# Contributions of Anthropogenic and Natural Forcing to Recent Tropopause Height Changes

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Observations indicate that the height of the tropopause—the boundary between the stratosphere and troposphere—has increased by several hundred meters since 1979. Comparable increases are evident in climate model experiments. The latter show that human-induced changes in ozone and well-mixed greenhouse gases account for  $\sim\!80\%$  of the simulated rise in tropopause height over 1979—1999. Their primary contributions are through cooling of the stratosphere (caused by ozone) and warming of the troposphere (caused by well-mixed greenhouse gases). A model-predicted fingerprint of tropopause height changes is statistically detectable in two different observational ("reanalysis") data sets. This positive detection result allows us to attribute overall tropopause height changes to a combination of anthropogenic and natural external forcings, with the anthropogenic component predominating.

The tropopause represents the boundary between the troposphere and stratosphere and is marked by large changes in the thermal, dynamical, and chemical structure of the atmosphere (1-3). Increases in tropopause height over the past several decades have been identified in radiosonde data (2, 4), in optimal combinations of numerical weather forecasts and observations ("reanalyses") (3, 5), and in climate models forced by combined natural and anthropogenic effects (6). Model experiments suggest that such increases cannot be explained by natural climate variability alone (6, 7).

To date, no study has quantified the contributions of different anthropogenic and natural forcings to 20th-century tropopause height changes. We estimate these contributions here and demonstrate the usefulness of the thermally defined tropopause as an integrated indicator of human-induced climate change. We also identify a model-predicted "fingerprint" of tropopause height changes in reanalysis data. Detection of this fingerprint enables us to attribute tropopause height changes to the combined effects of anthropogenic and natural forcing.

The anthropogenic forcings we consider are changes in well-mixed greenhouse gases (G), the direct scattering effects of sulfate aerosols (A), and tropospheric and stratospheric ozone (O). The natural forcings considered are changes in solar irradiance (S) and volcanic aerosols (V). All of these factors are likely to have modified the thermal structure and static stability of the atmosphere (6-11), thus affecting tropopause height. To isolate the thermal responses that drive tropopause height changes, we analyze the effects of G, A, O, S, and V on temperatures averaged over broad layers of the stratosphere and troposphere.

Model and reanalysis data. We used the Department of Energy Parallel Climate Model (PCM) developed by the National Center for Atmospheric Research (NCAR) and Los Alamos National Laboratory (12). A wide range of experiments has been performed with PCM, of which seven were analyzed here for tropopause height changes. In the first five experiments, only a single forcing is changed at a time; for example, G varies according to historical estimates of greenhouse gas concentration changes, whereas A, O, S, and V are all held constant at preindustrial levels (13-18). In the sixth experiment (ALL), all five forcings are varied simultaneously. Both natural forcings are changed in the seventh experiment (SV). G, A, O, and S commence in 1872, whereas V, SV, and ALL start in 1890. The experiments end in 1999. To obtain better estimates of the underlying responses to the imposed forcings, four realizations of each experiment were performed. Each realization of a given experiment has identical forcing but commences from different initial conditions of an unforced control run.

The detection part of our study requires estimates of observed tropopause height changes, which were obtained from two reanalyses (19, 20). The first is from the National Center for Environmental Prediction and the National Center for Atmospheric Research (NCEP); the second is from the European Centre for Medium-Range Weather Forecasts (ERA). Reanalyses use an atmospheric numerical weather forecast model with no changes over time in either the model itself or in the observational data assimilation system. NCEP data were available from 1948 to 2001, but data before January 1979 were excluded because of well-documented inhomogeneities (21, 22). ERA spans the shorter period from 1979 to 1993.

We estimate tropopause height using a standard thermal definition of  $p_{\rm LRT}$ , the pressure of the lapse-rate tropopause (23). This definition has the advantage that it can be applied globally and is easily calculated from vertical profiles of atmospheric temperature under most meteorological conditions (24, 25). The algorithm that we used to compute  $p_{\rm LRT}$  primarily monitors large-scale changes in the thermal structure of the atmosphere.

