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On the distribution and variability of ozone in the tropical upper troposphere: Implications for tropical deep convection and chemical-dynamical coupling

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[1] Tropical ozonesonde measurements display events of substantially reduced or near-zero ozone in the upper troposphere that can be coherent over broad spatial scales. Available observations indicate that these events occur most frequently between about 300 and 100 mbar in the tropical southwest Pacific region. The spatial structure of the events suggests linkages to deep convection as the primary cause, with the potential for long-range transport from the southwest Pacific to other locations. Observations are sparse in time as well as space, but suggest possible long-term changes in tropical ozone transport and the frequency of deep convection there since the 1980s. **Citation:** Solomon, S., D. W. J. Thompson, R. W. Portmann, S. J. Oltmans, and A. M. Thompson (2005), On the distribution and variability of ozone in the tropical upper troposphere: Implications for tropical deep convection and chemical-dynamical coupling, *Geophys. Res. Lett.*, *32*, L23813, doi:10.1029/2005GL024323.

1. Introduction

[2] Ozone in the troposphere and stratosphere is linked to atmospheric transport processes as well as chemistry. The distribution and variability of ozone is central to the atmospheric thermal structure, and it is in tropopause region that ozone perturbations can exert their greatest influence on climate [e.g., *Thuburn and Craig*, 2002; *Forster and Joshi*, 2005]. Recent studies have underscored the importance of understanding the full vertical [e.g., *Fu et al.*, 2004] and latitudinal distributions [*Thompson and Solomon*, 2005] of temperature changes from the surface to the stratosphere, as a cornerstone in climate change detection and attribution.

[3] Ozone at tropical latitudes is sensitive to the deep convection that occurs there, which rapidly transports material from altitudes near the surface to the upper troposphere and feeds the lowermost stratosphere [*Thompson et al.*, 1996; *Kley et al.*, 1996; *Folkins et al.*, 2000; *Folkins and Martin*, 2005]. Such air may contain high ozone or ozone precursors associated, for example, with biomass burning or other sources of pollution [e.g., *Thompson et al.*, 1996]. On the other hand, ozone in very clean boundary layer air can be destroyed under wet, hot conditions in the deep tropics (where solar illumination is intense) by reactions such as those involving hydrogen radicals (OH, HO_2), leading to surface ozone abundances of only a few ppbv [*Johnson et al.*, 1990], in contrast to typical values of the order of 10–30 ppbv [*Thompson et al.*, 2003a, 2003b]. Balloon flights during ship transects first suggested that convective transport of ozone-poor boundary layer air can lead to "near-zero" ozone levels of a few ppbv or less in the region of tropical deep convective outflow near 200 mbar [*Kley et al.*, 1996].

[4] In this paper, balloon-borne electrochemical (ECC) ozonesonde observations are presented in order to gain further insight into the behavior of ozone close to the tropopause in the tropics (≈20°S–20°N). The focus is on gaining a better understanding of the coupling of ozone to dynamical processes as well as possible trends. We consider not just near-zero ozone episodes but also the frequency of observations of ozone below chosen thresholds of 20 and 30 ppbv depending upon location, referred to here as reduced ozone events. We demonstrate that the largest observed frequency of reduced ozone events occurs in the tropical southwest Pacific ("warm pool") region, providing new support for the key role of that region in convective transport. We also present evidence for long-term changes in reduced and near-zero tropical ozone events that are likely to be due to changes in deep convection, which may be linked to climate change. Some observations raise questions regarding whether contributions from other processes (e.g., chemical removal) may also play a role, and these are considered.

2. Distribution and Variability of Tropical Ozone

[5] Ozonesonde data are generally taken weekly, but sampling limitations due to instrument problems or data gaps imply that detailed differences between the sites should be interpreted with caution [see, e.g., *Thompson et al.*, 2003a, 2003b; *Oltmans et al.*, 1996]. Data quality checks have been performed in the observations shown here. Ozonesonde data provide unique information regarding vertical resolution (i.e., spatial scales of a few tens of meters) as well as very high local sensitivity to steep gradients and local values as low as a few ppbv.

