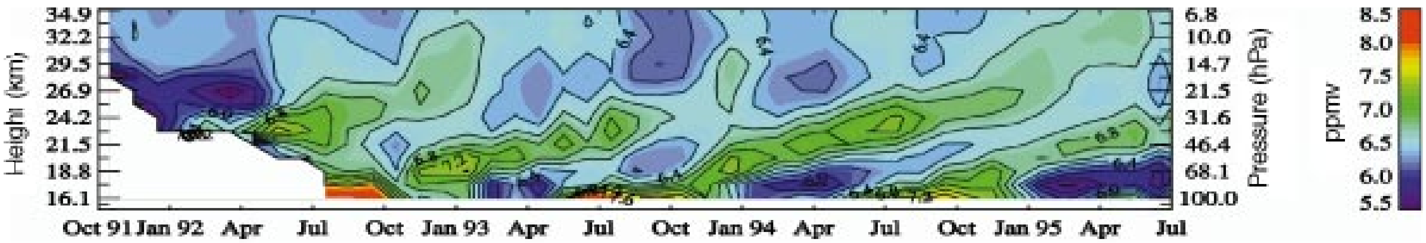


Figure 2.6 Time-height section of 2(CH_4)+(H_2O), the variable part of total hydrogen, between 12˚S and 12˚N, measured by the HALOE instrument on UARS. The vertically propagating oscillation is indicative of tropical ascent. (From Mote et al. [1996].)

1992; Fahey et al. 1996]. The lowest part of the extratropical stratosphere is also a distinctive region, because it is "ventilated" by isentropic surfaces that intersect the troposphere [Holton et al. 1995]. Accordingly, the current picture of stratospheric transport and mixing is far more complex than it was a decade ago, and in some ways more fragmented. A schematic of the various elements in the picture is provided in Figure 2.5.

2.4 Tropics

The tropics play an important role in establishing ozone abundance in a number of ways. First, the tropical stratosphere is the principal source region for ozone, which the Brewer-Dobson circulation then transports into the extratropics. Second, the lower tropical stratosphere represents the principal stratospheric gateway for ozone-depleting substances. A current issue of great concern in this respect is the potential impact of NO_x emissions from proposed high-altitude civil aircraft flying in the extratropical lower stratosphere [NASA 1995]. If the NO_x gets directly entrained into the tropical ascent region, instead of being diluted in the troposphere, then the potential for ozone destruction is enormously greater (see Chapter 3). Third, the temperature of the tropical tropopause regulates the amount of water vapour in the stratosphere through Brewer's "cold trap" effect, and this affects the temperature threshold for PSC formation. In this way, a long-term trend in the tropical tropopause temperature could have an indirect impact on ozone destruction.

A significant development of the past few years has been the realization that the ascent that occurs in the tropical branch of the Brewer-Dobson circulation is relatively isolated. This was first seen in aerosol measurements following the eruption of Mount Pinatubo in 1991 [Trepte and Hitchman 1992; Grant et al. 1994], which showed limited transport of aerosol into the extratropics above about 22 km. Further evidence of a subtropical transport barrier is evident in latitudinal profiles of the chemical correlations of long-lived species [Fahey et al. 1996], which suggest distinct mixing regions in the tropics and extratropics. But perhaps the most dramatic evidence comes from satellite measurements of water vapour in the tropical lower stratosphere, which reveal a meridionally confined, vertically propagating oscillation, shown in Figure 2.6; Mote et al. [1996] argue that this represents the imprint of the annual cycle of tropical tropopause temperatures on the water vapour content of the air being carried up by the mean meridional circulation – in other words, an annual modulation of the "cold trap" effect, carried upwards.

However, there is considerable variability in the existence and strength of the subtropical transport barriers; they depend on season, and are significantly modulated by the QBO. This is consistent with the notion that they are explainable in terms of the spatial structure of the horizontal velocity field: transport barriers correspond to quasi-zonal streamlines, without stagnation points [Polvani et al. 1995]. It is well established that the midlatitude stratosphere is disturbed by breaking planetary waves, forming a "surf zone" [McIntyre and Palmer 1983]. The nature of this breaking is consistent with the predictions of Rossby-wave critical-layer theory. A particularly striking example is shown in Figure 2.7, at about 21 km