Multidecadal changes in  $p_{\mathrm{LRT}}$  are generally smaller than the vertical resolution of the atmospheric models used in PCM and the two reanalyses (Fig. 1A). This raises concerns regarding the sensitivity of our estimated  $p_{\rm LRT}$  trends to the vertical resolution of the input temperature data in the vicinity of the tropopause. Three independent pieces of evidence support the reliability of our  $p_{LRT}$ changes. First, calculations of NCEP  $p_{\mathrm{LRT}}$ trends performed at high and low vertical resolution are in close agreement (26). Second, climate models with different vertical resolutions yield similar  $p_{LRT}$  trends in response to external forcing (6). Finally,  $p_{\rm LRT}$ trends computed from ERA and NCEP are in good agreement with  $p_{LRT}$  changes obtained from high-resolution radiosonde temperature soundings (4, 6). All three lines of evidence enhance confidence in the robustness of our procedure for estimating  $p_{LRT}$  changes.

At each grid point in the model and reanalysis data, we compute  $p_{\rm LRT}$  from the monthly mean temperature profile at discrete pressure levels (25). For PCM, we also calculate stratospheric and tropospheric temperatures equivalent to those monitored by channels 4 and 2 (T4 and T2) of the satellite-based Microwave Sounding Unit (MSU) (22, 27–29). The simulated MSU data are useful in relating changes in  $p_{\rm LRT}$  to broad changes in atmospheric thermal structure. Direct comparisons with observed MSU T4 and T2 data are given elsewhere (30).

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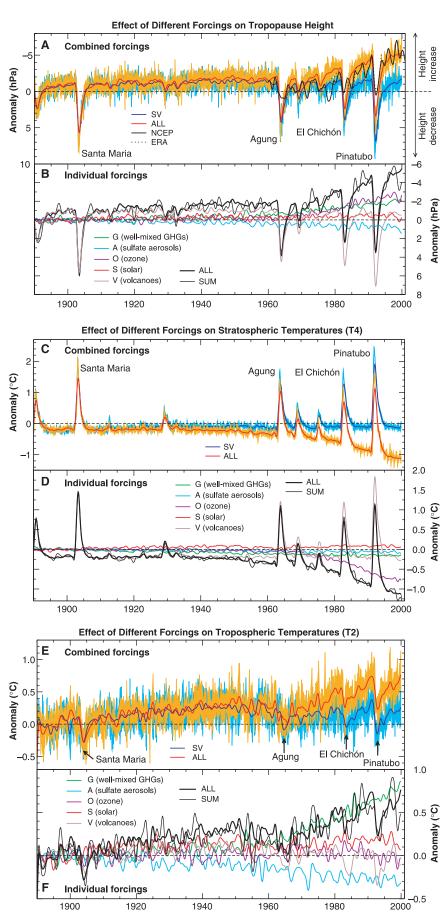


Fig. 1. Time series of global mean, monthly mean anomalies in tropopause pressure ( $\rho_{\rm LRT}$ )(A and B), stratospheric temperature (T4) (C and D), and mid- to upper tropospheric temperature (T2) (E and F). Model results are from seven different PCM ensemble experiments (12, 14, 16, 17). Five experiments use a single forcing only (G, A, O, S, or V). Two integrations involve combined forcing changes, either in natural forcings (SV), or in all forcings (ALL). There are four realizations of each experiment. In (B), (D), and (F), only low-pass filtered ensemble means are shown. In (A), (C), and (E), both the low-pass filtered ensemble mean and the (unfiltered) range between the highest and lowest values of the realizations are given. All model anomalies are defined relative to climatological monthly means computed over 1890–1909. Reanalysis-based  $\rho_{LRT}$ estimates from NCEP (19) and ERA (20) were filtered in the same way as model data (A). NCEP  $p_{LRT}$  data are available from 1948-2001, but pre-1960 data were ignored because of deficiencies in the coverage and quality of assimilated radiosonde data (22). The ERA record spans 1979-1993. NCEP (ERA) was forced to have the same mean as ALL over 1960-1999 (1979-1993). The SUM results [(B), (D), and (F)] are the sum of the filtered ensemble-mean responses from G, A, O, S, and V.