[6] Figure 1 shows the distribution of the sites considered in this paper, along with the average ozone profiles observed during 1998–2004 at illustrative stations as well as representative profiles showing typical reduced or near-zero ozone measurements. Most of these stations began systematic ozone measurements in the late 1990s in

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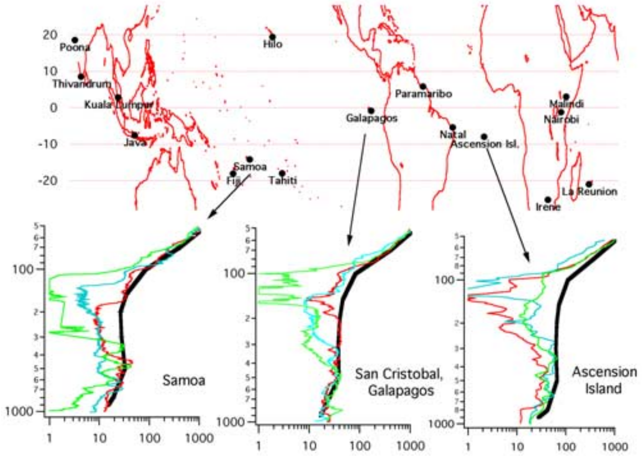

Figure 1. Ozonesonde profiles at illustrative tropical stations (mixing ratio in ppbv versus pressure). Heavy solid black lines show average profiles for 1998–2004, while colored lines display illustrative near-zero and reduced ozone events. Similar shapes were observed for such events at other locations.

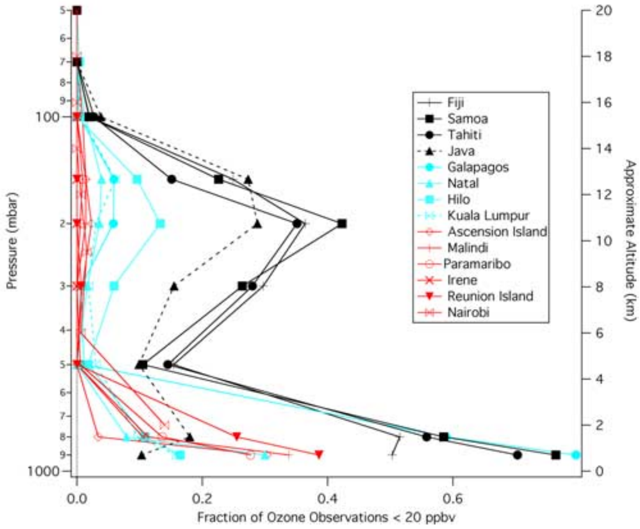

Figure 3. Vertical profile of frequency of occurrence of ozone mixing ratios below 20 ppbv at tropical ozonesonde stations, for the period from 1998–2004.

association with the SHADOZ network. The average profiles represent the means of the raw data over bins of ±10 mbar for 900, 800, 500, 300, and 200 mbar, ±5 mbar at 150 and 100 mbar, and ±2 mbar at 70 and 50 mbar. The same binning is used in Figures 2, 3, and 4 and in Table 1. Figure 1 illustrates illustrative reduced and near-zero ozone events, which occur in the upper troposphere at many different locations in the tropics examined in this paper. Their character is similar at a remarkable range of stations: such events extend over broad rather than narrow layers, ranging from about 300 to 100 mbar, rather than at lower or higher altitudes.

[7] A particularly extensive and deep layer in which ozone abundances less than 8 ppbv from about 300 to 100 mbar was observed at Samoa in March of 1998 (green line in Samoa panel of Figure 1). Measurement of ozone abundances below 10 ppbv poses an observational challenge, due to the possibility of instrumental background signals. Figure 2 presents observations at 150 mbar at three neighboring sites in the tropical southwest Pacific at Tahiti, Fiji, and Samoa. These data show that near-zero ozone events were observed nearly simultaneously at all three sites in early 1998, and the three data sets track one another at many other times as well. This provides new and unique

