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A Tropical Atlantic Paradox: Shipboard and Satellite Views of a Tropospheric Ozone Maximum and Wave-one in January-February 1999.

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Abstract. During the Aerosols99 trans-Atlantic cruise from Norfolk, VA, to Cape Town, South Africa, daily ozonesondes were launched from the *R/V Ronald H Brown* between 17 January and 6 February 1999. A composite of tropospheric ozone profiles along the latitudinal transect shows 4 zones, nearly identical to the ozone distribution during a January-February 1993 trans-Atlantic cruise [Weller et al., 1996]. Sondes from the cruise and Ascension Island (8S, 14.5W), as well as the Earth-Probe (EP)/TOMS satellite instrument, show elevated tropospheric ozone (> 35 Dobson Units) throughout the south Atlantic in January 1999. Ozone layers associated with biomass burning north of the ITCZ (Intertropical Convergence Zone) are prominent at 0-5 km from 10-0N, but even higher ozone (100 ppbv, 5-15 km) occurred south of the ITCZ, where it was not burning - an ozone "paradox" that contributes to a wave-one zonal pattern in tropospheric ozone. Back trajectories, satellite observations and shipboard tracers suggest that the south Atlantic ozone results from a combination of interhemispheric transport, aged stratospheric-upper tropospheric air, and possibly from ozone supplied by lightning nitric oxide.

1. Introduction: Tropical tropospheric ozone from satellites and sondes.

Tropospheric ozone (O_3) over the tropical Atlantic displays several paradoxes that point to a complex interaction of dynamical and photochemical influences. High-resolution tropospheric O_3 and aerosol data from TOMS satellites [Herman et al., 1997; Thompson and Hudson, 1999] reveal that the highest tropospheric column O_3 in the south tropical Atlantic occurs in September-October, 2-3 months after the peak of regional biomass burning that produces much of the free tropospheric O_3 [Thompson et al., 1996]. The satellite record (site: <http://metosrv2.umd.edu/~tropo/14yr_d.d>) also shows that during the northern hemisphere (NH) biomass burning season, tropical tropospheric O_3 does not attain O_3 column amounts seen during southern hemisphere burning even though NH burning is as intense.

In-situ data confirm these features. Two trans-Atlantic cruises that launched ozonesondes in the January-April period [Smit et al., 1989; Weller et al., 1996] showed more free tropospheric O_3 south of the ITCZ (Intertropical Convergence Zone) than north of it, downwind from savanna fires. We refer to this phenomenon as an "ozone reversal" or "tropical Atlantic paradox." Aircraft observations in January 1991 [Jonquières and Marenco, 1998] also showed this feature. At Ascension Is. (8S, 14.5W; <http://code916.gsfc.nasa.gov/Data_services/shadoz>), integrated tropospheric O_3 from sondes [Thompson et al., 1996] can reach 50 DU (1 DU = 2.69×10^{16} molec-cm^{-2}) in January and February.

The Aerosols99 oceanographic cruise on the *R/V Ronald H Brown*, from Norfolk, Virginia (37N, 76W) to Cape Town, South Africa (34S, 22E) in January-February 1999, offered the first opportunity to look at the NH-SH (southern hemisphere) ozone transition during a northwest-to-southeast Atlantic crossing with a suite of chemical and aerosol instrumentation. Earth-Probe/TOMS tropospheric O_3 and aerosol products complemented the cruise, giving a regional view of elevated O_3. Tropospheric O_3 observed during the cruise resembled a January-February 1993 *R/V Polarstern* cruise [Weller et al.,1996], with four latitude zones and the distinctive O_3 reversal. We offer evidence for photochemical O_3 sources (African biomass burning and lightning). However, in the SH, where mid-tropospheric O_3 > 100 ppbv, interhemispheric transport, convective mixing, and large-scale subsidence, appear to interact in ways that cannot be deconvoluted without additional free tropospheric tracer data.

2. In-situ and Satellite Measurements. Analysis.

Ozone and temperature profiles on the *R/V Ronald H. Brown* were determined with an electrochemical concentration cell ozonesonde (ENSCI 2Z) in combination with an RS-80/15 Väisäla radiosonde and a HumiCap humidity sensor; 1-sec data were recorded. The procedures for sonde preparation and data acquisition were developed by NOAA/Climate Diagnostics and Monitoring Lab. [Komhyr et.al, 1989]. All but four of 22 launches (17 Jan. to 6 Feb. 1999) reached 30 km.