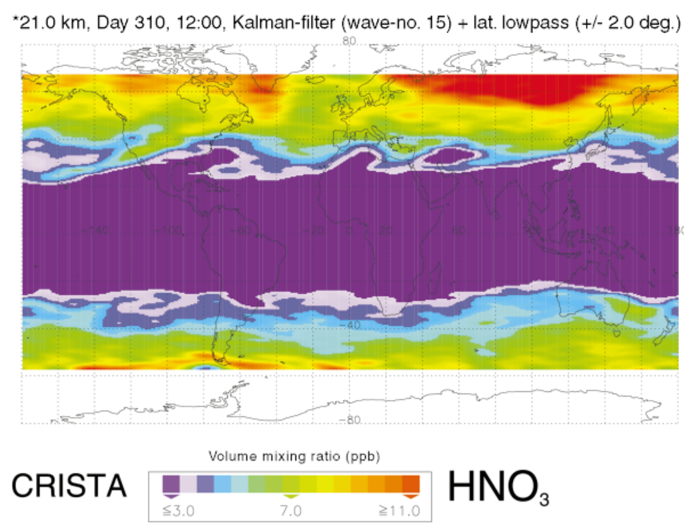


Figure 2.7 Horizontal cross-section of HNO_3 at about 21 km altitude on 6 November 1994, measured by the CRISTA instrument on the Space Shuttle (personal communication, D. Offermann, Wuppertal University, Germany). The field shows several cat's-eye structures in the northern subtropics.

altitude; the tracer field exhibits the characteristic "cat's eye" structure of a Rossby-wave critical layer. Within the cat's eyes, strong horizontal mixing occurs, but on either side of the cat's eyes, tracer contours remain quasi-zonal [Ngan and Shepherd 1997a]. From this perspective, the subtropical transport barrier represents the tropical limit of the midlatitude surf zone. (Similarly, the polar vortex transport barrier represents the polar limit of the surf zone; see Section 2.5.) Its existence, therefore, depends on whether or not breaking planetary waves extend into the tropics. This, in turn, depends on the amplitude and phase speed of the planetary waves and the meridional shear of the underlying zonal flow. Both depend on season, while the latter also depends on the phase of the QBO.

Because of the subtropical transport barriers, the transport and mixing characteristics of the tropics are qualitatively different from those of midlatitudes. An extreme idealization is that of a "tropical pipe" [Plumb 1996], where tropical air is assumed to ascend with no exchange with midlatitudes. Certainly the water vapour signal shown in Figure 2.6 suggests there is some validity to this concept. However, detailed chemical modelling within the tropics shows that the observed vertical profiles of chemical species cannot be reconciled with a strict interpretation of the tropical pipe; some dilution from midlatitudes is required [Avallone and Prather 1996], especially below 22 km or so. There have been a number of recent attempts to quantify the amount of dilution by balancing the dilution against vertical upwelling and chemical processing [e.g., Volk et al. 1996]. Most calculations estimate that, at altitudes above 22 km, 50% or more of the air in the tropical upwelling region is of midlatitude origin. Such calculations are subject to considerable uncertainty. First, they rely on an extreme idealization of lateral exchange between two homogeneous mixing regions; second, they depend quite sensitively on the estimate of the tropical upwelling rate.

The upwelling rate cannot be directly measured, and must be inferred from the estimated radiative heating rates in the tropics. (The vertical propagation rate of an annual cycle, such as that seen in water vapour in Figure 2.6, is not the same as the mean upwelling rate if there is significant dilution [Hall and Waugh 1997].) However, this is a very difficult calculation, as it depends on a small difference of large terms, and is thus prone to sampling errors – especially in the lower stratosphere where radiative timescales are long. The solution is also generally non-unique. A number of recent studies [e.g., Rosenlof 1995] have used the UARS data set to attempt to make accurate estimates of the upwelling rate, and while there is broad agreement in the results, uncertainties remain at about the 50% level in the lower stratosphere and increase as one approaches the tropopause.

A further difficulty is that one cannot assume a simple balance between radiative heating and vertical upwelling in the tropics, because of inertial adjustment that is expected to occur in response to radiatively forced inertial instability [Dunkerton 1989]. (This is the horizontal analogue of convective adjustment in the vertical.) The way in which these processes interact to determine tropical temperatures in the real stratosphere has yet to be established at a quantitative level suitable for diagnostic analysis. The resulting difficulty in relating radiative heating to vertical upwelling provides a major limitation on our understanding of the tropical branch of the Brewer-Dobson circulation.