Global-scale changes. For any specified forcing, the ensemble members define an envelope of possible climate trajectories (Fig. 1A). The time at which the ALL and SV envelopes separate completely (and remain separated) is a simple qualitative measure of the detectability of an anthropogenic signal in PCM data. For tropopause height, this separation occurs in the mid-1980s. Divergence of the ALL and SV solutions occurs earlier for T4 data [before the eruption of Mt. Agung in 1963 (Fig. 1C)] and later for T2 [in the early 1990s (Fig. 1E)].

The short-term (3- to 4-year)  $p_{LRT}$  signatures of major volcanic eruptions are clearly evident in reanalyses and the SV and ALL experiments (Fig. 1A). Large explosive volcanic eruptions warm the lower stratosphere and cool the troposphere (Fig. 1, C and E). Both effects decrease tropopause height and increase  $p_{\rm LRT}$  (6). In PCM,  $p_{\rm LRT}$  changes after the eruptions of Santa Maria, Agung, El Chichón, and Pinatubo are large, relative to both the "between realization" variability of  $p_{\rm LRT}$  and the  $p_{\rm LRT}$  variability during volcanically quiescent periods.

The total linear  $p_{LRT}$  decrease in ALL is 2.9 hPa over 1979-1999, corresponding to a tropopause height increase of roughly 120 m (Fig. 1A). Tropopause height in NCEP rises by  $\sim$ 190 m. The smaller increase in PCM is primarily due to two factors. The first is the unrealistically large stratospheric cooling in NCEP (31). Because stratospheric cooling tends to increase tropopause height (4, 6), excessive cooling amplifies NCEP's height increase. The second factor is PCM's overestimate of volcanically induced stratospheric warming (30). Excessive stratospheric warming yields a volcanically induced tropopause height decrease that is too large, thus reducing the overall height increase in ALL.

The largest contributions to PCM's overall tropopause height increases are from changes in

1900

1920

1940

1960

1980

well-mixed greenhouse gases and ozone (Fig. 1B). To quantify these contributions, we examine the total linear  $p_{\rm LRT}$  trends in the ensemble means of the G, A, O, S, and V experiments (32). Linear trends are computed over four different time horizons: the 20th century, 1900-1949, 1950-1999, and the satellite era (1979-1999) (Fig. 2A). Contributions of individual forcings are expressed as a percentage of the total linear change in ALL (33). Because some forcings increase tropopause height and others lower it, the contributions of individual forcings can in some cases exceed 100%. This simply means that in the absence of other influences, the trend due to a specific forcing can be larger than that obtained when all five forcings act in

During the 20th century, G and O account for 77 and 88% (respectively) of the total increase in tropopause height in ALL. Their combined percentage contribution of 165% is partly offset by the smaller decreases in height caused by A and V (-32 and -39%). Solar irradiance changes make a small positive contribution to the height increase in ALL. The relative contribution of each forcing varies with time. Whereas G and O make roughly equivalent contributions to tropopause height change over 1900-1949, ozone becomes more important in the second half of the century and during the satellite era (Fig. 2A). Similarly, S and V together explain 74% of the total tropopause height change over 1900-1949, but their net contribution to ALL is only -40% over 1950-1999. The large influence of V over 1900-1949 arises from the Santa Maria eruption in 1902 (Fig. 1A).

Decadal-scale increases in the height of the tropopause are driven by temperature changes above and below the tropopause. Temperatures in this region are influenced primarily by ozoneand greenhouse gas-induced cooling of the stratosphere and greenhouse gas-induced warming of the troposphere. Both of these effects tend to raise tropopause height (4, 6). To first order, the global mean changes in  $p_{IRT}$  over a stipulated time period are linearly related to  $\Delta T4$  –  $\Delta$ T2. A key question, therefore, is whether the simulated height increase in ALL could have occurred solely through stratospheric cooling and without significant anthropogenically induced warming of the troposphere. We address this question by estimating the contributions of individual forcings to T4 and T2 changes.

