

Dynamics of Transport Processes in the Upper Troposphere

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A number of key problems in atmospheric chemistry are shaped by the strength and character of the various mechanisms acting to move and mix air in the upper troposphere. These transport processes are examined from a mechanistic perspective, with primary emphasis on the tropopause and middle-troposphere regions in the extratropics. The roles of vertical and horizontal transport “barriers” are explored, including the processes by which such barriers are created and are overcome. These transport considerations lead to a hypothesis concerning the processes that shape the tropopause itself. Some perspectives are offered on the still immature subject of transport in the upper troposphere of the tropics.

The upper troposphere is a transition zone separating the distinctly different chemistries of the stratosphere from those of the lower troposphere. This region exchanges air with the stratosphere and the lower troposphere through various atmospheric transport processes. The differing efficiencies of these transport processes determine, to a significant degree, the effectiveness of the photochemical processes that occur there.

The effects of anthropogenic influences on ozone and aerosol concentrations in the upper troposphere are of current interest because of their potential contributions to climate change. For example, observed sharp ozone losses in the lower stratosphere can influence chemical and climate changes in the upper troposphere by means of transport across the tropopause. Ground-level anthropogenic sources of nitrogen oxides and hydrocarbons could play a substantial chemical role in changing the ozone concentrations in the upper troposphere. Also, upper tropospheric effects of particulate and nitrogen oxide emissions from subsonic aircraft are receiving increased research emphasis.

Tools and Methods

Multiple tools are available to study transport processes in the extratropical upper troposphere. Most importantly, this region is rather well sampled, mainly for weather-forecasting purposes. Specifically, measurements of temperature, pressure, water vapor, and horizontal wind speed and direction are available from the international network of

radiosonde balloons, which are launched twice daily over roughly a thousand locations. Ozone is routinely measured by balloon at about 25 stations, roughly once a week. Temperature and water vapor measurements are also available from satellites. Water vapor remains difficult to measure in the upper troposphere from either balloons or satellites. Other chemical measurements are much more sporadic in character. The best upper tropospheric measurements of chemically active substances—such as oxides of nitrogen, oxides of hydrogen, hydrocarbons, and particles—typically are available only from specific field campaigns designed to understand chemical processes. A detailed review of such measurements (1) and their significance is outside the scope of this paper.

Over the past two decades, examination of upper tropospheric transport and chemical behavior through self-consistent mathematical models has become increasingly powerful. These global, three-dimensional (3D) models solve the equations of classical physics (conservation of momentum, heat, and matter) explicitly on computational grids with typical horizontal spacings of 100 to 500 km and vertical spacings of 1 to 2 km. They have been rather successful in providing an objective basis for improved weather forecasting, as well as for increasingly credible simulations of the climate and its variations (2).

Structure of the Upper Troposphere

The upper troposphere is a distinct region, both chemically and dynamically, because of its distance from the very active processes near Earth’s surface and its proximity to the tropopause. The tropopause is the boundary

zone between the troposphere (3) and the stratosphere (4). At times, the tropopause can be marked by a virtual discontinuity in vertical temperature gradient, and frequently is so in the tropics, but at high latitudes, a single specific tropopause point is normally difficult to locate in a vertical profile measurement. The traditional concept of a single tropopause point becomes problematic in regions where active cross-tropopause transport processes are under way.

Understanding the physical basis of the tropopause and its role in transport is central to accurate modeling of upper tropospheric chemical processes. Strong indications can be found in the nearly universal observation that trace constituents with net sources or sinks in the middle stratosphere (for example, ozone and nitrous oxide) exhibit strong vertical gradients above the tropopause and much smaller vertical gradients in the troposphere; that is, sharp changes in vertical gradients are seen, similar to those observed in temperature profiles. This observation leads to the simple inference that vertical transport is much less efficient in the stratosphere than in the troposphere. This assumption is reasonable because the static stability (5) of the stratosphere is much larger than that of the troposphere. In effect, the strong vertical packing of stratospheric surfaces of constant potential temperature (5) (Fig. 1) makes it more difficult for diabatic processes to move air efficiently across such surfaces.

