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A barrier to vertical mixing at 14 km in the tropics: Evidence from ozonesondes and aircraft measurements

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Abstract. We use ozonesondes launched from Samoa (14°S) during the Pacific Exploratory Mission (PEM) Tropics A to show that O_3 mixing ratios usually start increasing toward stratospheric values near 14 km. This is well below the tropical tropopause (as defined either in terms of lapse rate or cold point), which usually occurs between 16 and 17 km. We argue that the main reason for this discrepancy in height between the chemopause and tropopause is that there is very little convective detrainment of ozone-depleted marine boundary layer air above 14 km. We conjecture that the top of the Hadley circulation occurs at roughly 14 km, that convective penetration above this altitude is rare, and that air that is injected above this height subsequently participates in a slow vertical ascent into the stratosphere. The observed dependence of ozone on potential temperature in the transitional zone between the 14-km chemopause and the tropical tropopause is consistent with what would be expected from this hypothesis given calculated clear-sky heating rates and typical in situ ozone production rates in this region. An observed anticorrelation between ozone and equivalent potential temperature below 14 km is consistent with what would be expected from an overturning Hadley circulation, with some transport of high O_3/low θ_e air from midlatitudes. We also argue that the positive correlations between O_3 and N_2O in the transitional zone obtained during the 1994 Airborne Southern Hemisphere Ozone Experiment/Measurements for Assessing the Effects of Stratospheric Aircraft) (ASHOE/MAESA) campaign support the notion that air in this region does have trace elements of stratospheric air (as conjectured previously), so that some of the ozone in the transitional zone does originate from the stratosphere rather than being entirely produced in situ.

1. Introduction

One of the defining characteristics of the troposphere is that it is considered to be vertically well mixed on a timescale of 1-2 weeks. One would therefore not expect chemical tracers with lifetimes longer than 1-2 weeks to exhibit significant vertical gradients within the troposphere. It has become clear, however, that in the tropics, and especially outside regions of active deep convection, that the onset of vertical gradients in many chemical tracers (sometimes called the chemopause) frequently occurs significantly below the lapse rate tropopause. There is a transition zone between ~14 and 17 km in which air starts to assume some of the chemical characteristics of stratospheric air. This depression of the chemopause below the lapse rate (or cold point) tropopause has, at times, been attributed to the transport of stratospheric air into the upper tropical troposphere. For example, O_3 enhancements several kilometers below the tropical tropopause observed above Christmas Island (2°N) during the 1993 Central Equatorial Pacific Experiment (CEPEX) campaign were attributed to stratospheric influx [*Kley et al.*, 1996]. Data from the 1994 Airborne Southern Hemisphere Ozone Experiment/Measurements for Assessing the Effects of Stratospheric Aircraft (ASHOE/MAESA) campaign were used to demonstrate that the onset of vertical gradients in long-lived species such as CFC-11 and N_2O usually occurred several kilometers below the tropical tropopause, and to argue that there was a "standing reserve" of stratospheric air throughout much of the upper tropical troposphere [*Tuck et al.*, 1997]. *Fujiwara et al.* [1998] used ozonesondes and rawindsonde measurements from Indonesia to argue that ozone enhancements in the upper tropical troposphere were

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sometimes associated with breaking equatorial Kelvin waves, which resulted in a downward mixing of stratospheric air into the upper troposphere.

It is also possible, however, that the intermediate chemical characteristics of the transition zone (14 - 17 km) can be attributed to a suppression of the vertical mixing associated with tropical deep convection above 14 km. The notion that tropical deep convection rarely penetrates above 14 km has gained more attention recently [*Highwood and Hoskins*, 1998]. A barrier to vertical mixing near 14 km would be anticipated on thermodynamic grounds since it is near the altitude at which potential temperatures first become equal to the highest equivalent potential temperatures realized in the marine boundary layer (~355-360 K). It is therefore near the maximum altitude an air parcel from the boundary layer can reach by undiluted, nonovershooting ascent [*Reid and Gage*, 1981].