Previous model-based work suggests that ozone forcing is the major driver of recent stratospheric cooling (10, 34). A similar result holds in PCM: Ozone changes account for 123% of the total linear change in T4 over 1950–1999 and for 82% of the T4 decrease during the satellite era (Figs. 1D and 2B). The corresponding contributions of greenhouse gas forcing to T4 changes are only 9 and 3%, respectively (6). In contrast, well-mixed greenhouse gases are the major contributor to PCM's tropospheric warming and explain 174% of the total change in T2

over 1950–1999 (Figs. 1F and 2C). The ozone component of T2 changes is small over this period (-6%) (35). In PCM, therefore, the main effect of well-mixed greenhouse gases on tropopause height is through warming the troposphere rather than cooling the stratosphere.

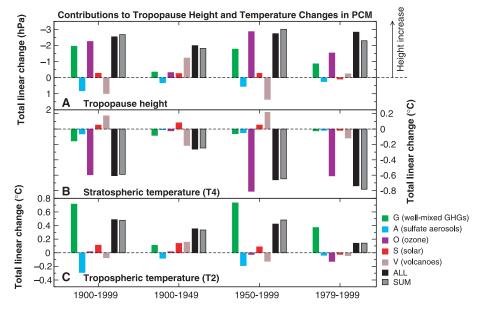
Several additional features are noteworthy. Anthropogenic sulfate aerosols decrease tropopause height, primarily by cooling the troposphere (Figs. 1F and 2C). In the model, sulfate aerosols cause only a small decrease in T4 (Figs. 1D and 2B). Solar irradiance changes over the 20th century warm both the troposphere and the stratosphere, with offsetting effects on tropopause height. The sign of the solar effect on  $p_{\rm LRT}$  must therefore depend on the relative magnitudes of solar-induced stratospheric and tropospheric warming. The small rise in tropopause height in S (Fig. 1B) suggests that for solar forcing, tropospheric warming is more important.

A key assumption in many detection studies is that the sum of the individual climate responses to several different forcing mechanisms is equal to the response obtained when these forcings are varied simultaneously (8, 9, 36). This implies that there are no strong interactions between individual forcings. We tested this assumption for  $p_{LRT}$ , T4, and T2 by comparing ALL results with the sum of the individual responses to G, A, O, S, and V (SUM). ALL and SUM show very similar global mean changes (Fig. 1, B, D, and F). For the three variables considered here, the estimated linear changes in SUM are within 10% (37) of the corresponding ALL values (Fig. 2). For these global-scale changes, additivity is a reasonable assumption.

Fingerprint analysis. We next used a standard detection method (38, 39) to determine whether a model-predicted spatial pattern of externally forced  $p_{LRT}$  changes can be identified in reanalysis data. Our detection method assumes that the searched-for signal (the fingerprint,  $\vec{f}$ ) is well represented by the first empirical orthogonal function (EOF) of the ALL ensemble mean (30). We search for an increasing expression of  $\vec{f}$  in the NCEP and ERA  $p_{\mathrm{LRT}}$  data and estimate the detection time—the time at which  $\vec{f}$  becomes consistently identifiable at a stipulated 5% significance level. The climate noise estimates required for assessing statistical significance are obtained from 300year control runs performed with PCM and the ECHAM model of the Max-Planck Institute for Meteorology in Hamburg (40).

Detection times are computed both with and without the global mean component of tropopause height change (30). If the spatial mean is included, detection results can be dominated by large global-scale changes. Removing the spatial mean focuses attention on smaller-scale model-data pattern similarities and provides a more stringent test of model performance.

We first describe the patterns of tropopause height change in PCM and reanalyses. Maps of linear  $p_{\rm LRT}$  trends over 1979–1999 are similar in NCEP and PCM (Fig. 3, A and C). Both show spatially coherent increases in tropopause height (negative trends in  $p_{\rm LRT}$ ), with the smallest changes in the tropics and the largest increases toward the poles, particularly in the Southern Hemisphere (6). Tropopause height changes in ERA (Fig. 3B) are less coherent than



**Fig. 2.** Total linear changes (32) in global mean, monthly mean tropopause height (A), stratospheric temperature (B), and tropospheric temperature (C) in PCM experiments with individual forcings (G, A, O, S, and V) and combined natural and anthropogenic forcings (ALL). Linear changes are computed over four different time intervals using the (unfiltered) ensemble-mean data from Fig. 1. For each time period, anomalies were defined relative to climatological monthly means computed over 1900–1999. SUM denotes the sum of the linear changes in G, A, O, S, and V.