While most interest has focused on lateral inmixing into the tropics from midlatitudes, one expects this process also to be associated with lateral outmixing. There is clear evidence of outmixing events in the middle stratosphere, corroborated by tracer advection calculations [Randel et al. 1993]. Aerosol measurements also suggest significant outmixing in the lower stratosphere [Grant et al. 1994]. Figure 2.8 shows an aerosol cross-section from the Lidar In-Space Technology Experiment (LITE), which flew on the space shuttle [McCormick 1996]. Given that stratospheric aerosol is injected in the tropics (most notably from the eruption of Mount Pinatubo in 1991), this figure shows poleward spreading just above, and following, the tropopause. In addition, measurements of water vapour [Dessler et al. 1995], carbon dioxide [Boering et al. 1994], and ozone [Folkins and Appenzeller 1996] in the midlatitude

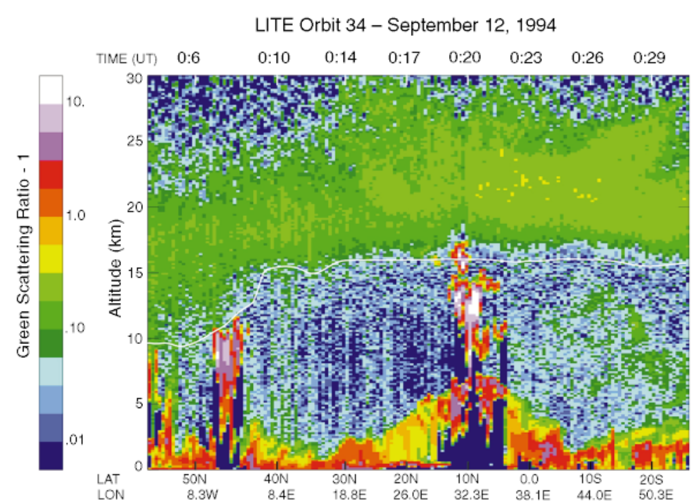


Figure 2.8 Aerosol scattering ratio cross-section on 12 September 1994 measured by the LITE experiment on the Space Shuttle [Hoff and Osborn 1996]. The aerosol layer in the lower stratosphere is largely due to the remnants of the Mount Pinatubo eruption in 1991 (green); in the lower troposphere, high concentrations of aerosol from Europe to east of Africa can be seen near the surface (red-yellow). The thin line indicates the location of the tropopause from US NMC data.

lower stratosphere point to a direct transport pathway from the tropical upper troposphere/lower stratosphere into the midlatitude lower stratosphere, which amounts to a "short-circuiting" of the Brewer-Dobson circulation. In fact, the lowest part of the tropical stratosphere is revealing itself as a particularly complex part of the atmosphere, which plays a critical role in determining the global structure of stratospheric transport and mixing. It remains one of the most poorly understood regions of the atmosphere. In particular, the way in which the rapid convective dynamics of the tropical troposphere connect to the slower dynamics of the tropical stratosphere, over what is properly regarded as a tropical tropopause layer (rather than a simple interface), represents a major gap in our theoretical understanding. For example, most current conceptual models completely ignore the effect of strong longitudinal asymmetries such as the monsoon circulation.

It should be emphasized that inmixing and outmixing are distinct processes. They are equivalent in the limit of small eddies, as is assumed in traditional mixing-length approaches, but – as noted in Section 2.3 – the subtropical transport barriers represent precisely the opposite situation, where large eddies act on a sharp tracer gradient. The scales involved are exemplified by Figure 2.7. If planetary-wave breaking is largely unidirectional, then one can have significant outmixing with little inmixing, or vice-versa; it is well established that this is the case in the wintertime polar vortex (see Section 2.5). Our understanding in the tropics is much more limited, in comparison, because of the current unavailability of reliable wind measurements in the tropical stratosphere. Estimates of stratospheric winds are largely derived from temperatures, assuming gradient-wind balance, but this balance breaks down in the tropics. A high priority for future measurement capabilities in the stratosphere is the development of reliable wind measurements, in order to provide better constraints on our understanding of transport and mixing. While direct wind measurements would be the most obvious solution, there is some hope that advanced data assimilation techniques might be able to obtain reliable wind information from the time evolution of tracer fields, such as that shown in Figure 2.7.

2.5 Wintertime Polar Vortex

The strong polar vortex that develops in the winter hemisphere provides the basic dynamical setting for polar ozone loss. The strength of the vortex is directly related, by thermal-wind balance, to the cold temperatures required for ozone-depleting heterogeneous chemistry; at the same time, the vortex provides a transport barrier that isolates the polar air from midlatitude dilution. As noted in Section 2.2, these effects are much more pronounced in the Antarctic than in the Arctic, for reasons that are reasonably well understood, and this accounts for the fact that the most dramatic polar ozone loss has occurred in the Antarctic.

From the perspective of transport and mixing, there are many useful parallels between the wintertime polar vortex and the tropics. In both cases, the Brewer-Dobson circulation provides one-way vertical transport (up in the tropics, down in the polar vortex), while a transport barrier is established at the relevant edge of the midlatitude surf zone. As in the tropics, the polar-vortex transport barrier is not perfect, and much interest has focused on quantifying the extent of inmixing and outmixing. Our quantitative understanding of transport and mixing processes is, however, considerably greater for the wintertime polar vortex than it is for the tropics for a number of quite different reasons. First, since the discovery of the Antarctic ozone hole there have been numerous aircraft missions directed at polar regions (both Arctic and Antarctic). Second, global wind measurements are far more reliable in the polar regions than they are in the tropics, thus making possible the calculation of detailed air-parcel trajectories. Third, much tighter theoretical constraints are available on the mean meridional mass circulation in the polar case, because of the strong meridional gradient in angular momentum [Holton et al. 1995].