In this paper, we first use ozonesondes launched during the Pacific Exploratory Mission (PEM) Tropics A campaign to demonstrate that there is indeed a chemical transition zone between 14 and 17 km above Samoa (14°S). We then show that the vertical variation in lapse rate above Samoa, and its correlation with ozone, is consistent with an inhibition of tropical deep convection above 14 km. We introduce a very simple model to show that the dependence of ozone on potential temperature in the transition region is consistent with a realistic rate of in situ ozone production and calculated clear-sky heating rates. In Section 3 of the paper, we use correlations of O_3 with N_2O from ASHOE/MAESA

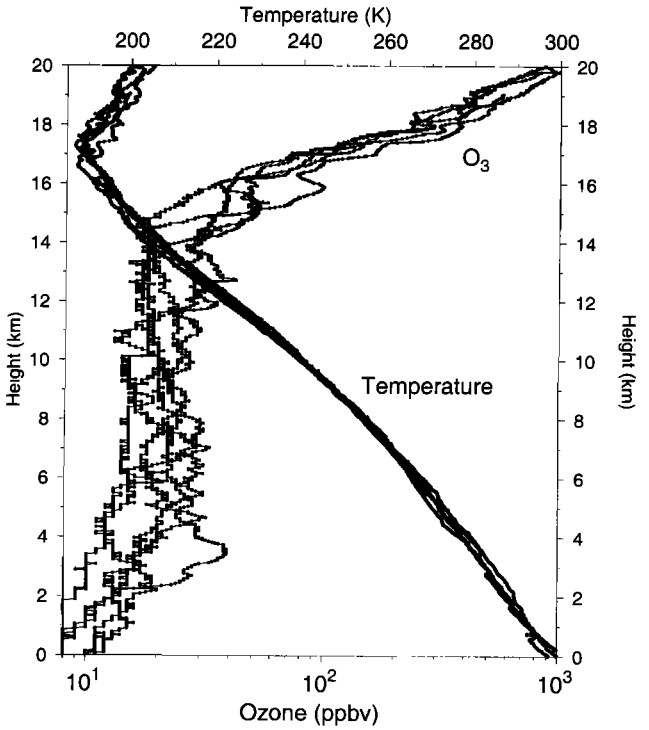

Figure 1. Profiles of temperature and O_3 taken from five ozonesondes launched Samoa in March 1996.

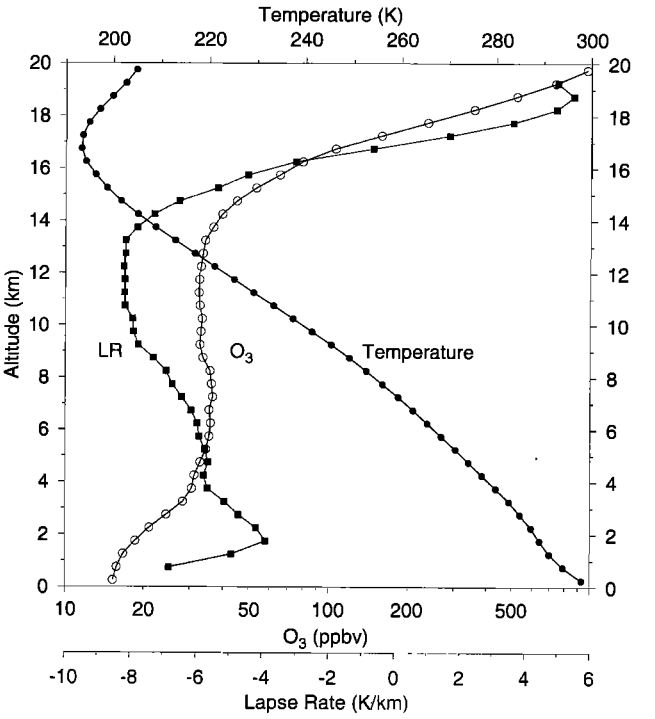

Figure 2. Average profiles of temperature, O_3, and lapse rate(LR) from all 108 Samoan ozonesondes.

to show that stratospheric inmixing is also likely to have some impact on the vertical variation of ozone in this region.

2. Ozonesondes From Samoa

Figure 1 shows temperature and O_3 profiles from five ozonesondes launched from Samoa in March 1996. The cold point tropopause (temperature minimum) is usually between 16.5 and 17.5 km. The standard definition of the tropopause is based on lapse rate, and is defined as the lowest altitude at which the lapse rate decreases to ≤ 2°C/km, provided that the average lapse rate between this height and all higher altitudes within the next 2 km does not exceed 2°C/km [*Craig*, 1965]. The lapse rate tropopause is usually near 16 km. On the other hand, Figure 1 shows that the onset of vertical gradients in O_3 is usually near 14 km, so that the chemopause is typically 2-3 km lower than the tropopause.