in NCEP or PCM, with slight decreases in height in the tropics. This largely reflects the shorter record length of ERA, which ends at a time when Pinatubo decreased tropopause height (Fig. 1, A and B). The ALL "mean included"  $p_{LRT}$  fingerprint is similar to the PCM and NCEP linear trend patterns, with uniform sign, largest loadings at high latitudes in the Southern Hemisphere, and strong zonal structure (Fig. 3D). Removal of the spatial means emphasizes the strong equator-to-pole gradients and hemispheric asymmetry of the fingerprint (Fig. 3E).

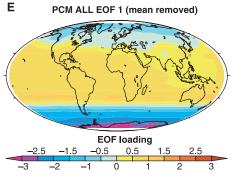
When our detection method is applied with spatial means included, the ALL tropopause height fingerprint is consistently identifiable in both reanalyses (fig. S1). Although ERA tropopause height increases are less coherent than in NCEP and are visually less similar to the ALL fingerprint,  $\vec{f}$  is still readily detectable. In all four cases considered (41), fingerprint detection occurs in 1988, only 10 years after the start of our analysis period (1979). With our strategy, 1988 is the earliest time at which detection can be achieved.

The removal of spatial means still yields an identifiable fingerprint, but only in NCEP. This positive result arises from model-data similarities in the equator-to-pole gradient and the hemispheric asymmetry of tropopause height changes (Fig. 3, A and E). Detection of  $\vec{f}$  in NCEP data occurs in 1995 (fig. S1), 7 years later than in the "mean included" case and after the end of the ERA record. This suggests that the ERA record is simply too short to achieve positive detection of subglobal features of the predicted tropopause height changes.

Conclusions. Our results are relevant to the issue of whether the "real-world" troposphere has warmed during the satellite era. PCM provides both direct and indirect evidence in support of a warming troposphere. The direct evidence is that in the ALL experiment, the troposphere warms by 0.07°C/ decade over 1979-1999 (30). This warming is predominantly due to increases in wellmixed greenhouse gases (Fig. 2C). We have previously shown (30) that the T2 fingerprint estimated from ALL is identifiable in a sat-

Α В NCEP linear trend (1979-1999) ERA linear trend (1979-1993) Linear trend (hPa/decade) Linear trend (hPa/decade) -6 C D PCM ALL linear trend (1979-1999) PCM ALL EOF 1 (mean included) Linear trend (hPa/decade) **EOF** loading -0.9 -1.8-1.2 -0.61.8 Ε PCM ALL EOF 1 (mean removed)

Fig. 3. Least-squares linear trends and fingerprints of tropopause height change. Trends in global mean, monthly mean  $p_{LRT}$  data (in hPa per decade) were computed over 1979–1999 for NCEP (A) and the PCM ALL experiment (C) and over 1979-1993 for ERA (B). The PIRT climate change fingerprints were computed from the ensemble-mean ALL results (D and E). Results are shown for analyses with the spatial mean included (D) and with the mean removed (E).



ellite data set with a warming troposphere (27) but not in a satellite data set with little overall tropospheric temperature change (28).

The second (and more indirect) line of evidence relies on the relation between changes in tropopause height and changes in tropospheric temperature. Our detection results show consistency between the patterns of tropopause height increase in ALL and reanalyses. The PCM individual forcing experiments help to identify the main drivers of this change. Over 1979-1999, roughly 30% of the increase in tropopause height in ALL is explained by greenhouse gas-induced warming of the troposphere (Fig. 2A). Anthropogenically driven tropospheric warming is therefore an important factor in explaining modeled changes in tropopause height. Without this tropospheric warming effect in the model, simulated height changes would be markedly reduced, and the correspondence that we find between modeled and observed  $\Delta p_{\mathrm{LRT}}$  would be substantially degraded. The inference is that human-induced tropospheric warming may also be an important driver of observed increases in tropopause height. Both the direct and indirect lines of evidence support the contention that the troposphere has warmed during the satellite era.