Transport Across the Tropopause

The earliest attempt toward understanding cross-tropopause transport was made nearly a half century ago. The discovery that the stratosphere is more than three orders of magnitude drier than the lower troposphere led to the hypothesis of an overturning circulation with rising motion in the tropics and sinking in higher latitudes (6). Such a circulation forces the air entering the stratosphere from below to be “cold trapped” to extreme dryness (~3 parts per million by volume) by its passing through the very cold (−80° to −85°C) tropical tropopause. This qualitative observation remains valid today.

In the 1950s and 1960s, it was discovered that substantial extrusions of stratospheric air into the mid-latitude troposphere occur quasi-adiabatically (5), under near conservation of potential vorticity (7), in association with extratropical cyclones (large-scale weather systems) (8–11). Later, it was determined that this extrusion process is directly related to the unstable growth of extratropical cyclones (commonly termed as cyclogenesis) (12).

These early studies led to the general

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assumption that the observed seasonal variation of surface radioactive fallout (resulting from detonations of nuclear weapons that initially deposit their radioactive debris into the stratosphere) is related to the seasonal variation in the intensity of extratropical cyclones (13). Later it was found that the cyclogenesis process is active throughout the year in its movement of lower stratospheric air into the troposphere, thus leading to the assertion that the tropospheric seasonal radioactivity cycle and its spring peak depend mainly on the seasonal variations in the extratropical downward mass flux to the vicinity of the tropopause (14). Other analyses have indicated that the seasonal variation of the flux of stratospheric air into the troposphere also depends on the seasonal variation of the mass of the stratosphere; in effect, it depends on the altitude of the tropopause (10, 15). It is not yet clear whether these two mechanisms are physically distinct.

Understanding of the nature of this seasonal effect was greatly enhanced by the realization that the stratospheric overturning is essentially driven by zonal momentum forcing (strongest in the winter), which is produced by atmospheric disturbances that propagate upward from the troposphere, dissipate, and thus act to decelerate the strong eastward winds (16). For reviews and explanations of these processes and their implications, see (17, 18). More recently, it has been demonstrated that a higher resolution atmospheric circulation model (≈ 100 -km horizontal grid length) can generate the dynamical processes required to simulate the strong downward mass flux in the region above and through the higher latitude tropopause in mid- and late winter (19). Also, such models can simulate the dynamics of tropospheric ex-

tratropical cyclone growth well enough to provide a roughly correct irreversible flux of air into the troposphere in response to such events. That is, the flux estimates per event in (8–12) agree reasonably well with those of similar events in the GFDL (Geophysical Fluid Dynamics Laboratory) SKYHI model (Fig. 2). This correspondence appears to be valid, although the finest structural details of such extrusions into the troposphere (9, 20) cannot yet be adequately simulated in even today's higher resolution global models. Achieving such detailed simulations would require models with horizontal grid spacings smaller than about 60 km and vertical grid spacings of less than about 100 m.

In effect, the downward exchange of air into the extratropical upper troposphere can be regarded as the end result of overcoming two barriers. First, in the stratosphere, the potential-temperature barriers discussed in the previous section are circumvented by momentum forcing that decelerates the winter season's strong eastward flow, induces a north-south overturning circulation (Fig. 1) that warms the higher latitudes mainly by adiabatic compression, and excites radiatively induced diabatic cooling. These effects produce a net mean downward diabatic mass flux across surfaces of constant potential temperature (16) toward and through the higher latitude tropopause. Second, along the potential-temperature surfaces that strongly intersect the tropopause region (Fig. 1), the north-south gradients of potential vorticity are strongly concentrated near the meandering mid-latitude jet stream. A region of concentrated potential-vorticity gradient on a potential-temperature surface is relatively stable to north-south displacements of air on that surface (17). This region thus provides a barrier to quasi-horizontal