The height offsets shown in Figure 1 between the chemopause and tropopause for several individual profiles also appear quite strongly as climatological features. Figure 2 shows the temperature and O_3 mixing ratio profiles obtained by averaging over all 108 Samoan ozonesondes from August 8, 1995, to February 20, 1998. One again sees in the average O_3 profile the onset of an O_3 increase near 14 km and in the temperature profile a tropopause near 17 km.

Figure 2 also shows the vertical variation in lapse rate (defined here as $\Gamma = dT/dz$) above Samoa during this time period. The 14 km O_3 mixing ratio increase is

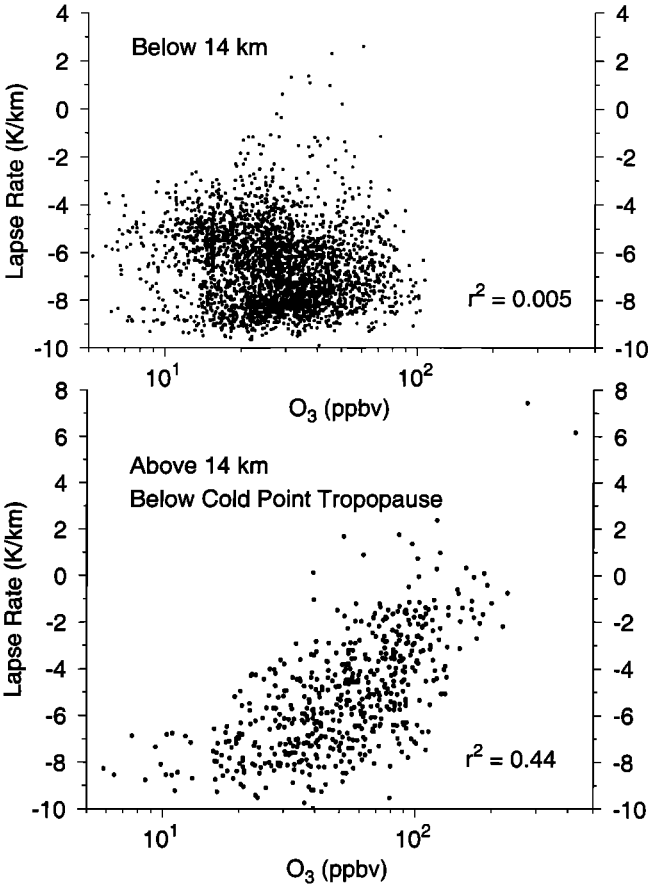

Figure 3. (top) Lapse rate plotted against O_3 mixing ratio. Both sets of measurements were taken from the Samoan ozonesondes. Only altitudes < 14 km were considered. Measurements were first averaged into 0.5 km bins. The correlation coefficient between the lapse rate and the logarithm of the O_3 mixing ratio is 0.005. Temperature and O_3 measurements from each of the 108 ozonesondes used in these plots were first averaged into 0.5-km bins. (bottom) Same as Figure 3, top, except that the measurements shown were taken from between 14 km and the cold point tropopause. The correlation coefficient was 0.44.

coincident with an increase in lapse rate. In the upper tropical troposphere, lapse rates are maintained near the dry adiabatic value of ~ -10 K/km by deep convection. The proximity of the O_3 mixing ratio and lapse rate increases therefore suggests that the O_3 increase is associated with a reduction in the convective flux of O_3 depleted marine boundary layer above this altitude. *Atticks and Robinson* [1983] have previously noted the existence of lapse rate changes in the upper tropical troposphere.

The attribution of the 14-km O_3 mixing ratio increase to a reduction in convective overturning above 14 km is supported by the fact that while variations in O_3 mixing ratio are not correlated with variations in lapse rate below 14 km, they are strongly correlated above 14 km. Figure 3 (top) shows that O_3 mixing ratio and lapse rate are not significantly correlated below 14 km ($r^2 = 0.005$). Figure 3 (bottom) shows that O_3 mixing ratio and lapse rate rate are significantly correlated in the transition zone ($r^2 = 0.44$), defined here as above 14 km but below the cold point tropopause.