One of the principal difficulties in quantifying transport and mixing in and around the wintertime polar vortex has been the definition of the vortex edge. The dynamical edge is generally considered to be the location of the maximum gradient in potential vorticity. Since this is a highly derived quantity, and therefore difficult to measure (especially on an aircraft flight track), researchers tend to use the maximum zonal velocity instead. The distinction appears to make quantitative rather than qualitative differences in transport diagnostics. A kinematical edge can be defined in terms of the structure of the velocity field, as the edge of the region of quasi-zonal streamlines without stagnation points. This should correspond most closely to a transport barrier. Observations often show the dynamical and kinematical edges to be distinct entities. In fact, the kinematical edge is just the outer limit of a barrier region of finite extent [Bowman 1993]. This behaviour is represented in Figure 2.9, which shows the evolution of tracer particles on a single (quasi-horizontal) isentropic surface in the Canadian Middle Atmosphere Model. If the barrier region extends well into the vortex, this creates the possibility of further edges in tracer fields within the vortex itself. In particular, measurements sometimes reveal a chemical edge inside the vortex, associated with chemical processing in the polar night within a confined region.

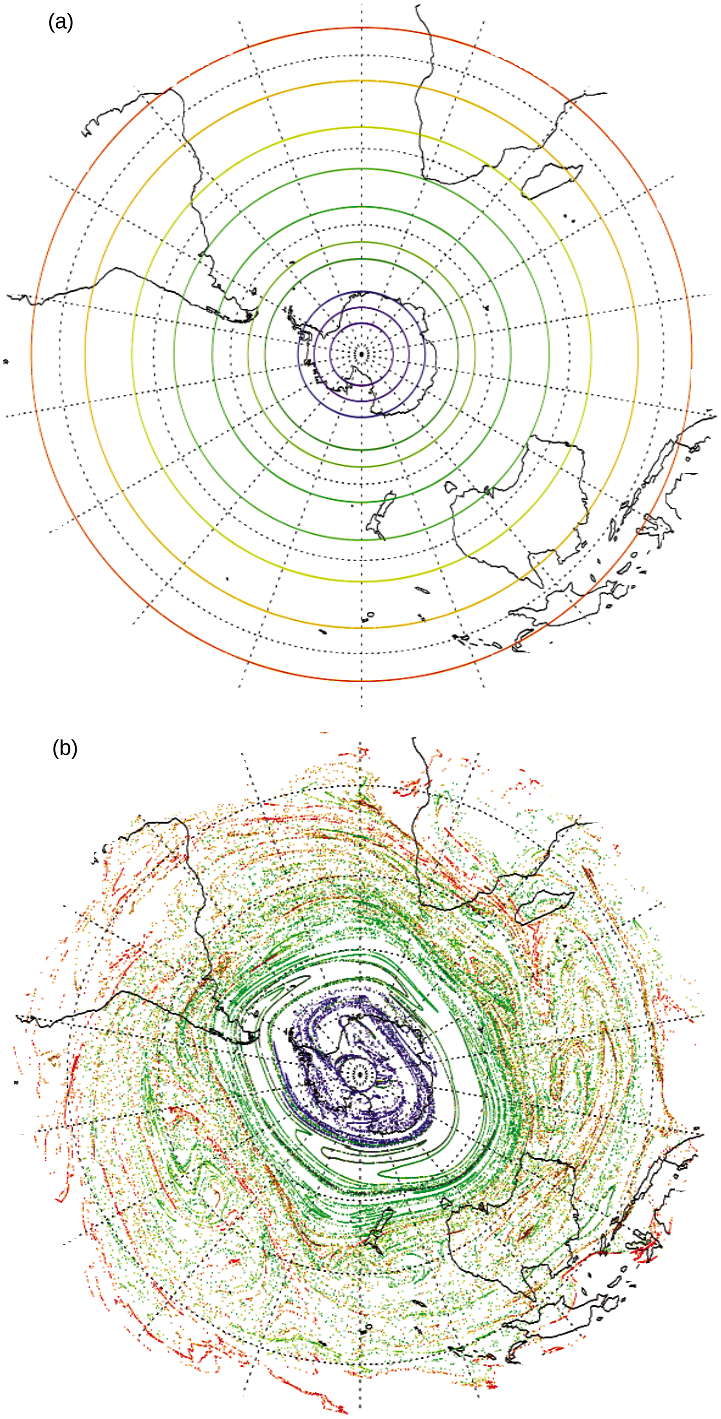


Figure 2.9 Evolution of passive tracer particles in the lower stratosphere during the southern hemisphere winter (July) from the Canadian Middle Atmosphere Model. (a) shows the initial distribution, and (b) the distribution after 30 days using quasi-horizontal winds interpolated onto the 450 K isentropic surface, which is located at about 17 km altitude. Thus, diabatic motion is neglected in this calculation. The particle evolution reveals strong mixing in the midlatitude "surf zone" (including the presence of several large cat's-eye structures), coexisting with comparatively regular behaviour in the outer part of the vortex, manifested in quasi-zonal wrapping of lines of particles. (Figure provided by Keith Ngan, University of Toronto.)

A striking feature of the polar vortex is that mixing very much tends to be outward rather than inward [Waugh et al. 1994]. Inmixing does occur on occasion in the Arctic, but it is associated with rare, large-amplitude perturbations that provide a major disruption of the vortex [Plumb et al. 1994] and are thus connected to sudden warmings such as those seen in Figure 2.2. There has been some debate in recent years over exactly how much outmixing occurs, especially in the Antarctic. This is a critical issue for ozone depletion, because the amount of ozone loss over a winter season is directly proportional to how much air can be chemically processed; a leaky vortex can support higher ozone destruction. (On the other hand, a leaky vortex requires greater vortex downwelling, which would warm the vortex and thus perhaps limit ozone destruction.) Mass conservation constraints from the mean meridional circulation appear to limit the amount of outmixing in the Antarctic middle stratosphere (down to about 16 km) to an amount consistent with the vortex being flushed out only once over the period of ozone loss. Similar kinds of considerations are of less relevance for the Arctic, because it is rarely intact through the winter season. Considerable uncertainty remains, however, with regard to the characterization of transport and mixing in the subvortex region (below about 16 km).