We have shown that both stratospheric cooling and tropospheric warming lead to increases in tropopause height. However, the relative importance of these two factors is uncertain. In the NCEP reanalysis, which has documented errors in stratospheric temperature (31), tropopause height increases even though the troposphere cools. In ERA (22) and PCM, the troposphere warms, and both stratospheric cooling and tropospheric warming contribute to tropopause height increases. Clarification of the relative roles of T2 and T4 changes in recent tropopause height increases is clearly required. This may be facilitated by second-generation reanalyses (such as the recently completed ERA-40 reanalysis), by further study of radiosonde-based temperature soundings, and by high-resolution measurements of the tropopause from Global Positioning System (GPS) data (42).

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- 32. The total linear change is defined as  $b \times n$ , where b is the slope parameter of the linear trend (in hPa per

- month or  ${}^{\circ}\text{C}$  per month) fitted by the standard least-squares method over a specified period of n months.
- 33. For forcing component x, this is calculated as  $(x/y) \times 100$ , where x is the total linear change in  $\rho_{LRT}$  over a stipulated time interval (due to x) and y is the linear change in  $\rho_{LRT}$  (over the same interval) due to ALL. This definition is not applicable if the linear change in ALL is close to zero, a situation that never arises here.
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#### Supporting Online Material

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Fig. S1

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### Dense Packing and Symmetry in Small Clusters of Microspheres

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When small numbers of colloidal microspheres are attached to the surfaces of liquid emulsion droplets, removing fluid from the droplets leads to packings of spheres that minimize the second moment of the mass distribution. The structures of the packings range from sphere doublets, triangles, and tetrahedra to exotic polyhedra not found in infinite lattice packings, molecules, or minimum—potential energy clusters. The emulsion system presents a route to produce new colloidal structures and a means to study how different physical constraints affect symmetry in small parcels of matter.

What defines an optimal packing of a set of n identical spheres? Although centuries old, this question remains both pertinent and pervasive in mathematics and science (I). For packings of an infinite number of spheres, the obvious measure of optimality is the bulk density. As Kepler conjectured and Hales proved (2, 3), the optimal infinite packing is the face-centered cubic (fcc) arrangement, which maximizes the density or, equivalently, minimizes the volume per sphere. But for a finite group of spheres there is no compelling definition of density, and optimality in

finite sphere packings can be defined by the minimization of any physically reasonable variable, such as potential energy or surface area, for example. Different minimization criteria can lead to dramatically different sequences of packings (4), with symmetries that are rarely consistent with that of an infinite (bulk) packing.

In nature, differences in symmetry between finite and bulk packings manifest themselves in a variety of phenomena. The interactions between atoms or particles in many systems, including metals, noble gases, and colloids, are nearly spherically isotropic, and the equilibrium structures at high density are fcc, a consequence of Kepler's conjecture. But finite packings of such particles, as found in isolated clusters, complexes, or local arrangements of bulk phases, frequently

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#### **PERSPECTIVES**

A deep and stormy lake. Lake Tanganyika, just south of the equator in tropical Africa, is one of the world's oldest and deepest lakes. Home to many animal species that live nowhere else, such as the snail Tiphobia horei (inset), Lake Tanganyika has been profoundly affected by global climate change.

insufficient light to support photosynthesis. Photosynthetic production of new organic material is controlled by water mixing, which returns some of the nutri-

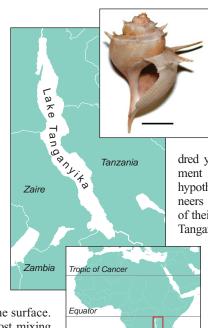
ents from deep water to the surface.

In Lake Tanganyika most mixing is seasonal, dependent on the southeast monsoon, which drives the light surface-water north and brings deep water close to the surface at the south end of the lake. The density gradient

of the stratified water column resists this mixing, and productivity of the lake is strictly controlled by the balance between mixing and stability.

