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Zonal asymmetries in southern hemisphere column ozone: Implications of biomass burning

J. R. Ziemke, S. Chandra, A. M. Thompson, and D. P. McNamara

NASA Goddard Space Flight Center, Greenbelt, Maryland

Abstract. This study compares Nimbus 7 total ozone mapping spectrometer (TOMS) total column ozone with ozone data from two different satellite instruments measuring vertical profiles of ozone in the stratosphere. The first instrument is the microwave limb sounder (MLS) onboard the upper atmosphere research satellite (UARS) and the second is the solar backscattered ultraviolet (SBUV2) nadir sounder aboard NOAA 11. Previous studies have shown that TOMS data exhibit a zonal wave maximum (amplitudes ~20–30 Dobson units) in total column ozone in the tropical South Atlantic region near 0° longitude. This wave structure occurs in all seasons but maximizes around August–October in association with intense biomass burning in South America and southern Africa. Results of this investigation show that MLS stratospheric column ozone integrated between 1 to 68 hPa shows no such feature. Horizontal structures of 1 to 68-hPa MLS column ozone are found to be incoherent with TOMS and SBUV2 total column data in the tropics in all seasons. This study provides the first evidence from a UARS data set that the southern tropical wave 1 peak in TOMS may have relatively small dependence on stratospheric ozone. Combined MLS, TOMS, SBUV2, and ozonesonde station data show that zonal asymmetries observed in total column ozone in the tropics originate primarily from tropospheric effects (dynamics coupled with biomass burning). Outside of the tropics, zonal patterns in total ozone originate mostly from stratospheric dynamics. Wave signatures in TOMS, SBUV2, and MLS column ozone all show generally coherent horizontal structures in middle and high latitudes, especially during seasons of large planetary wave events such as in the southern hemisphere during southern winter-spring.

Introduction

Using satellite ozone measurements, *Fishman et al.* [1991, 1992] identified a seasonal maximum in tropospheric ozone in the tropical South Atlantic region during southern spring. When this maximum occurs, it engulfs the South Atlantic, with anomalies extending far westward across the South American continent. The South Atlantic peak in tropospheric ozone is related to intense biomass burning in Africa and Brazil around July–October every year (A. M. Thompson et al., 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, submitted to *Journal of Geophysical Research*, 1996 (hereinafter referred to as Thompson et al., submitted manuscript, 1996)). These biomass burning products are a potential source for tropospheric ozone. In addition, *Krishnamurti et al.* [1993] showed that persistent tropospheric subsidence and horizontal winds in the tropical Atlantic also play a role in the formation of this ozone anomaly. Besides subsidence in the South Atlantic during October, lower (upper) level westward (eastward) winds transport biomass burning products into the South Atlantic from Africa (South America).

The method used by *Fishman et al.* [1991, 1992] was to subtract stratospheric column ozone from total column, forming a residual estimate of tropospheric ozone. Stratospheric and total column ozone measurements were derived from Stratospheric Aerosol and Gas Experiment (SAGE) and version 6 TOMS data, respectively. As noted by *Fishman et al.* [1992], the method will not be exact because of subtracting two large quantities, each with ~3% precision. In addition, although SAGE is capable of measuring the lowest portions of the stratosphere, it is a solar occultation instrument with poor areal and temporal coverage (especially in the tropics) when compared with total ozone mapping spectrometer (TOMS), solar backscattered ultraviolet (SBUV2), or microwave limb sounder (MLS). In the tropics, SAGE measurements occur approximately once (twice if using both sunrise and sunset measurements) every 25–30 days within a 5°–10° latitude band (see, for example, *McPeters et al.* [1994]), leading to only

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one or two samples for every monthly mean estimate. The episodic sampling of SAGE introduces additional errors in monthly estimates of SAGE ozone profiles in the tropics because of natural variabilities caused by persistent tropical waves in ozone [*Ziemke and Stanford*, 1994].

The purpose of this study is not to estimate tropospheric ozone as done by *Fishman et al.* [1992], but rather to determine the sources of tropical and southern hemisphere (SH) extratropical stationary waves in total ozone using lower stratospheric temperatures and ozone data from ozonesondes, MLS, TOMS, and SBUV2.

Data

The MLS instrument onboard the UARS satellite simultaneously measures temperature, water vapor, chlorine monoxide, and ozone. In this study, we use daily version 3 ozone volume mixing ratio measurements from the 205-GHz channel [*Froidevaux et al.*, 1994]. UARS pressure levels selected for this study are 68.1, 46.4, 31.6, 21.5, 14.7, 10, 6.81, 4.64, 3.16, 2.15, 1.47, and 1 hPa. MLS stratospheric column ozone ($\Delta\Omega$) was derived by integrating volume mixing ratio (X) over pressure (P) from 1 hPa to 68 hPa: $\Delta\Omega = A\int_{1hPa}^{68hPa} X dP$, where A is a constant ensuring Dobson units (DU) in the integration (A=7880 DU Pa^{-1}).