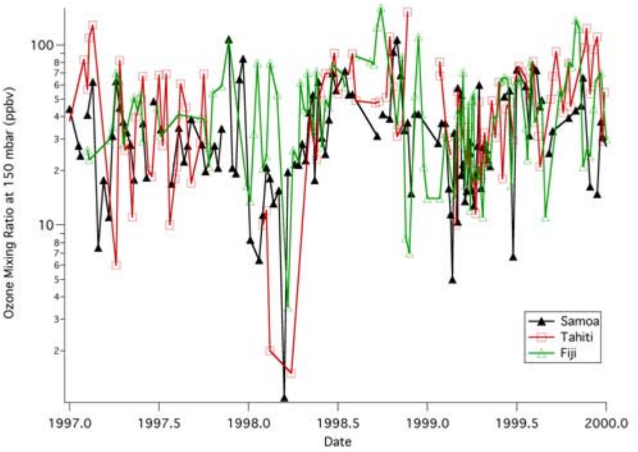

Figure 2. Ozone observations at 150 mbar at Samoa, Tahiti, and Fiji during the period of measurement overlap at all three southwest Pacific sites (1997–2000).

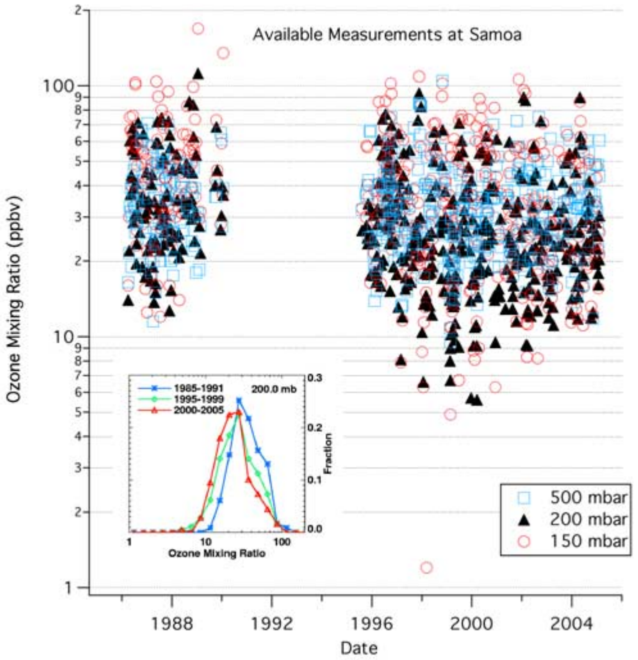

Figure 4. Time series of ozonesonde observations at Samoa at 500, 200, and 150 mbar. The inset shows the shift observed in the probability distribution function for the 200 mbar observations, averaged over available five-year intervals.

Table 1. Changes in Reduced Ozone in the Upper Troposphere From the Late 1970s to Present

Period	Number of Sondings	Percent of Reduced Ozone Events at 200 mbar
	Samoa	
1986–1990	115	10 (<20 ppbv)
1995–1996	96	18
1997–1999	82	47
2000–2002	104	42
2003–mid 2005	73	32
	Hawaii	
1982–1985	84	7 (<20 ppbv)
1986–1990	221	4
1991–1995	184	15
1996–1999	159	16
2000–2004	189	14
	Natal	
1979–1982	32	3 (<30 ppbv)
1990–1992	65	2
1997–2000	123	18
2001–2002	117	14
2003–mid 2005	103	5

evidence suggesting that such events are real rather than the result of instrumental artifacts.

[8] Figure 3 further probes the distribution and variability of reduced ozone abundances below 20 ppbv by showing measurements at all of the sites as a function of altitude during 1998–2004. While Figure 1 illustrates how events of similar character may be observed at many different sites, Figure 3 shows that their frequency of occurrence differs greatly from one location to another. Some broad patterns are evident. Reduced ozone events have never or only very infrequently been observed at some stations (e.g., Malindi, Nairobi, and Irene in Africa and Paramaribo, Surinam). The largest frequency of reduced ozone abundances occurs at Samoa, Fiji, Tahiti, and Java in the tropical southwest Pacific region. This area has long been known to be a region of enhanced tropical deep convection, linked to warm SSTs there [e.g., *Hartmann*, 1994]. Thus, the horizontal distribution of reduced tropical ozone events is qualitatively consistent with a primary link to deep convection.