Verburg and his colleagues have searched the sparse historical records for information about lake temperature, transparency, and fauna. Climate warming in the last century has raised the temperature of the lake more at the surface than in the depths. The sharper temperature gradient inhibits mixing, so productivity has fallen. The mass of planktonic organisms is now less than one-third of the mass when measurements were first made 25 years ago. Currently, the transparency is so great that a white disk can be seen in the water to a depth of 13 m instead of 6 m.

More intense stratification has led to drastic changes in water chemistry. Dissolved oxygen does not penetrate to the depth it once did, and one of the endemic snails, Tiphobia horei, which in 1890 was collected to a depth of 300 m, is now restricted to the upper 100 m of the water column (see the figure). Hydrogen sulfide could not be detected above 300 m in 1938; now it extends to a depth of 120 m. Soluble reactive phosphorus in the mixed layer has fallen from 0.29 µM to below the detection limit of 0.01 µM since the mid-20th century. The amount of dissolved silica in deep water has not changed, but in the upper 50 m it has trebled since 1975. Diatoms, which use silica to make their skeletons, have decreased so much that upwelling of silica, in spite of a sharpened density gradient, more than suffices for their needs.



Tropic of Capricorn

Although Verburg et al. have contributed substantially to our knowledge of the modern lake, their conclusion depends on observations by many early biologists. For a hun-

dred years, with no management goal and no scientific hypothesis to test, these pioneers used the best methods of their time to describe Lake Tanganyika. Their data, col-

lected intermittently when war and scientific funding allowed, are sparse, but are sufficient to demonstrate to a very high degree of statistical reliability that there has been a drastic change in mixing of the lake's waters.

New techniques

and conceptual advances (5) have rendered obsolete much of what freshwater biology

students were taught 20 years ago about the annual mixing of lake waters. The National Science Foundation has no program for the physical study of inland lakes and waters, despite long-standing rumors of imminent change. A modest injection of funds might help us to understand the mixing of lake waters as well as we comprehend the mixing of ocean waters.

Verburg *et al.* show that a deep tropical lake with a very rich fauna, which seemed resistant to global climate change, has already been profoundly affected by it. The best efforts of lake-shore nations and conservationists can do little to slow the continuing alteration of Lake Tanganyika. That will take determined international action, of which enforcement of the Kyoto protocols might be a reasonable first step.

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#### ATMOSPHERIC SCIENCE

## Climate Change at Cruising Altitude?

Brian J. Hoskins

limate model studies are widely used to estimate whether observed changes in the climate system are likely to be related to changes in greenhouse gases or other climate forcing agents. On page 479 of this issue, Santer *et al.* (1) report important climate model results of this kind.

The authors consider, separately and in combination, the anthropogenic forcings (caused by greenhouse gases, aerosols, and ozone) and natural forcings (resulting from changes in solar output and volcanic aerosols) that may have influenced climate in the 20th century. However, instead of taking the surface temperature as the measure of climate—as is done in most such studies—they consider a phenomenon characteristic of Earth's atmosphere: the tropopause.

Because Earth's atmosphere is nearly transparent to solar radiation, its tempera-

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ture is generally highest at the Earth surface. Atmospheric ozone, which absorbs solar ultraviolet radiation, creates another temperature maximum, near 0°C, at a height of about 50 km (see the figure). Weather systems and deep convection in the troposphere determine the rate at which the temperature falls with height (the lapse rate). Air masses in the troposphere can move substantially in height or latitude within days. In contrast, the temperature in the stratosphere generally does not change so much with height. Here, solar radiation is very important in the heat balance, motion is quasi-horizontal and less turbulent, and time scales are generally much longer.

The boundary between the troposphere and the stratosphere, the tropopause (see the figure), is generally sharp, whether viewed in terms of changes in lapse rate, potential vorticity [a fluid dynamic property measuring both stability and rotation (2)], or chemical characteristics (3). This sharpness is probably a result of mixing in

#### **PERSPECTIVES**

On a typical day. (Right) Schematic vertical section from equator to pole on a particular day, showing the tropopause with westerly jet streams, tropical convection, and transport and exchange. (Left) Typical vertical profile of atmospheric temperature.

the upper troposphere and of the different concentrations of radiatively important trace constituents on the two sides. The lower stratosphere contains much more ozone, but the troposphere is much moister.