equatorward and downward movement of stratospheric air along the potential-temperature surfaces that dip into the troposphere (Fig. 1). Moreover, this same barrier tends to impede the poleward and upward movement of tropospheric air along the same potential-temperature surfaces from tropospheric lower latitudes into the stratosphere. However, the tropospheric cyclogenesis process provides the key mechanism to overcome this final barrier to north-south and vertical transport, at least sporadically. In effect, strong cyclogenesis produces enough unstable amplification of tropospheric cyclone waves that they "break," thus inducing irreversible, quasi-adiabatic north-south and vertical mixing of potential vorticity and accompanying trace constituents (Fig. 2).

If a 3D atmospheric model simulates these large-scale unstable disturbances with sufficient accuracy, it is probable that its simulation of the north-south and vertical mass exchanges will be adequate. Most higher resolution 3D global models appear to simulate these processes reasonably well in comparison to analyses of data from the global weather-observing system. However, a clear link of such successful model simulations to accurate transport calculations is difficult because of the relative scarcity of available trace-constituent data. In fact, it is almost unthinkable that global observational data could be achieved on the spatial and temporal scales illustrated in Fig. 2. Thus, evaluation of the quantitative skill of 3D atmospheric models must depend on limited data from a small number of focused field experiments and on long-term measurements of trace constituents, wind, pressure, and temperature structure from a relatively limited number of locations.

Tropopause and Jet-Stream Transport Barriers

The above arguments address tropopause-level transport barriers and how they are broken but do not address the more fundamental questions concerning how the barriers to transport across the tropopause and the jet stream got there in the first place. As noted earlier (3), the tropopause exists mainly because of the convective processes excited by the destabilizing effect of solar heating, which occurs mainly at Earth's surface. The tropopause is thus approximately determined by the "capping" of the convection zone encountering the stably stratified stratosphere, which is heated from above.

A problem of detail with the above argument is that simple radiative-convective models of the atmosphere (21) predict a tropopause structure that is noticeably different from the observed. The observed tropopause has an average height of about

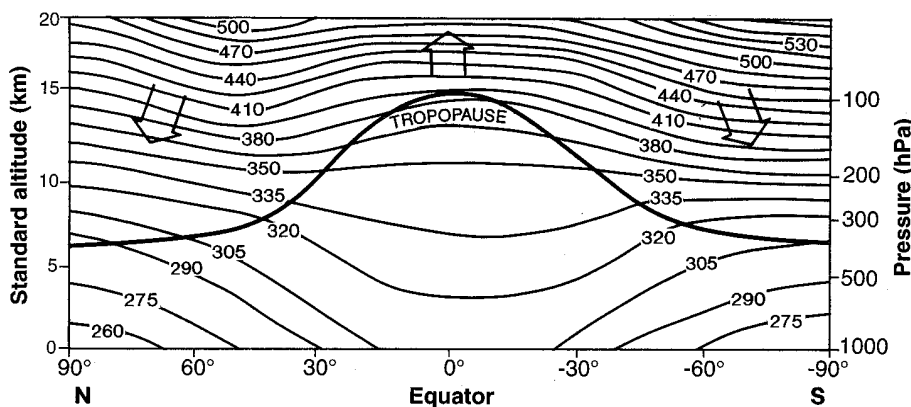


Fig. 1. Latitude-height distribution of mean potential temperature (in kelvin) for the month of January (5), averaged over longitude (thin lines). The mean position of the tropopause, which marks the troposphere-stratosphere boundary, is indicated by the thick, poleward-downward sloping line. The vertical spacing between lines of potential temperature is much closer in the stratosphere (above the tropopause). Note that many lines of potential temperature intersect the tropopause. The arrows denote the sense of the north-south overturning circulation in the stratosphere.