Because of mixing, there is an important distinction between the descent provided by the mean meridional mass circulation, which is in balance with mean radiative cooling, and the Lagrangian-mean descent of long-lived chemical species. Satellite measurements of chemical species [Russell et al. 1993] show clear evidence of unmixed descent in the Antarctic vortex, maximizing at the pole, which is consistent with air-parcel trajectory calculations [Fisher et al. 1993]. However, radiative cooling – and thereby the descent rate of the mass circulation – is greatest not over the pole, but over the vortex edge. This difference in the location of the peak descent rates can be understood by recognizing that the vortex edge represents a region of significant lateral mixing. Air parcels initially located at the vortex edge do not remain there, but perform large excursions into the midlatitude surf zone where they experience much less radiative cooling. Hence, the descent rate of the (Eulerian) mean mass circulation at the vortex edge is a significant overestimate of the actual Lagrangian descent of air parcels. It is worth remarking that this phenomenon arises from the fact that the diabatic cooling rate varies strongly (with latitude) over the length scale characterizing the air parcel motion; in the opposite, small-eddy limit – which is that assumed in traditional mixing-length approaches – the two descent rates would have to be essentially equivalent.

As noted in Section 2.2, dynamical variability of the wintertime polar vortex is much greater in the Arctic than in the Antarctic, and temperatures sufficiently cold to allow PSC formation therefore occur far less often. Consequently, Arctic ozone depletion is comparatively more limited and more variable from year to year. It may be that predictions of ozone loss in the Arctic will never be possible, because of the inherent unpredictability of wintertime dynamical variability. Nevertheless, it is still important to achieve sufficient understanding of the relevant dynamical processes so that reasonable bounds can be placed on the range of variability of Arctic ozone destruction. Understanding at this level remains poor. Even when constrained by observed winds and temperatures, stratospheric models with sophisticated ozone chemistry have been unable to account for the observed amounts of Arctic ozone depletion, with shortfalls in the range of 40%. Edouard et al. [1996] have argued that this discrepancy can be explained by insufficiently fine horizontal resolution in the models. Because the catalytic halogen species may be concentrated in filamentary structures, and because the chemistry of ozone depletion is strongly nonlinear with respect to the concentrations of those species, the rate of ozone destruction may therefore be underestimated if such structures are not well represented. (In other words, the same amount of halogen destroys ozone more rapidly when it is concentrated in intense filaments rather than spread out over a broad region.) Such considerations are of most relevance in the Arctic, because of the incompleteness of the ozone destruction; in the Antarctic, where polar ozone destruction is essentially complete, the rate is not relevant to the final result.