Shipboard O_3 and carbon monoxide (CO) were measured with a commercial UV photometer (TEI model 49) and a modified commercial nondispersive infrared gas filter correlation instrument (TEI model 48C), respectively [Parsons and Dickerson, 1999]. Hourly averages of 1-min data appear here. AOT (aerosol optical thickness) was read using a multichannel sun photometer (Solar Light Co.) on days with cloud-free midday conditions.

TOMS tropospheric O_3 data between 20N and 20S are provided by near-real-time processing at <http://metosrv2.umd.edu/~tropo>. Absorbing aerosol gridded data are from the TOMS archive. Lightning flash counts were obtained from the Lightning Imaging Sensor (LIS) aboard the Tropical Rainfall Measuring Mission (TRMM) satellite. Back trajectories were run with the GSFC isentropic trajectory model initialized from the ozonesonde launch locations. NCEP analyses from standard 2.5x2.5deg 0Z and 12Z fields were used in the model. These compared favorably to winds from shipboard radiosondes. To look at tracer origins, air parcels were followed along potential temperature surfaces at lower and middle tropospheric altitudes for 5 days.

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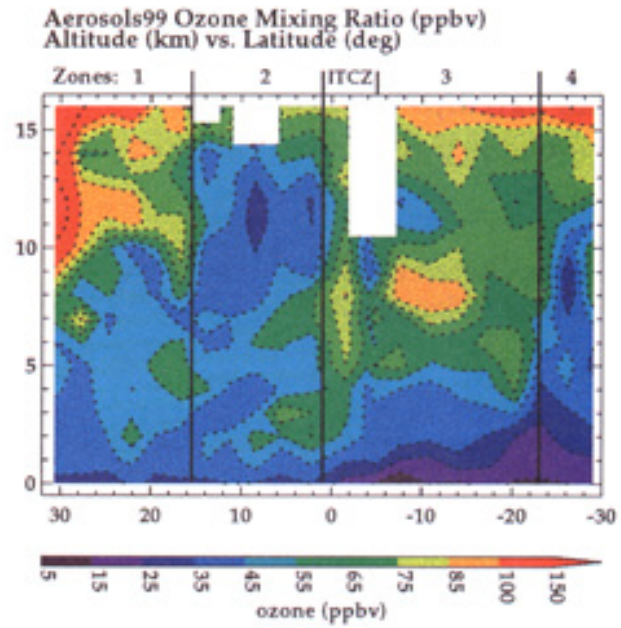

Figure 1. Cross-section of tropospheric/lower stratospheric O_3 mixing ratio based on 22 soundings taken during the *R/V R H Brown* Aerosols99 cruise. 1-km averages used; all but 3 soundings reached 10 hPa. Four zones: 1: 30-14N (17-22 Jan.); 2: 14N-0 (23-27 Jan.); 3: 0-23S (28 Jan.-3 Feb.); 4: 23-30S (4-6 Feb.).

3. Ozonesonde Latitudinal Cross-section and Other Tracers in Four Zones.

Figure 1 shows a latitudinal cross-section of tropospheric O_3 derived from the sondes. Other parameters - temperature profiles, convective cloud cover and pyrogenic tracers - were used to define the ITCZ. Four distinct O_3 profile types appeared along the transect. In the following discussion the terms lower, middle and upper tropospheric O_3 are used interchangeably with 0-5 km, 5-10 km, and 10-15 km altitude.

➳ Zone 1: NH mid-latitude air of mixed troposphere-stratosphere origins. For example, upper tropospheric O_3 > 100 ppbv around 30N coincided with low water vapor (not shown) traceable back to high latitudes.

➳ Zones 2 and 3 (14N to 23S): A broad tropical band straddles either side of the equator. Relative lower and middle tropospheric O_3, as depicted in 0-5 km and 5-10 km amounts, are reversed

Table 1. Integrated tropospheric ozone from sondes (in DU).

Integ. O_3*	P'stern Oct.-Nov.	P'stern Jan.-Feb.	Aerosols99	Pacific Jan.-Feb, 1999
0-5 km	11.2	15.4	13.7	5.8
5-10 km	13.4	13.6	12.8	4.6
10-15 km	8.9	8.9	7.0	2.8

*14N-20S; 1993 Polarstern data [Weller et al., 1996]; Jan.-Feb. 1999 Pacific from mean of Tahiti, Samoa sondes.