Clear sky radiative cooling plays a crucial role in the maintenance of the Hadley circulation because it provides a mechanism by which air parcels can diminish their potential temperature and sink back to the surface after detraining from convective clouds. In cloud-free regions, there is a widely used diagnostic relationship between clear-sky radiative cooling and subsidence [*Lin et al.*, 1998]:

$$-\omega\left(\frac{\alpha}{c_p} - \frac{\partial T}{\partial p}\right) \approx \frac{Q}{c_p} \qquad (1)$$

where ω is the pressure tendency of an air parcel, α the specific volume, c_p the specific heat capacity at constant

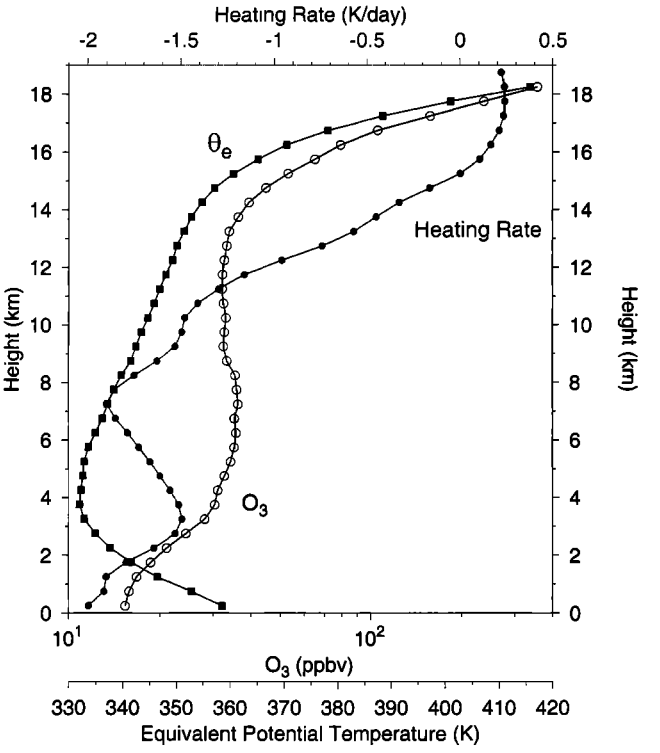

Figure 4. Vertical profiles above Samoa of O_3, equivalent potential temperature (θ_e), and 24-hour averaged clear-sky radiative heating rate. The O_3 and θ_e are averages, in 0.5-km bins, of all 108 ozonesondes from Samoa. θ_e was first calculated using the temperature, pressure, and relative humidity measurements of each ozonesonde. Relative humidity was often not available from the sondes above 10 km. In this case, θ_e was assumed to roughly equal θ. Above 10 km, water vapor concentrations are sufficiently low, even at 100 % relative humidity, that θ_e and θ will differ by <1 K and can be regarded as essentially equivalent. The clear-sky radiative heating rate was calculated by a radiative transfer model [*Fu and Liou*, 1992] using as inputs the averaged ozonesonde measurements of O_3, pressure, temperature, and relative humidity below 30 km, and an Air Force Geophysical Laboratory tropical climatology [*Anderson et al.*, 1986] above 30 km.