It should be emphasized that although models may require fine spatial resolution in order to represent chemical tracers, they do not necessarily require equally high resolution for the dynamical fields, because it is only the large-scale wind field that appears to be relevant to the development of realistic small-scale structure in the tracer field. This is quite consistent with our understanding of quasi-horizontal stirring and mixing (see also Section 2.6), and – it may be added – with the existence of transport barriers. A possible exception concerns the effect of small-scale temperature inhomogeneities on PSC formation, which has not yet been quantified.

The implications of inadequate spatial resolution in models are complex, because there are competing effects. If the spatial resolution is too coarse, and the tracer fields too diffuse, then reactions between distinct air masses (for example, following an intrusion of midlatitude air into the polar vortex) will occur too quickly, before the actual (i.e., the fine-grained) air masses have actually mixed. On the other hand, when they finally do mix, if the tracer concentrations are too diffuse, then the resulting chemical effects will be underestimated. Such considerations are especially relevant to the important question of the possible impact of polar ozone chemistry on midlatitude ozone (see Section 2.6).

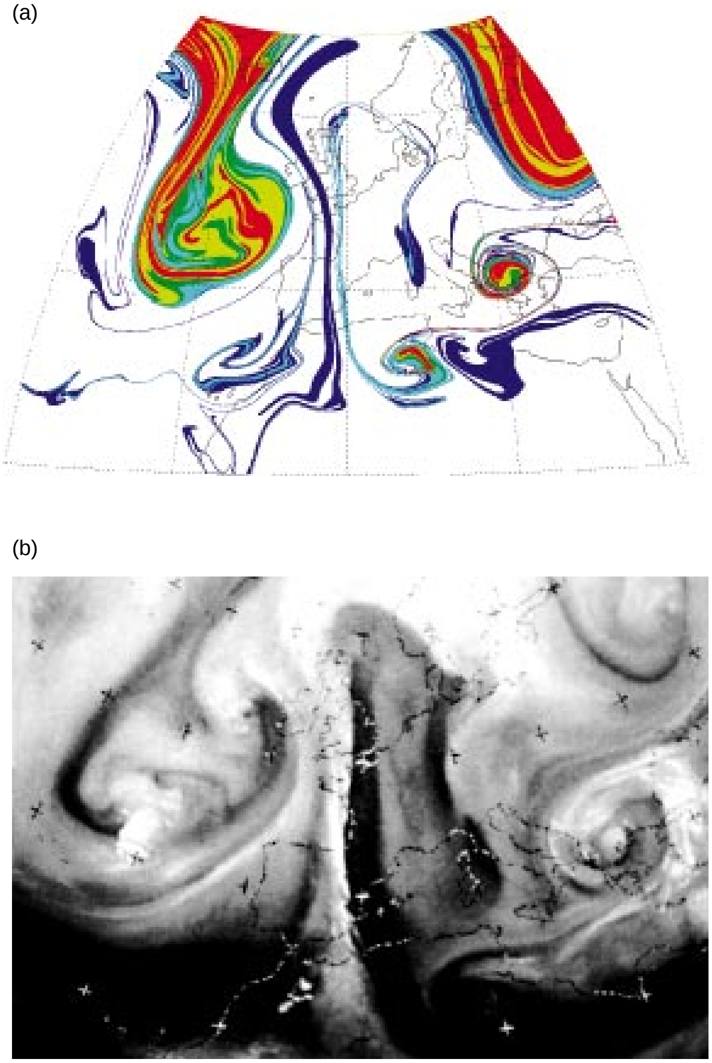
2.6 Midlatitudes

The greatest uncertainties in our present understanding of observed ozone changes have to do with the cause of midlatitude changes. There are many ways in which dynamical processes can be expected to be relevant. First, the height and structure of the midlatitude tropopause has a direct bearing on total ozone abundance. Second, the strength of the Brewer-Dobson circulation controls the transport of ozone from the tropical source region into the midlatitudes, and also controls the rate of mean descent through the lowermost stratosphere into the upper troposphere [Holton et al. 1995]. Third, the transport of chemically processed air out of the wintertime vortex – either in a major vortex-breaking event, such as a sudden warming, or in the final breakdown of the vortex – provides a strong chemical perturbation in midlatitudes. The last example contains competing effects; although breakdown and flushing out of the vortex ultimately limit ozone loss, there are short-term losses in midlatitudes as processed air reaches to lower latitudes and receives more sunlight. The extent of midlatitude ozone loss can be expected to depend sensitively on how rapidly this processed air gets diluted in the surf zone, which, like the vortex breakdown itself, depends on the large-scale flow.