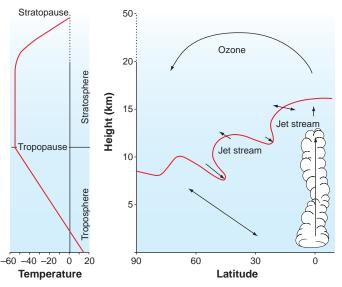
The tropopause typically slopes from about 9 km near the poles to about 18 km near the equator. However, because it is distorted by weather sys-

tems, on any particular day and longitude the tropopause is likely to be far from smooth, with thin tongues of stratospheric air sometimes moving deep into the troposphere (4).

The tropopause is important on a daily basis because it puts a lid on tropospheric convection. It also plays a very active role in the development of large weather systems: One classic view of the growth of weather systems involves the interaction of waves propagating on temperature gradients at the tropopause and at Earth's surface (5).

Not only the amounts of trace chemicals but also the dominant chemical processes differ widely in the regions on either side of the tropopause. For these reasons, there has been much interest in the extent to which the tropopause is permeable, allowing stratosphere-troposphere exchange (6). Such exchange influences stratospheric ozone reduction, tropospheric pollution, and global warming.

The tropospheric air that eventually moves high into the stratosphere crosses



the tropopause in the equatorial region. The extreme dryness of the stratosphere suggests that this must occur mainly over the Indonesian region, where the temperature is low enough to "freeze-dry" it. However, the winds are such that while the air crosses the tropopause it must also move a considerable horizontal distance. A picture of the three-dimensional motion in this region is starting to emerge (7). In the subtropics, there is two-way exchange, whereas at higher latitudes, streamers and cut-off pools of stratospheric air are produced by weather systems and later absorbed into the troposphere (8).

On the basis of the contrasting thermodynamic budgets of the weather-dominated troposphere and radiation-dominated stratosphere, the height of the tropopause can be estimated (9, 10). More simply, assuming that the vertical temperature profile is like that shown in the figure and that the lapse rate in the troposphere does not change, tropopause height changes are related to changes in tropospheric and lower

stratospheric temperatures. An increase in the former and a decrease in the latter—as expected, for example, with increases in carbon dioxide concentrations—both lead to a higher tropopause. In each case, a 1°C change leads to a rise in the tropopause by about 160 m. Stratospheric ozone reduction also leads to stratospheric cooling and, hence, a higher tropopause. In contrast, sulfate aerosols are calculated to cool the troposphere and therefore lower the tropopause. Other natural and anthropogenic forcings will also act to change its height.

The analysis of Santer et al. suggests that over the past 20 years, the observed globally averaged tropopause height has risen by ~200 m. Their model results show that this rise is consistent with the changes in natural and anthropogenic forcings over this period, and that greenhouse gas and ozone changes are mostly responsible for it. Continuing changes in the properties of the tropopause as a result of human activity could have wide-ranging implications because of its physical and chemical roles in the climate system.

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CHEMISTRY -

### Colloidal Molecules and Beyond

Alfons van Blaaderen

bout a hundred years ago, Jean Perrin's experiments on colloids particles a few nanometers to a few micrometers in diameter—convinced even the skeptics that matter consists of atoms and molecules. More recently, the analogy between atoms and colloids has led to in-

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sights into crystal nucleation and growth, the glass transition, and the influence of the range of particle-particle interactions on phase behavior. Moreover, the ability to manipulate colloid crystallization has led to advanced materials such as photonic crystals.

To date, most studies of colloids have used spherical particles or particles with simple shapes, such as rods and plates. Syntheses of nonspherical colloids generally yield a broad size distribution. On page 483 of this issue, Manoharan et al. (1) report a method for making large quantities of identical colloidal particles with complex shapes consisting of equal-sized colloidal spheres.

The authors made these particles by drying the oil out of an oil-in-water emulsion in which spherical colloids were adsorbed to the surface of the oil droplets. Subsequent centrifugation yielded an intriguing sequence of colloidal structures (including the tetrahedral tetramer in the top left panel of the figure). Each structure consists of 2 to 15 spheres. For a given particle size, the spheres are arranged identically in all particles (1).