15 km in the tropics and a height of roughly 8 to 10 km at higher latitudes; the tropopause predicted by simple radiative-convective models has a tropical height of about 14 km (with temperatures of about -60°C , which is too warm) and rather low, indistinct heights at higher latitudes (22). These differences in structure and temperature are qualitatively consistent with the hypothesis that the mass fluxes across the tropopause are mainly determined by a process dubbed “downward control” (23), in which the integrated zonal momentum forcing of the eastward winds above the tropopause determines the magnitude of net upward mass flux in the tropics and the net downward mass flux at higher latitudes. In effect, the dynamically forced mean rising motion through the tropical tropopause cools the region further and elevates the tropopause somewhat higher than it would have been in the absence of this process. This theoretical prediction is still regarded as controversial by many atmospheric scientists, because the mean rising motion within the tropical troposphere (the “Hadley” circulation) is attributable to dynamical and thermodynamical processes acting within the troposphere itself. The tropical tropopause thus appears to be a boundary zone marking the transition from tropospheric to stratospheric “control” of the mean upward air movement through it. It therefore is not surprising that the specific dynamical determinants of the observed cold tropical tropopause temperatures remain unresolved.

At extratropical latitudes, the tropopause height is not adequately defined by radiative-convective arguments (21) alone. The vertical mixing processes induced by unstable extratropical cyclones may play a significant role in raising the higher latitude tropopause to heights significantly above the low values predicted by simple radiative-convective models (24). However, there appears to be another key piece to the story. For over three decades, piecemeal measurements in the mid-latitude lower stratosphere have revealed that the north-south slopes of quasi-conservative-trace-constituent surfaces strongly parallel the north-south slope of the tropopause itself (Fig. 1), even as high as 10 km above the tropopause. Moreover, these trace-constituent slopes are nearly parallel to the north-south potential-vorticity (7) slopes in this region. The equilibrium north-south slopes of stratospheric tracer isolines result from a balance between the downward (upward) diabatic mass flux in higher (lower) latitudes (a slope-steepening effect) and the quasi-adiabatic north-south mixing induced by cyclone-scale and planetary-scale waves propagating upward from the troposphere (a slope-flattening effect) (25). It is thus hy-

pothesized here that the north-south slope of the extratropical tropopause may be controlled, to a first approximation, by these same dynamically driven transport processes that so strongly shape the higher latitude lower stratosphere. The higher latitude tropopause appears to be “forced downward,” especially during the winter, to a lower altitude than it would be in the absence of such stratospheric forcing. This hypothesis is consistent with the calculation of radiative cooling rates in the winter season that are quite small, in the sense that the radiative relaxation times are roughly

on the same time scale as that of the winter season itself. Thus, the potential-temperature surfaces can be “pushed downward” by the stronger downward control mechanism operative during the winter season.

The net downward flux of stratospheric trace constituents tends to “pile up” in the higher latitude lower stratosphere, finally constrained by the remaining potential-temperature and potential-vorticity barriers. The remaining question is concerned with why the tropopause-level potential-vorticity (jet stream) barrier is so strong. Some previous studies suggest that the un-