The arguments given in Section 2.5 concerning the scale dependence of halogen-induced chemical ozone loss in the Arctic apply equally to midlatitudes, if not more so. Such scale dependence reflects the fact that large-scale, quasi-horizontal motion is the dominant contributor to mixing in the stratosphere, and that “stirring” by such motion generically leads to filamentary structure. An example from the lower stratosphere is shown in Figure 2.10. Stratospheric mixing is, therefore, quite opposite to diffusive mixing: homogenization is achieved first at large scales, and only subsequently at small scales; extremes in the concentration field are preserved even as small scales develop; and the advective development of small scales is a precondition for mixing. Because of the nonlinearity of ozone chemistry, it thus becomes crucial to achieve a good understanding of the small-scale structure of ozone and of ozone-depleting substances.

The existence of widespread filamentary structure in the stratosphere, as shown in Figure 2.10, is well established from measurements and has been clearly linked to large-scale advection acting on the sharp gradients characterizing the vortex edge [Waugh and Plumb 1994]. On the other hand, vertical profiles of ozone taken by ozonesondes have long

(a)



(b)

Figure 2.10 (a) Pattern of selected potential vorticity contours on the 320 K isentropic surface at 1200 UT on 14 May 1992, derived using the passive contour advection technique. The calculation begins 96 hours earlier (on 10 May 1992) from analyzed potential vorticity. (b) The satellite radiance image taken at the same time as (a) confirms the physical reality of the predicted filamentary and vortical structures. (From Appenzeller et al. [1996a].)

shown evidence of "laminae," namely isolated layers of anomalous values [Dobson 1973]; some examples are provided in Figure 2.11. Until recently, it had been widely assumed that these laminae result from inertia-gravity waves – though it was never clear why a wave would produce a vertically isolated, layered perturbation to an ozone profile. (Interestingly, *subtropical* laminae have been linked for some time to quasi-horizontal advective intrusions of tropical air [e.g., Andrews et al. 1987, Section 9.5.1].) It is rather surprising that the connection between filaments and laminae has only recently been made, because they are clearly two sides of the same coin: because of vertical shearing of the large-scale quasi-horizontal flow, the filaments seen in tracer structure at a given level are just the horizontal manifestations of sloping sheets of tracer, whose vertical manifestations are laminae. This explanation of ozone laminae has recently been established by several groups [e.g., Orsolini 1995]. The process has been quantified by Haynes and Anglade [1997], who argue that for conditions characteristic of the lower stratosphere, the asymptote of the ratio of the horizontal to the vertical length scales, in the long-time limit, should be somewhere around 250. The limiting process is small-scale vertical diffusion.

The existence of ozone filaments and laminae is consistent with the fact that horizontal spectra of ozone calculated from aircraft profiles in the stratosphere exhibit roughly a k^{-2} power-law scaling, where k is the horizontal wavenumber [Bacmeister et al. 1996]. The most obvious explanation of this scaling is that it is the singularity spectrum arising from the presence of a finite number of jumps in ozone concentration, associated with filamentation of the vortex edge [Ngan and Shepherd 1997b]. (The same scaling is found for the TOMS data [Sirovich et al. 1995], where it is presumably associated with the vortex edge itself.) The scaling holds down to the smallest resolvable length scales, of about 0.5 km, which suggests that we have yet to determine the ultimate scale at which three-dimensional effects overcome two-dimensional advection in the stratosphere. This sets a lower limit on vertical diffusion, and on the mix-down time for passive tracers; the latter must be about two weeks or greater. This is comparable to the estimated chemical lifetime of ozone filaments in midlatitudes [von der Gathen et al. 1995].