[9] The shapes of the profiles display a sharp maximum in reduced ozone events near 300–150 mbar, while the overall range of reduced ozone events extends from a lower boundary near 300 mbar to a few events reaching as high as the base of the stratosphere at 100 mbar, suggesting that these events can influence lower stratospheric as well as tropospheric ozone abundances. Figure 3 is consistent with the vertical profile expected for links to outflow from deep convection, which maximizes near 200–150 mbar and extends from about 300 to 100 mbar [e.g., *Folkins et al.*, 2000; *Folkins and Martin*, 2005; *Thuburn and Craig*, 2002; *Gettelman and de Forster*, 2002]. Thus, both the horizontal and vertical distributions of reduced ozone events support a link to convection.

[10] What is the origin of near-zero ozone events at locations outside the tropical southwest Pacific? Galapagos is the only site outside of the southwest Pacific with frequent low ozone events in the boundary layer, but the much more limited number of low ozone events at higher altitudes there as compared to Tahiti, Fiji, and Samoa supports the view that convection dominates the observed behavior. The similarity of the vertical profiles at Fiji, Samoa and Tahiti to those obtained at Java and Hawaii in spite of greater pollution in the boundary layer at the latter two locations probably suggests a role for horizontal transport. However, it is not clear why such transport would yield near-zero ozone events at these locations but never at others such as Malindi. This raises the question of whether other factors may contribute to reducing or enhancing local upper tropospheric ozone, such as chemistry related to high clouds or convection. Reactions on, or accommodation in, ice cloud particles [e.g., *Roumeau et al.*, 2000] or in association with pollutants contained in cloud, such as soot from biomass burning may play some role. While the structure of the observations in longitude and altitude suggests that convection is the dominant cause of reduced and near-zero ozone values, other factors could play a secondary role.

3. Long Term Ozonesonde Records in the Tropics

[11] Systematic ozonesonde measurements were taken prior to the 1990s at only three sites in the deep tropics. The earliest data are from Natal beginning in the late 1970s, while observations at Samoa and Hawaii began in the 1980s.

[12] Figure 4 shows the time series of observations of ozone at Samoa at 500, 200 and 150 mbar. At 500 mbar, no obvious long-term changes are observed despite some interannual variability. At 200 and 150 mbar on the other hand, the figure reveals that the frequencies of occurrence of reduced and near-zero ozone air at Samoa have changed substantially over time. The inset illustrates the pronounced change in the probability distribution function (PDF) observed at 200 mbar. It should be emphasized that such observations provide information on deep convection, but not on total convection.

[13] Table 1 shows the changes observed at Samoa, Hawaii, and Natal at 200 mbar along with the number of available observations for each period at the three sites. Sampling is very limited at Natal in the early years. Observations are more extensive at Samoa and Hawaii, where the frequency of occurrence of reduced ozone events shows an evident increase between the 1980s and the mid-1990s. Hourly surface ozone data at Samoa and Hawaii do not display a long-term increase in reduced ozone events at the ground over this period (not shown). There are no known instrumental or reporting changes that could produce the trends shown in Figure 4 and Table 2, and it is noteworthy that the data from Natal versus Samoa represent sites maintained by different groups. Thus, the observed increases in the frequency of occurrence of reduced and near-zero ozone events suggest changes in ozone transport and increases in the frequency of tropical deep convection since about the mid-1990s at the available sites in the tropics.

4. Discussion and Conclusions

[14] We have presented observations from a range of sites in the tropics displaying new information regarding near-

zero and reduced ozone in the tropical upper troposphere. Observations at neighboring sites document similar events (Figure 2), demonstrating that these remarkable events reflect real changes rather than instrumental artifacts. The available data do not allow complete coverage in latitude, longitude, nor time, but they indicate that the greatest frequency of occurrence of reduced ozone occurs in the tropical southwest Pacific region near 300 to 100 mbar, consistent with links to the frequency of deep convection there (Figure 3). The coherent horizontal and vertical structures support the view that reduced and near-zero ozone events are primarily due to convection via transport of air that has been depleted through chemical processing in very clean tropical boundary layer air (as pointed out in the seminal papers by *Kley et al.* [1996], and *Folkins et al.* [2000]).

[15] The data are sparse, but the available observations at Samoa, Hawaii, and Natal indicate changes in tropical ozone transport at these locations through increases in the frequency of occurrence of reduced and near-zero ozone events from the 1980s to the period after about the mid-1990s.