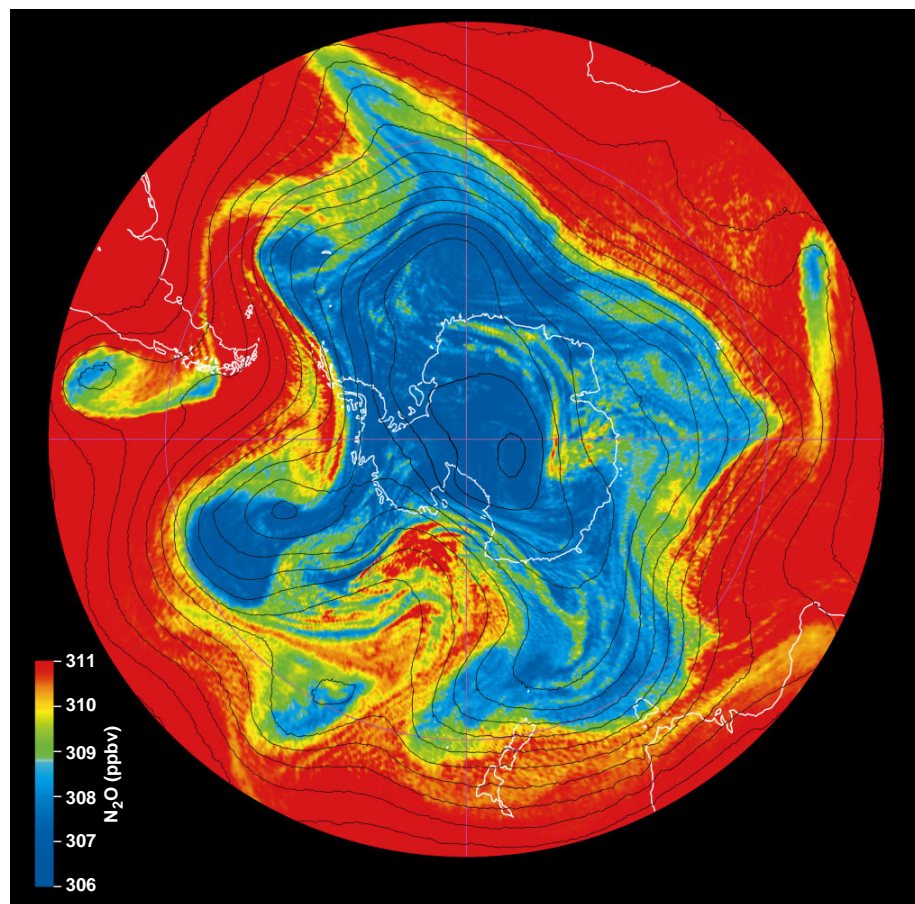


Fig. 2. Plot of nitrous oxide (30) mixing ratio in the Southern Hemisphere on the 320-K potential-temperature (5) surface for model date 13 June, 00 GMT. Units are in parts per billion by volume. Continental boundaries are indicated in white. The thin black lines are the Montgomery stream function ($M = c_p T + gZ$, where g is the acceleration of gravity and Z is geopotential height) (contour interval is $1000 \text{ m}^2/\text{s}^2$). The balanced (geostrophic) wind speed is proportional to the gradient of M divided by the vorticity of Earth’s solid-body rotation; closely spaced M lines thus indicate the locations of jet-stream winds and of cyclonic and anticyclonic vorticity (7). Data are from an experimental fine-mesh ($1/3^{\circ}$ -latitude horizontal grid spacing) version of the NOAA GFDL 40-level SKYHI atmospheric circulation model (32). The 320-K potential-temperature surface intersects the tropopause in mid-latitudes and the 4-km level in the tropics. The sharp gradient of nitrous oxide (blue edge) marks the meandering jet stream and pronounced potential-vorticity (7) transport barrier at the tropopause. Unstable amplification of cyclone-scale waves produces large quasi-adiabatic equatorward and downward excursions of (blue) stratospheric air that “cut off,” a portion of which irreversibly mixes into the mid-latitude and subtropical troposphere. Because of the dynamically driven downward mass flux across potential-temperature surfaces in the high-latitude lower stratosphere, poleward of the jet-stream axis, the net transport of (red and yellow) tropospheric air into the lower stratosphere as a result of the two processes together is considerably less efficient. Note, however, the thin “streamers” of (yellow and green) tropospheric N_2O values well within the (blue) stratospheric region.

stable cyclonic growth process at extratropical latitudes produces a north-south "stirring" of potential vorticity that, somewhat counterintuitively, concentrates the remaining north-south potential vorticity gradient into a rather narrow band (26). This stirring process arguably leads to the very existence of the sharp, meandering, and intermittent mid-latitude jet stream system (Fig. 2) that separates the transient cyclones and anticyclones so familiar to viewers of television weather forecasts.