(Figure 2). Four features support a Zone 2-Zone 3 demarcation at the equator: 1) north of the equator, a sharp 17.5 km tropopause is defined thermally (not shown) or by the steepest O_3 gradient, but the SH tropopause is less definite (Figure 3); 2) there are more O_3 peaks > 60 ppbv at 0-5 km in the NH than in the SH (Figure 3); 3) the NH has a more extended low O_3 layer from 8-14 km, suggesting convective transport from the MBL (marine boundary layer); 4) at the equator, MBL O_3 decreases sharply, from ~35 ppbv in the NH to < 20 ppbv in the SH (Figure 1).

➳ Zone 4: Subtropical SH (23-30S). The 5-10 km O_3 peaks disappear and surface O_3 is 10-20 ppbv ("ship ozone," Figure 2), consistent with typical remote SH O_3.

Figure 2 shows three tracers measured from the ship, AOT at 380 nm, surface O_3 mixing ratio and CO mixing ratio (scaled by a factor of 0.5). Shipboard O_3 falls more distinctly into two zones (NH mean ~30 ppbv, SH mean ~15 ppbv) than four, except for the variability denoted by peaks in convectively active regions near the equator. AOT, which is hard to classify due to sparse data, roughly corresponds to the latitude divisions of the ozonesondes. However, the zones of CO are somewhat offset from the ozonesondes, resembling other shipboard tracers (aerosol nitrate, K, black carbon; Bates et al., 2000). From 5-18S, CO varied about a 60 ppbv mean, implying some mixing with the free troposphere.

4. Regional Views of Tropospheric Ozone and Wave-One. Comparison with Other Cruises.

Integrated tropospheric O_3 from the Zone 2 and Zone 3 sondes (mean = 36 DU) agrees with the corresponding satellite value in Figure 4A to within the +/-4 DU error of the modified-residual method [Hudson and Thompson, 1998]. The Jan.-Feb., 1993 *Polarstern* sondes at the same latitudes averaged 39 DU (Table 1). Tropospheric O_3 from sondes at Ascension Is. and Natal, Brazil (6S, 35W) during Jan. and Feb. 1999 averages 37 DU. This

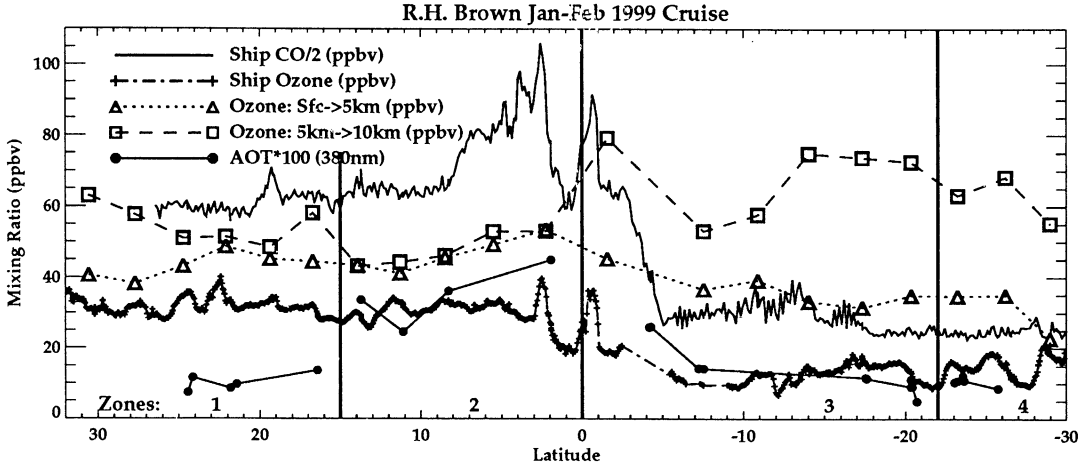

Figure 2. Tracers along *R/V Ronald H Brown* track. Surface O_3 and CO (x1/2) mixing ratios in ppbv, from ship measurements; 0-5 km, 5-10 km integrated O_3 from sondes. AOT (at 380 nm) from clear-sky, midday sun photometer readings.