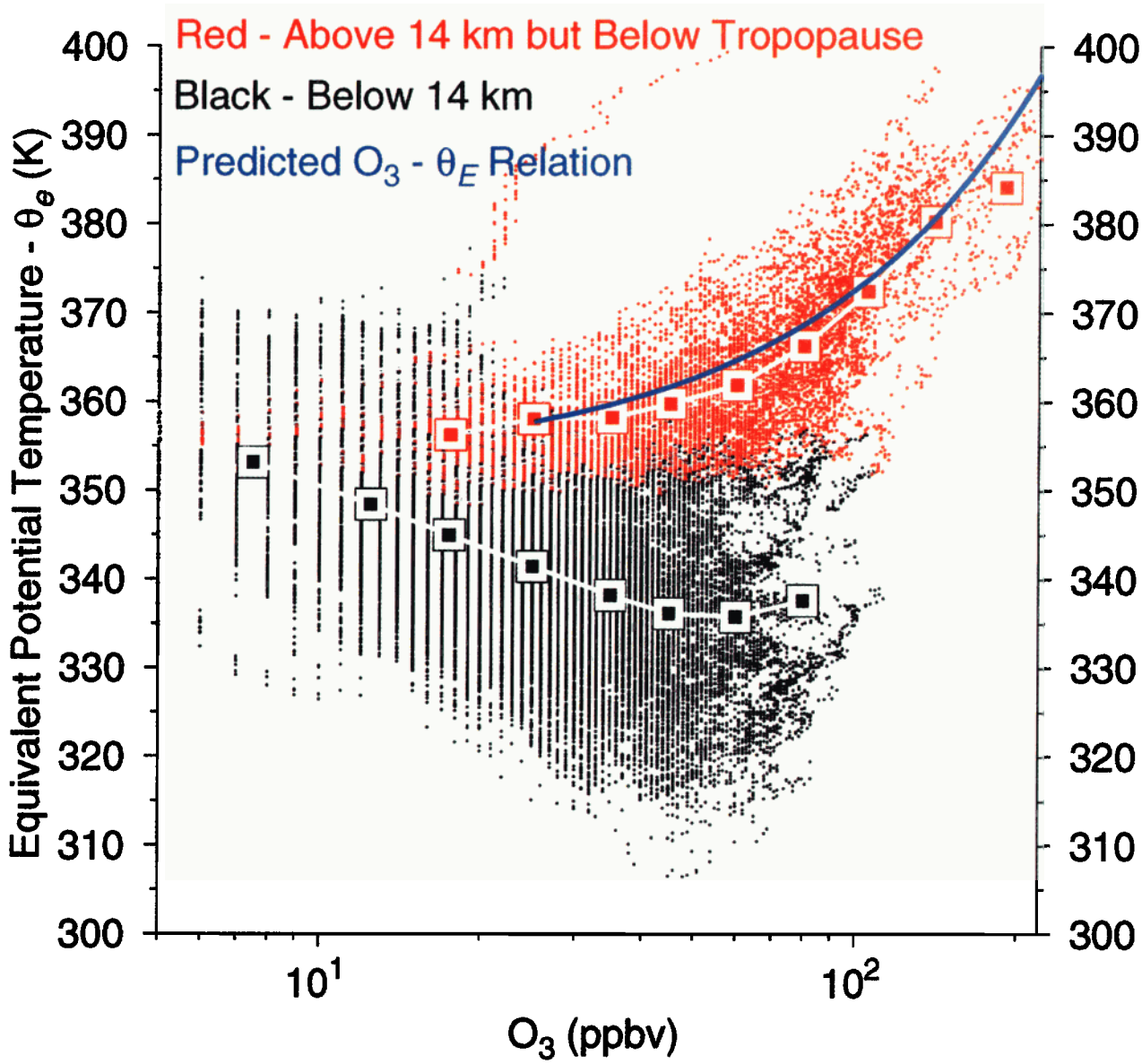

Plate 1. Equivalent potential temperature (θ_e) versus O_3 mixing ratio from all 108 Samoan ozonesondes. Data points shown in red were taken from heights above 14 km but below the cold point tropopause. Data points shown in black are from below 14 km. Squares denote average θ_e values within prescribed O_3 bins.

pressure, T the temperature, p the pressure, Q/c_p the heating rate, and $\frac{\alpha}{c_p} - \frac{\partial T}{\partial p}$ a measure of the static stability of an air parcel. From this expression, clear-sky cooling ($Q < 0$) implies downward motion ($\omega > 0$).

The vertical variation of the clear-sky heating rate above Samoa is shown in Figure 4. There is a transition from clear-sky cooling to heating at 15 km. Provided (1) is valid, this would imply that there is also a transition from large-scale downward to upward vertical motion at the same altitude. A downward mass flux above 15 km would be possible, but it would have to be associated with cloud processes such as evaporative or cloud top cooling. Positive radiative heating rates above 15 km do not preclude a convective mass flux through this altitude, but do suggest that most of the air detraining from convective clouds above 15 km will subsequently rise into the stratosphere rather than sinking back to the surface. The transition zone would therefore not be part of the overturning Hadley circulation as it is conventionally understood.

The θ_e profile shown in Figure 4 also supports the notion that convection above 14 km is rare. In the absence of convective overshooting, or mixing with higher θ_e air, the maximum attainable height of a convecting air parcel is the height at which its θ_e becomes equal to that of the background θ_e. Above this height, air parcels will be negatively buoyant. Figure 4 shows that θ_e is rarely higher than 355 K in the marine boundary

layer. After a minimum in the midtroposphere, equivalent potential temperatures again reach 355 K near 14.5 km. On thermodynamic grounds one would therefore not expect most convective clouds in the tropics to rise higher than this altitude.

The rapid increase in θ_e above 14.5 km is probably associated with some combination of reduced convective input of lower θ_e air from below, the near-simultaneous transition to clear-sky radiative warming (providing a source of θ_e), and, possibly, an increased inmixing of higher θ_e stratospheric air (see below).