[16] Other studies have suggested long-term changes in tropical deep convection and the strength of the associated circulation using different approaches, such as convective available potential energy (CAPE) data [*Gettelman et al.*, 2002], changes in the cold point [*Zhou et al.*, 2001] and ERBE data [*Chen et al.*, 2002]. The results presented in this paper show that ozonesonde observations represent independent and complementary evidence to elucidate spatial and temporal changes in tropical deep convection that is likely to affect ozone and hence temperatures in the tropical upper troposphere and lowermost stratosphere, and may link to climate change.

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References

Chen, J., B. E. Carlson, and A. D. Del Genio (2002), Evidence for strengthening of the tropical general circulation in the 1990s, *Science*, *295*, 838–841.

Folkins, I., and R. V. Martin (2005), The vertical structure of tropical convection and its impact on the budgets of water vapor and ozone, *J. Atmos. Sci.*, *62*, 1560–1573.

Folkins, I., S. J. Oltmans, and A. M. Thompson (2000), Tropical convective outflow and near surface equivalent potential temperature, *Geophys. Res. Lett.*, *20*, 2549–2552.

Forster, P., and M. Joshi (2005), The role of halocarbons in the climate change of the troposphere and stratosphere, *Clim. Change*, *71*, 249–266.

Fu, Q., C. M. Johanson, S. G. Warren, and D. J. Seidel (2004), Contribution of stratospheric cooling to satellite-inferred tropospheric temperature trends, *Nature*, *429*, 55–58.

Gettelman, A., and P. M. F. de Forster (2002), A climatology of the tropical tropopause layer, *J. Meteorol. Soc. Jpn.*, *80*, 911–924.

Gettelman, A., D. J. Seidel, M. C. Wheeler, and R. J. Ross (2002), Multidecadal trends in tropical convective available potential energy, *J. Geophys. Res.*, *107*(D21), 4606, doi:10.1029/2001JD001082.

Hartmann, D. L. (1994), *Global Physical Climatology*, 408 pp., Elsevier, New York.

Johnson, J. E., et al. (1990), Ozone in the marine boundary layer over the Pacific and Indian oceans: Latitudinal gradients and diurnal cycles, *J. Geophys. Res.*, *95*, 11,847–11,856.

Kley, D., et al. (1996), Observations of near-zero ozone concentrations over the convective Pacific: Effects on air chemistry, *Science*, *274*, 230–233.

Oltmans, S. J., et al. (1996), Tropospheric ozone during Mauna Loa Observatory photochemistry experiment 2 compared to long-term measurements from surface and ozonesonde observations, *J. Geophys. Res.*, *101*, 14,569–14,580.

Roumeau, S., P. Bremaud, E. Riviere, S. Baldy, and J. L. Baray (2000), Tropical cirrus clouds: A possible sink for ozone, *Geophys. Res. Lett.*, *27*, 2233–2236.

Thompson, A. M., et al. (1996), Where did tropospheric ozone over southern Africa and the tropical Atlantic come from in October 1992? Insights from TOMS, GTE/TRACE-A and SAFARI-92, *J. Geophys. Res.*, *101*, 24,251–24,278.

Thompson, A. M., et al. (2003a), Southern Hemisphere additional ozonesondes (SHADOZ) 1998–2000 tropical ozone climatology: 1. Comparison with Total Ozone Mapping Spectrometer (TOMS) and ground-based measurements, *J. Geophys. Res.*, *108*(D2), 8238, doi:10.1029/2001JD000967.

Thompson, A. M., et al. (2003b), Southern Hemisphere additional ozonesondes (SHADOZ) 1998–2000 tropical ozone climatology: 2. Tropospheric ozone variability and the zonal wave one, *J. Geophys. Res.*, *108*(D2), 8241, doi:10.1029/2002JD002241.

Thompson, D. W. J., and S. Solomon (2005), Recent stratospheric climate trends as evidenced in radiosonde data: Global structure and tropospheric linkages, *J. Clim.*, in press.

Thuburn, J., and G. C. Craig (2002), On the temperature structure of the tropical substratosphere, *J. Geophys. Res.*, *107*(D2), 4017, doi:10.1029/2001JD000448.

Zhou, X. L., M. A. Geller, and M. Zhang (2001), Cooling trend of the tropical cold point tropopause temperatures and its implications, *J. Geophys. Res.*, *106*, 1511–1522.

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