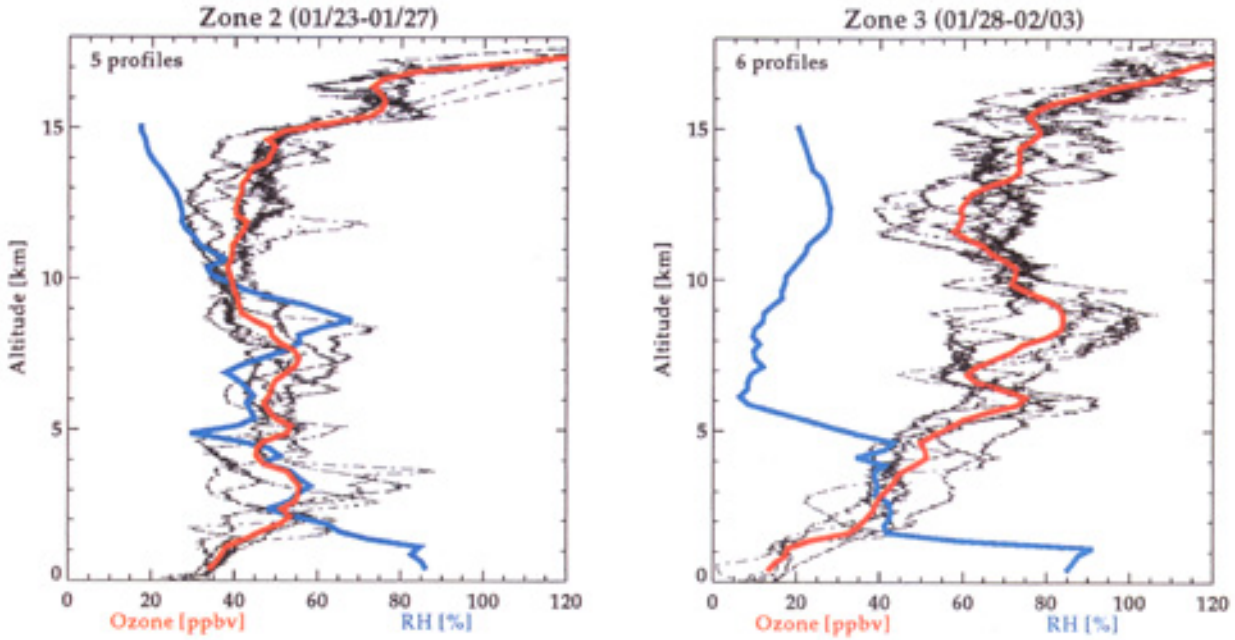

Figure 3. O_3 (ppbv, mixing ratio) and relative humidity (%, mean, blue) for Zones 2 and 3. Individual O_3 profiles (black) plotted along with 0.25 km interval means (red). Near-zero mixing ratio in one Zone 3 profile at 0-0.4 km occurred when sonde passed through ship stack plume. O_3 profiles during Jan.-Feb., 1999 at Ascension and Natal, Brazil [Thompson and Witte, 1999] include peaks similar to those in Zone 3.

supports the satellite view of elevated tropospheric O_3 extending across the Atlantic.

Figure 4A shows tropospheric O_3 over the Atlantic in January 1999 to be greater than over the adjacent continents and the rest of the equatorial band (not shown). This gives rise to a zonal wave-one pattern [Shiotani, 1992; Hudson and Thompson, 1998]. This is seen in Table 1 that shows O_3 from the Zone 2-Zone 3 latitudes on the Aerosols99 cruise and from January-February 1999 Pacific sondes at Samoa (14S) and Tahiti (18S). Ozone layers on two *Polarstern* cruises (Table 1) are similar to those of Aerosols99, implying that the wave-one persists during the October-February period. Relatively high low-mid tropospheric O_3 is expected over the Atlantic from continental burning and subsidence, but upper tropospheric O_3 also contributes to the Atlantic-Pacific contrast.