Figure 4 shows that θ_e and O_3 both start increasing rapidly above 14 km. In Plate 1, individual θ_e and O_3 measurements have been plotted against one another to demonstrate the existence of a strong positive correlation between θ_e and O_3 in the transition zone. Simultaneous measurements of θ_e and O_3 mixing ratio above 14 km are shown in red. The measurements from below 14 km are shown in black. The red and black squares denote average θ_e values in prescribed O_3 bins.

The emergence of a positive correlation between O_3 and θ_e in the transition zone can be attributed to a slow large-scale ascent in this region. We demonstrate this using a very simple heuristic calculation. The rate of change of O_3 with respect to θ_e can be written

$$\frac{dO_3}{d\theta_e} = \frac{dO_3/dt}{d\theta_e/dt}. \quad (2)$$

We will assume that the only mechanism for changing O_3 concentrations in the transition region is in situ chemical production, and that the only source or sink of θ_e is radiative heating. We also assume that the rates of ozone production and radiative heating are constant between $\theta_e = 357$ K (about 14.75 km) and $\theta_e = 390$ K (about 17.25 km). In this case, the dependence of O_3 on θ_e between these two equivalent potential temperatures can be written

$$O_3(\theta_e) = O_3(357\text{ K}) + \frac{dO_3/dt}{d\theta_e/dt} \times (\theta_e - 357\text{ K}). \quad (3)$$

where $O_3(357\text{ K})$ is the average ozone mixing ratio for $\theta_e = 357$ K. Plate 1 indicates that $O_3(357\text{ K}) \approx 20$ ppbv. A reasonably representative value for diurnally averaged ozone production in the background tropical atmosphere at 14 km is 0.5 ppbv/d [*Folkins et al.*, 1997a, *Wennberg et al.*, 1998]. We will assume here that $dO_3/dt = 0.5$ ppbv/d, though it should be kept in mind that dO_3/dt will increase dramatically above 16 km as the rate of O_2 photolysis increases. Figure 4 indicates that a reasonably representative radiative heating rate between 14.75 and 17.25 km is 0.1 K/d. In this case, (3) reduces to

$$O_3(\theta_e) = 20\text{ ppbv} + 5\text{ ppbv/K} \times (\theta_e - 357\text{ K}). \quad (4)$$

This curve is shown in Plate 1 as the solid blue line. It reproduces the average dependence of O_3 on θ_e in the transition region extremely well. This indicates that a slow upward ascent in the transition region (associated with a radiative warming), in concert with an in situ ozone production, is, in principle, able to explain the depression of the chemopause below the tropopause shown in Figure 2.

The main origin of the anticorrelation between θ_e and O_3 below 14 km shown in Plate 1 is probably that the marine boundary layer is both a chemical sink of O_3 and a source of θ_e. O_3 is destroyed quite rapidly (on a timescale of about a week) under the low NO, high water vapor, and moderately high ultraviolet light conditions prevalent of the remote marine boundary layer [e.g., *Thompson et al.*, 1993]. On the other hand, the θ_e of an air parcel entrained into the marine boundary layer will usually increase as it picks up moisture from the sea surface. These processes account for the decrease in O_3 and the increase in θ_e below 3 km shown in Figure 4.

A second and perhaps equally significant reason for the θ_e-O_3 anticorrelation below 14 km is transport from the midlatitude upper troposphere. Air moving toward Samoa from midlatitudes will ordinarily descend along quasi-isentropic surfaces [e.g., *Danielsen et al.*, 1987]. Owing to its descent, this air will have a lower relative humidity and lower θ_e than more humid tropical air on the same isentropic surface. This air also tends to be enhanced in O_3 because it originates close to the midlatitude tropopause [*Harris and Oltmans*, 1997].

A third but probably less important reason for the anticorrelation between θ_e and O_3 below 14 km is that the free troposphere below 14 km will tend to be a θ_e sink (clear-sky radiative cooling) and an O_3 source (generally positive O_3 production rates). The anticorrelation between θ_e and O_3 below 14 km shown in Plate 1 therefore reflects the sources and sinks of θ_e and O_3 an air parcel experiences within the overturning Hadley circulation, and the fact that there is a downward transport of air into the tropics from the upper mid-latitude troposphere.