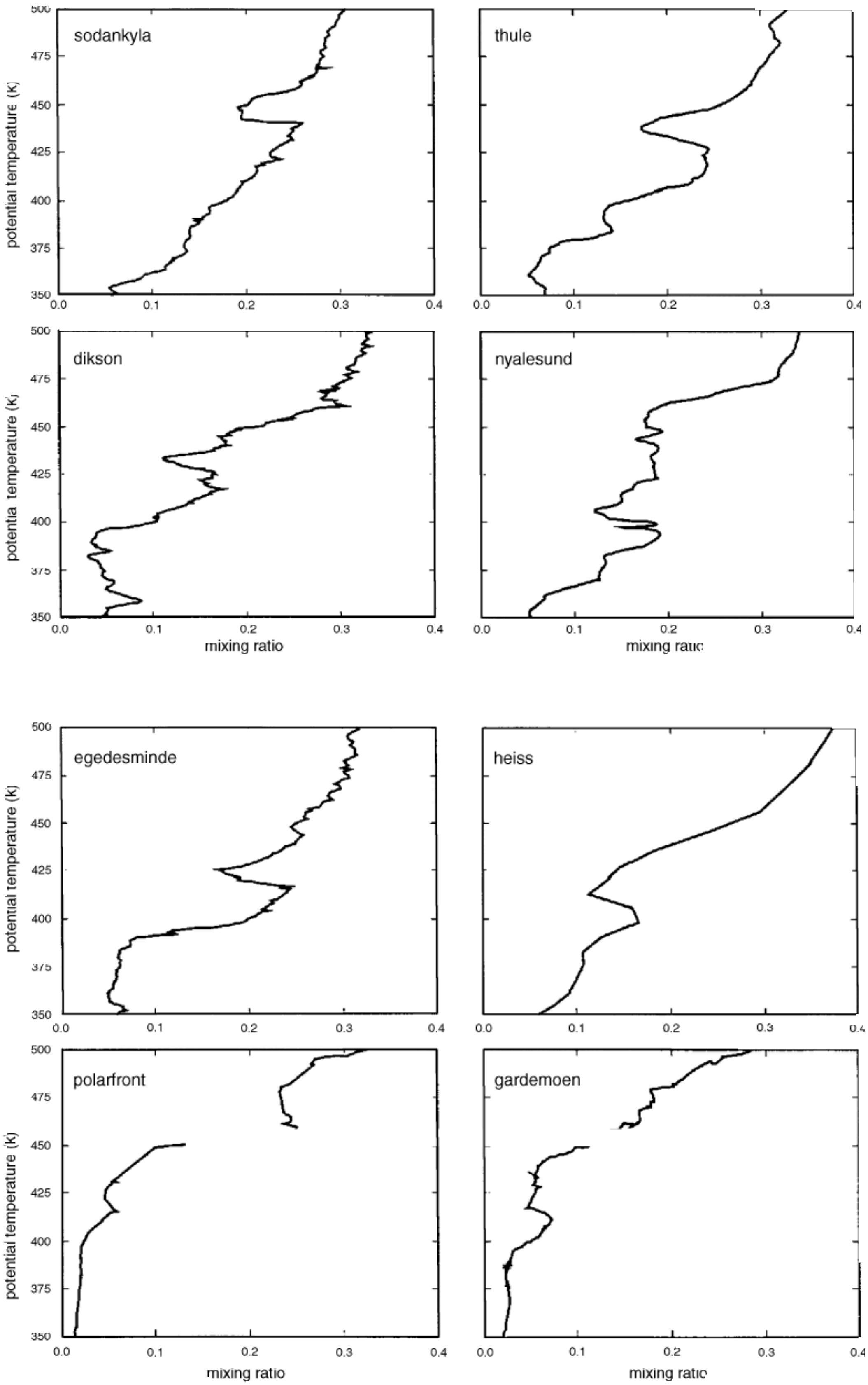


Figure 2.11 Ozone mixing-ratio profiles (in 10 parts per million by volume) taken over northern Europe and the northern Atlantic by ozonesondes on 29 January 1992. Height is measured by potential temperature (K). (After Orsolini [1995].)

When they first develop, ozone filaments will be associated with anomalous values of potential vorticity, because both ozone and potential vorticity are materially conserved on short timescales. However, the potential vorticity signature of these filaments will be increasingly damped – not diffused – by radiative cooling as their vertical scale decreases. Haynes and Ward [1993] have estimated the resulting horizontal cut-off scale in the lower stratosphere to be around 50 km, which is considerably larger than the cut-off scale estimated above for tracer filaments. (Tracers are not directly affected by radiative processes.) It follows that one should expect to find chemical filaments with no corresponding potential-vorticity signature. Such structures have indeed been found in the lowermost stratosphere in water vapour [Dessler et al. 1995] and in ozone [Folkins and Appenzeller 1996].

Any global budget of ozone in the extratropical lower stratosphere must take account of ozone transport into and out of this region. A recent diagnostic analysis by Appenzeller et al. [1996b] has shown that there is a seasonal cycle in the structure of the tropopause that leads to a seasonal cycle in the mass of the extratropical lower stratosphere; in the northern hemisphere (though not in the southern hemisphere) there is a corresponding lag between the mass flux in and the mass flux out, which must be accounted for in order to explain the seasonal cycle of stratosphere-troposphere exchange seen in tracers.

In order to determine the cause of midlatitude ozone depletion, it is essential to quantify the relative role of dynamical effects, chemical destruction, and the short-term impact of aerosol from volcanic eruptions (which complicates the recent record). Purely advective dynamical effects should be capable of quantification, but others are far more complex: for example, the impact of temperature changes on heterogeneous chemistry, or the impact of changes in the frequency of sudden warmings, or in the timing of the final vortex breakdown, on the propagation of polar ozone loss into midlatitudes. Such coupled dynamical-chemical processes are the most difficult to unravel. Since they may well have strong sensitivity to horizontal resolution (i.e., strong scale dependence), they are also extremely difficult to quantify with any confidence.

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