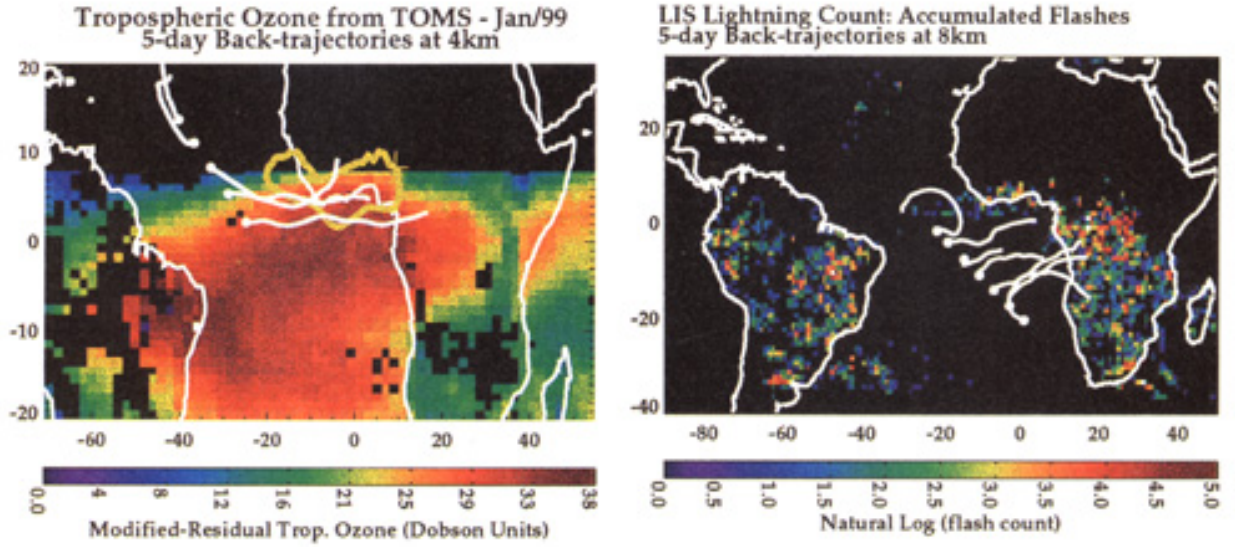

Figure 4. (A) TOMS tropospheric O_3, January 1999 average. Satellite O_3 valid where modified-residual method is reliable (~5N to 20S). Gaps in ozone correspond to clouds, where TOMS data are not used; compare lightning locations in (B). Five-day back trajectories from position of Zone 2 ozonesonde launches (23-27 Jan.) along ship track, initialized at 4 km, overlie gold-contoured area of high TOMS smoke aerosol, averaged from 20-27 Jan. 1999. **(B)** same as (A) except for trajectories at 8 km, superimposed over total LIS lightning flashes for 28 Jan.-3 Feb. 1999. Starting points from Zone 3 sonde launches. Clusters of back trajectories (start time = 12Z) were run to capture uncertainties associated with analyzed winds (NCEP 2.5 x 2.5 deg). Line shown corresponds to weighted central point in each cluster.

5. Discussion: Interpretation of Ozone in Zones 2 and 3.

Section 3 pointed out signatures of convective mixing and biomass burning in Zone 2. Figure 4A shows a northeast-to-southwest orientation of the tropospheric O_3 maximum, suggestive of transport from western African biomass fires, the latter denoted by TOMS smoke aerosol. Back trajectories initialized from the Zone 2 sonde launch positions at 4 km (0-5 km peaks in Figure 3, left) show transport over biomass burning regions.

Upper tropospheric high-ozone features above 13 km in Zone 3 are attributed to stratospheric influences in subtropical sondes (Réunion, 21S, 55E; Baray et al., 2000; Samoa, 14S, 171W; Folkins et al.,1999) and aircraft O_3 observations [Gouget, 2000].

In Zone 3, the greatest complexity in O_3 is in the middle troposphere, where shipboard tracers cannot resolve chemical and dynamical processes. Figure 3 (right, relative humidity) shows that this region is relatively dry, implying subsidence of stratospheric or aged tropospheric air. Figure 1 shows two O_3 peak regions, one at the ITCZ and one from 8-18S. The ITCZ feature corresponds to 3 soundings, one at 2.2N, a sounding to 600 hPa at 0.5S and one six hours later at 1.6S. Vertical redistribution of O_3 (and presumably O_3 precursors) taking place at the ITCZ may extend southward. Air parcels at relatively constant altitude (e.g. 6-km O_3 peak in Figure 3, right) could be advected northeast to southwest, a process observed in airborne O_3 and other tracers by Jonquières and Marenco [1998]. Figure 3 (right) shows a second distinct O_3 peak in the vertical (100 ppbv at 8-10 km). This could be introduced by convection with detrainment at 8-10 km and interhemispheric transport. An alternative explanation for the 8-10 km O_3 feature is lightning nitric oxide (NO), directly injected into the middle troposphere, where 5-15 ppbv O_3/day may form over the south Atlantic [Thompson et al., 1996]. Superposition of 8-km back trajectories (Figure 4B) initiated from sonde locations at which 8-10 km O_3 exceeded 90 ppbv (Figure 3) shows parcels intersecting African areas with high satellite lightning flash counts 4-5 days earlier. The lifetime of upper tropospheric NO allows 4-8 DU O_3 to form during transit to the ship.