3. Correlations of O_3 With N_2O From ASHOE/MAESA

One way of determining whether the O_3 molecules of a particular region originate in the stratosphere or troposphere is by examining the extent to which variations in O_3 correlate with N_2O. In the stratosphere the ultraviolet wavelengths that split O_2 and produce O_3 also account for a majority of the N_2O destruction. This helps give rise to a strongly negative correlation between O_3 and N_2O. In the troposphere, where N_2O is essentially inert and its distribution is near uniform, variations in O_3 are not coupled to variations in N_2O unless a significant fraction of the O_3 is of stratospheric origin. In this section, we use measurements of O_3 and N_2O from the 1994 ASHOE/MAESA campaign to argue that at least some fraction of the O_3 in the transition zone is likely to have originated in the stratosphere.

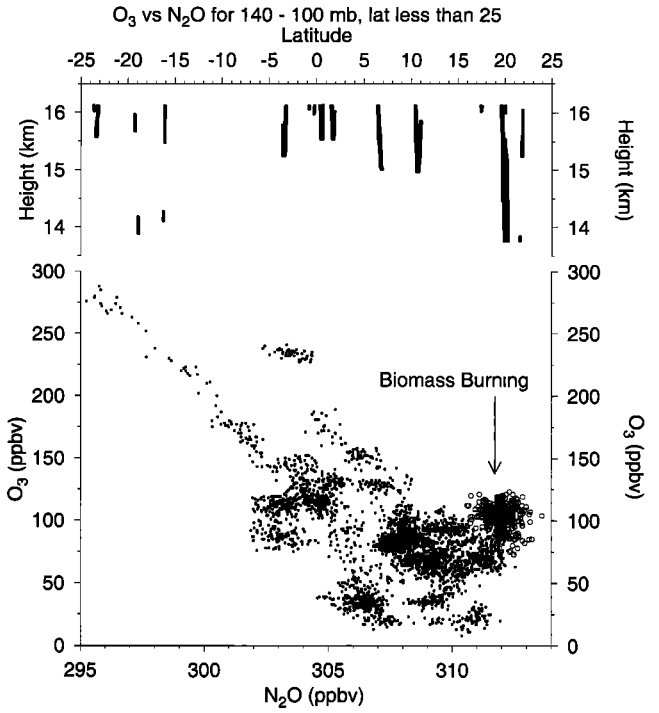

Figure 5. (top) Locations (in pressure height and latitude) of the O_3 and N_2O measurements from the ER-2 shown in the bottom panel. (bottom) O_3 versus N_2O for all measurements in the upper tropical troposphere (140 - 100 mbar, and latitude $< 25°$) from ASHOE/MAESA. Data points denoted by open circles were taken from within a biomass burning plume.

The NASA ER-2 usually flies at a pressure of $\sim$ 55 mbar (19-20 km). During ASHOE/MAESA the ER-2 took measurements in the tropics while flying from bases in Hawaii (22°N) and Fiji (14°S). Measurements in the vicinity of the tropical tropopause were taken during ascents and descents from these bases, as well as during a number of dips to 110 mbar from the cruising pressure of 55 mbar. These dips usually occurred near the equator. In Figure 5, all O_3 measurements from ASHOE/MAESA between 140 and 100 mbar (14.5 - 16.5 km), and within 25°of the equator, are plotted against simultaneous measurements of N_2O. Figure 5(top) shows the pressure altitude and latitudes of the measurements. There is a strong overall anticorrelation between O_3 and N_2O throughout the transition zone. Enhancements in O_3 are usually associated with decreases in N_2O. An exception is the cluster of points indicated by open circles. These points, also enhanced in CO and NO, have been previously identified as belonging to a biomass burning plume originating from the Indonesian fires of 1994 [*Folkins et al.*, 1997b]. Because the O_3 enhancements within this plume are of tropospheric origin, N_2O assumes its typical tropospheric value of 312 ppbv.

4. Discussion and Conclusions

We have shown that there are a number of changes occurring near 14 km above Samoa, and possibly throughout the tropics, which collectively indicate a suppression of tropical deep convection above 14 km: the simultaneous onset of rapid increases in O_3 mixing ratio and lapse rate, an increase in θ_e beyond the characteristic marine boundary layer value of 355 K, the emergence of positive correlations between O_3 and lapse rate, increased evidence for stratospheric inmixing as given by the positive correlation between O_3 and N_2O, and a transition from net clear-sky radiative cooling to heating. The enhanced thermal stratification above 14 km, and the suppression of vertical mixing with which it is associated, helps to maintain the existence of a reasonably well-defined chemopause, defined in terms of O_3, near 14 km. At least at Samoa the tropical tropopause, as commonly defined either in terms of lapse rate or cold point, does not correspond to the upper limit of the well-mixed troposphere, as is commonly assumed.