Exchange with the Lower Troposphere

The above conclusions lead to an assertion that the transport of trace constituents by the mean diabatic mass flux at higher latitudes and by quasi-adiabatic mixing by the large-scale cyclonic and anticyclonic disturbances of the mid-latitude upper troposphere and lower stratosphere is reasonably well understood. Higher-resolution, comprehensive atmospheric models probably simulate the transport processes there adequately, say within roughly $\pm 30\%$ of that of the real world.

Within the mid-latitude troposphere, these same large-scale "weather" disturbances produce effective north-south lifting and sinking of air along surfaces of constant potential temperature that slope sharply poleward and upward (Fig. 1). This process produces relatively efficient north-south and vertical mixing throughout the extratropical troposphere. Moreover, for air parcels that ascend from the lower troposphere, move poleward, and cool nearly adiabatically, water vapor saturation and a concomitant release of latent heat of condensation is virtually inevitable. This process produces a net upward flux of heat that works against the mean vertical gradient of potential temperature and adds to the effectiveness of the tropospheric vertical mixing. These efficient unstable weather processes apparently constitute the dominant vertical and north-south transport mechanisms in the mid-latitude troposphere above the surface boundary layer and throughout the troposphere. This hypothesis is generally consistent with the relative success of global 3D models in simulating such transport and the premise for their widespread use (1, 27). Other mid-latitude processes such as isolated small-scale moist convective events are likely to be of smaller magnitude. However, such moist convective processes may be locally important, almost certainly so in the summertime, when small-scale convective storms are stronger and the north-south temperature gradient (the main energy source for unstable extratropical cyclonic disturbances) is weaker.

A current leading topic of discussion in the tropospheric chemistry community focuses on the degree to which chemicals released near Earth's surface influence the chemistry of ozone in the upper troposphere (1). Models that add small-scale convective mechanisms designed to move air even more efficiently through the mid- and higher latitude middle troposphere do indeed calculate a more substantial influence of surface sources on the chemistry of the upper troposphere (1). A question remains, however, as to whether or not such aggressive prescribed vertical mixing is correct. The obvious approach to determining what can be considered correct would be to use measurements of an atmospheric trace constituent with known sources and sinks to evaluate the various model performances. Unfortunately, there appear to be no trace constituents with known sources and sinks for which sufficient measurements exist to provide quantitative tests of 3D transport models (1); therefore, carefully considered extratropical tropospheric measurements of the vertical and horizontal structure of appropriately chosen trace constituents should be given a high research priority.

Transport in the Tropical Upper Troposphere

Transport in the upper troposphere of the tropics is currently receiving increased attention, although the subject is still in a relatively immature state. The exchanges of tropical upper troposphere air with that from higher latitudes of both hemispheres and with air from the lower troposphere are relevant to some key atmospheric chemistry problems. Most significant is the ozone-generating role of nitric oxides produced by lightning and by tropical biomass burning. Also, the role of interhemispheric exchange in solving the "missing sink" problem of anthropogenic carbon dioxide (28) is being actively pursued. The 3D models indicate that the dominant mechanisms for exchanging air between the Northern and Southern hemispheres are likely to be in the tropical upper troposphere (28, 29).

The subject of transport within and across the tropical upper troposphere is less scientifically mature than that of the extratropics and is thus discussed only minimally here. Eventually, however, improved fundamental understanding of transport in this region will have to deal with low-latitude transport barriers and how they are circumvented.

The connection between the upper and lower troposphere in the equatorial region is fast and efficient because of the dominant role of moist convection. In the subtropics, however, such processes become quite inef-

fective. It is probable that the slow processes related to the mean sinking motion associated with the north-south overturning Hadley circulation will begin to dominate there. Also, the east-west overturning associated with the Walker circulation is likely to shape tropical transport in significant ways. It is likely to be true as well for the now well-known El Niño Southern Oscillation (2- to 6-year time scale).