6. Summary

The first northwest-to-southeast Atlantic cruise with O_3 soundings (Jan.-Feb., 1999), displayed four zones, similar to three *Polarstern* Atlantic cruises. SH O_3 was elevated through the middle and upper troposphere (mean column, 0-15 km = 37 DU, in agreement with TOMS), a feature that satellite and sonde measurements show extending across the south Atlantic. The appearance of higher O_3 in the SH during the NH savanna burning season is difficult to interpret with only shipboard tracers. It appears to be due to a combination of interhemispheric advection of NH lower and mid-tropospheric O_3, enrichment by stratospheric O_3 and possibly augmentation by a lightning NO source of mid-tropospheric O_3.

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References

Baray, J.-L., V. Daniel, G. Ancellet, and B. Legras, Planetary-scale tropopause folds in the southern subtropics, *Geophys. Res. Lett.*, 27, 353-356, 2000.

Bates, T. S., et al., Regional physical and chemical properties of the marine boundary layer aerosol across the Atlantic during Aerosols99: An overview, *J. Geophys. Res.*, submitted, 2000.

Folkins, I., M. Loewenstein, J. Podolske, S. J. Oltmans, and M. Proffitt, A 14 km mixing barrier in the tropics: Evidence from ozonesondes and aircraft measurements, *J. Geophys. Res.*, 104, 22095-22102, 1999.

Gouget, H., Case study of a tropopause fold and of subsequent mixing in the subtropics of the Southern Hemisphere, *Atmos. Environ.*, 34, 2653-2658, 2000.

Herman, J. R., P. K. Bhartia, O. Torres, C. Hsu, C. Seftor, and E. Celarier, Global distribution of UV-absorbing aerosols from Nimbus 7/TOMS data, *J. Geophys. Res.*, 102, 16911-16922, 1997.

Hudson, R. D., and A. M. Thompson, Tropical tropospheric ozone (TTO) maps from TOMS by a modified-residual method, *J. Geophys Res.*, 103, 22129-22145, 1998.

Jonquières, I. and A. Marenco, Redistribution by deep convection and long-range transport of CO and CH_4 emissions from the Amazon basin, as observed by the airborne campaign TROPOZ II during the wet season, *J. Geophys. Res.*, 103, 19075-19091, 1998.

Komhyr, W. D., S. J. Oltmans, P. R. Franchois, W. F. J. Evans, and W. A. Matthews, The latitudinal distribution of ozone to 35 Km altitude from ECC ozonesonde observations, 1985-1987, *Ozone in the Atmosphere*, ed. R. D. Bojkov and P. Fabian, , pp. 147-150, 1989.

Parsons, R. L., and R. R. Dickerson, NOAA Ship Ronald H. Brown: World global climate study, *Sea Technology*, 40, 39-45, 1999.

Shiotani, M. Annual, quasi-biennial and El Nino-Southern Oscillation (ENSO) time-scale variations in Equatorial total ozone, *J. Geophys. Res.*, 97, 7625-7634, 1992.

Smit, H., D. Kley, S. McKeen, A. Volz, and S. Gilge, The latitudinal and vertical distribution of tropospheric ozone over the Atlantic Ocean in the southern and northern hemispheres, in *Ozone in the Atmosphere*, ed. R. D. Bojkov and P. Fabian, pp. 419-422, 1989.

Thompson, A. M. and R. D. Hudson, Tropical tropospheric ozone (TTO) Maps from Nimbus 7 and Earth-Probe TOMS by the modified-residual method: Evaluation with sondes, ENSO signals and trends from Atlantic regional time series, *J. Geophys. Res.*, 104, 26961-26975, 1999.

Thompson, A. M. and J. C. Witte, SHADOZ (Southern Hemisphere ADditional Ozonesondes): A new ozonesonde data set for the Earth Science Community, *Earth Observer*, 11 (4), 27-30, 1999.

Thompson, A. M., K. E. Pickering, D. P. McNamara, M. R. Schoeberl, R. D. Hudson, J. H. Kim, E. V. Browell, V. W. J. H. Kirchhoff, and D. Nganga, 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, 24251-24278, 1996.

Weller, R., R. Lilischkis, O. Schrems, R. Neuber, and S. Wessel, Vertical ozone distribution in the marine atmosphere over the central Atlantic Ocean (56°S-50°N), *J. Geophys. Res.*, 101, 1387-1399, 1996.

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