We have also argued that while the anticorrelation between θ_e and O_3 below 14 km is consistent with an overturning Hadley circulation, the positive correlation above 14 km is most consistent with a slow large-scale ascent in which θ_e and O_3 increase simultaneously due to positive radiative heating and in situ chemical production respectively. This positive correlation may also be enhanced by the horizontal or vertical inmixing of higher θ_e and O_3 air from the stratosphere. In Figure 6, we have summarized the changes in the properties of the tropical troposphere associated with the emergence of a transition zone at 14 km, as indicated in the Samoan ozonesondes. Figure 6 presents a similar perspective on the vertical structure of the tropical troposphere as given in Figure 13 of *Highwood and Hoskins* [1998] but focuses more on the sources and sinks of ozone and θ_e an air parcel experiences within the overturning Hadley circulation and in the more stably stratified transition zone between 14 and 17 km.

It is important to note that our conclusions, based largely on measurements from Samoa, may not be representative of all tropical regions, in particular those closer to the inter tropical convergence zone or over continents where convection may reach higher altitudes. It would be worthwhile investigating if the relationships discussed here apply to other regions. More measurements would also be desirable to help construct a more comprehensive ozone budget of the transition zone than the simplified analysis discussed here. Determining the origin of O_3 in this zone is important for climate since O_3 in this region exhibits a strong greenhouse forcing [*Lacis et al.*, 1990].

Danielsen [1993] has argued that the overshooting of deep convective clouds into the lower tropical stratosphere and the irreversible mixing between tropospheric and stratospheric air with which they are associated

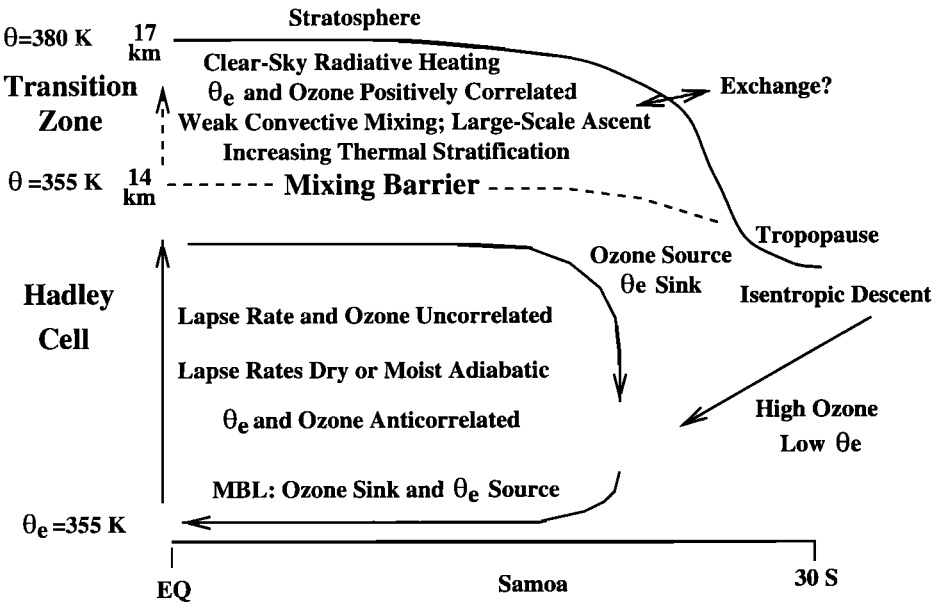

Figure 6. Cross section of the tropical troposphere illustrating some of the differences between the tropical transition zone (14-17 km) and the tropical troposphere below 14 km. Also shown are some of the sources and sinks of θ_e and O_3 within the Hadley circulation.

play an important role in the upward flux of tropospheric air into the stratosphere. We have suggested that the large-scale upward ascent in the lower tropical stratosphere associated with the Brewer Dobson circulation penetrates into a transition zone in the upper tropical troposphere. This is roughly consistent with a previous argument based on vertical temperature correlations that the upward motion in the lower tropical stratosphere extends as low as 125 mbar ($\sim$ 15.25 km, the same altitude at which the heating rate shown in Figure 4 goes to zero) [*Reid and Gage*, 1996]. If true, the overshooting of of tropical deep convection into the lower stratosphere would not be a prerequisite for cross tropopause transport. Perhaps the convective mass flux into the transition zone is approximately that needed to sustain the Brewer Dobson circulation.

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