As in the extratropics, north-south exchange of trace constituents, essentially along potential-temperature surfaces, is subject to the constraints of north-south gradients of potential vorticity (7). Because of the opposite sense of rotation in the Northern and Southern hemispheres, the potential vorticities of the two hemispheres are of opposite sign. Thus, large nonconservative processes are fundamentally required to produce net interhemispheric exchange. These constraints necessarily limit the efficiency of the exchange process. In addition, the subtropical jet streams, located at roughly 30°N and 30°S latitudes in the winter seasons, are also locations of concentrated north-south gradients of potential vorticity. Consequently, they are significant barriers to upper tropospheric transport between the tropics and the extratropics.

It has been empirically established that the time scale of exchange from the northern to the southern extratropics is roughly one year (28). Leading candidates for the mechanisms that allow these barriers to be crossed are the Hadley circulation and its seasonal variations, the Asian summer and winter monsoon circulations, low-latitude incursions of extratropical cyclonic disturbances (Fig. 2), and transient weather disturbances throughout the tropics.

Interestingly, the two GFDL 3D models (29, 30) devoted to transport studies performed reasonably well in recent (yet unpublished) experiments designed to evaluate interhemispheric exchange for application to the carbon-cycle problem. Unfortunately, the early level of analysis of the model experiments and the low quality of available measurements of trace constituents in the tropics allow only a qualitative understanding of the operative transport process in the tropical upper troposphere. The availability of better field measurements of key trace constituents within the tropics could markedly improve the rudimentary understanding of transport mechanisms within this region.

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3. The troposphere is the part of the atmosphere that is generally characterized by sharply decreasing temperatures with increasing altitude, roughly 6°C/km. Its thickness ranges from roughly 8 km in higher latitudes to 16 km in the tropics. Because the cloudless atmosphere is nearly transparent to incoming solar radiation, the troposphere is essentially heated from below, with dynamically induced vertical mixing shaping the vertical temperature structure. The upper limit of the troposphere is conventionally called the tropopause.
 4. The stratosphere begins at the tropopause and extends to an altitude of about 50 km. Its temperature slowly increases with altitude, mainly resulting from the absorption of solar ultraviolet radiation by ozone.
 5. Under adiabatic conditions (no addition or subtraction of heat), an air parcel conserves its entropy, or equivalently its potential temperature, as it undergoes expansion or compression. From direct manipulation of the first law of thermodynamics, potential temperature is defined as $\Theta = T(P_0/P)^\kappa$, where T is temperature, P is pressure, P_0 is a reference pressure at Earth's surface, $\kappa = R/mc_p$, R is the universal gas constant, m is the molecular weight of dry air, and c_p is the specific heat of air at constant pressure. Under these idealized circumstances, the first law of thermodynamics adopts the simple form $d\Theta/dt = 0$; that is, potential temperature is conserved following an air parcel over time t . In the atmosphere, Θ generally increases with altitude, markedly so in the stratosphere. Long-term transport between the troposphere and stratosphere must be tightly constrained by the magnitude of net diabatic processes, ultimately of dynamical origin, moving air across surfaces of constant potential temperature. Static stability is defined here for convenience as the vertical pressure gradient of potential temperature ($-\partial\Theta/\partial P$) and is a measure of the resistance of air to vertical displacement. Use of potential temperature as a vertical coordinate allows straightforward use of the radiosonde-based meteorological network of wind, temperature, and pressure data to calculate 3D air trajectories in the troposphere, to the extent that the air moves nearly adiabatically. This substitution is a powerful diagnostic tool for addressing tracer movements, because it evades the problem that standard meteorological measurements do not measure vertical velocity directly (8–11).
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Throughout the competition period, readers are invited to nominate papers appearing in the Reports, Research Articles, or Articles sections. Nominations must be typed, and the following information provided: the title of the paper, issue in which it was published, author's name, and a brief statement of justification for nomination. Nominations should be submitted to the AAAS–Newcomb Cleveland Prize, AAAS, Room 1044, 1200 New York Avenue, NW, Washington, DC 20005, and **must be received on or before 30 June 1997**. Final selection will rest with a panel of distinguished scientists appointed by the editor-in-chief of *Science*.

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