It should be noted that the 68.1-hPa version 3 MLS ozone data are not direct measurements but instead are derived by averaging the retrievals at 46.4 and 100 hPa. Ozone data for 100 hPa are not recommended for scientific study since they may have biases caused by being partly climatology rather than measurement. In short, while version 3 MLS ozone data properly extend down to 46.4 hPa, data at 68.1 hPa should be used with caution since they are part climatology. Examination (D. Deboer and J. Gleason, personal communication, 1996) of both Halogen Occultation Experiment (HALOE) and SAGE ozone data seasonally averaged between 20°S and the equator shows approximately 8 DU ozone column between 68 and 100 hPa and around 25 DU between 46 and 100 hPa. These estimates are consistent with ozonesonde data (discussed later).

Total column ozone used in this study are Nimbus 7 version 6 and preliminary version 7 (R. D. McPeters, personal communication, 1996) TOMS and NOAA 11 SBUV2 [*Planet et al.*, 1994] data. In contrast to version 6 TOMS data, version 7 includes correction for errors induced by low marine stratus clouds. These errors in TOMS version 6 measurements are discussed by *Thompson et al.* [1993] and *Hudson et al.* [1995] for the tropical South Atlantic region in southern spring; biases in TOMS version 6 were found to be ~+20 and +5 DU in this region for daily and monthly measurements, respectively.

The TOMS spectrometer measures total column ozone in a broad scanning motion about nadir, providing nearly complete global coverage (except for latitudes in polar night) each day. In comparison, SBUV2 provides retrievals for both total column and vertical ozone profiles but for nadir measurements only. Overlapping time periods between MLS, SBUV2, and TOMS are from September 16, 1991, through April 30, 1993. Temperature fields were computed (via standard five-point finite differencing) from daily (1200 UTC) standard National Meteorological Center (NMC) geopotential height analyses.

For consistency, all retrieved satellite data other than SBUV2 were binned to equivalent 5° latitude (85°S to 85°N) by 15° longitude block structures. SBUV2 data were gridded to coarser 10° latitude by 30° longitude blocks. Spurious and missing data were replaced using a subjective three-dimensional (latitude, longitude, day) Gaussian weighting procedure followed by linear interpolation in time. Monthly and seasonal averaging of data did not include days when MLS experienced missing data. The number of days used in the averages thus depends on the number of measurement days of MLS at a given latitude. Over the course of the entire MLS record, approximately one half of the data are missing poleward of ±30° because of the alternating satellite yaw maneuvers that occur roughly every 36 days. Note that there are some conspicuous missing MLS data even in the tropics (see, for example, *Froidevaux et al.* [1994]); we find in our analyses approximately only 2% of daily MLS tropical ozone data missing during September 16, 1991 to April 30, 1993.

Comparisons between total ozone and ground-based ozonesonde measurements were made using several tropical stations. Specific stations and time intervals are discussed later.

Zonal Asymmetries in the Southern Hemisphere During Southern Spring

Plate 1 shows anomaly fields of SH version 6 TOMS and MLS column ozone for September 1992. These anomalies represent zonal wave perturbations relative to zonal means and are shown in percent. The TOMS percentages shown in Plate 1 are mostly independent of which version (6 or 7) is used. (Percentages for version 6 TOMS shown in Plate 1 were found to be identical everywhere to version 7 results, except in the South Atlantic near 0° longitude where the 7% anomaly in version 6 was instead ~5–6% in version 7.)

TOMS total ozone (Plate 1, top) shows a clear zonal wave 1 structure that is out of phase between the tropics and extratropics. MLS column ozone anomalies (Plate 1, bottom) are seen to be nearly identical to TOMS in the middle and high latitudes. This strong wave 1 outside the tropics in TOMS and MLS ozone is a manifestation of vertical and horizontal advection effects that cause a similar wave pattern in lower stratospheric temperatures [*Wirth*, 1993]. Although MLS stratospheric column ozone explains most zonal variability (~80%) of TOMS wave 1 in high latitudes, the tropical wave 1 in TOMS is clearly not present in MLS. This latter observation appears to be generally consistent with the SAGE results of *Shiotani and Hasebe* [1994], where again there was no similar TOMS tropical wave 1

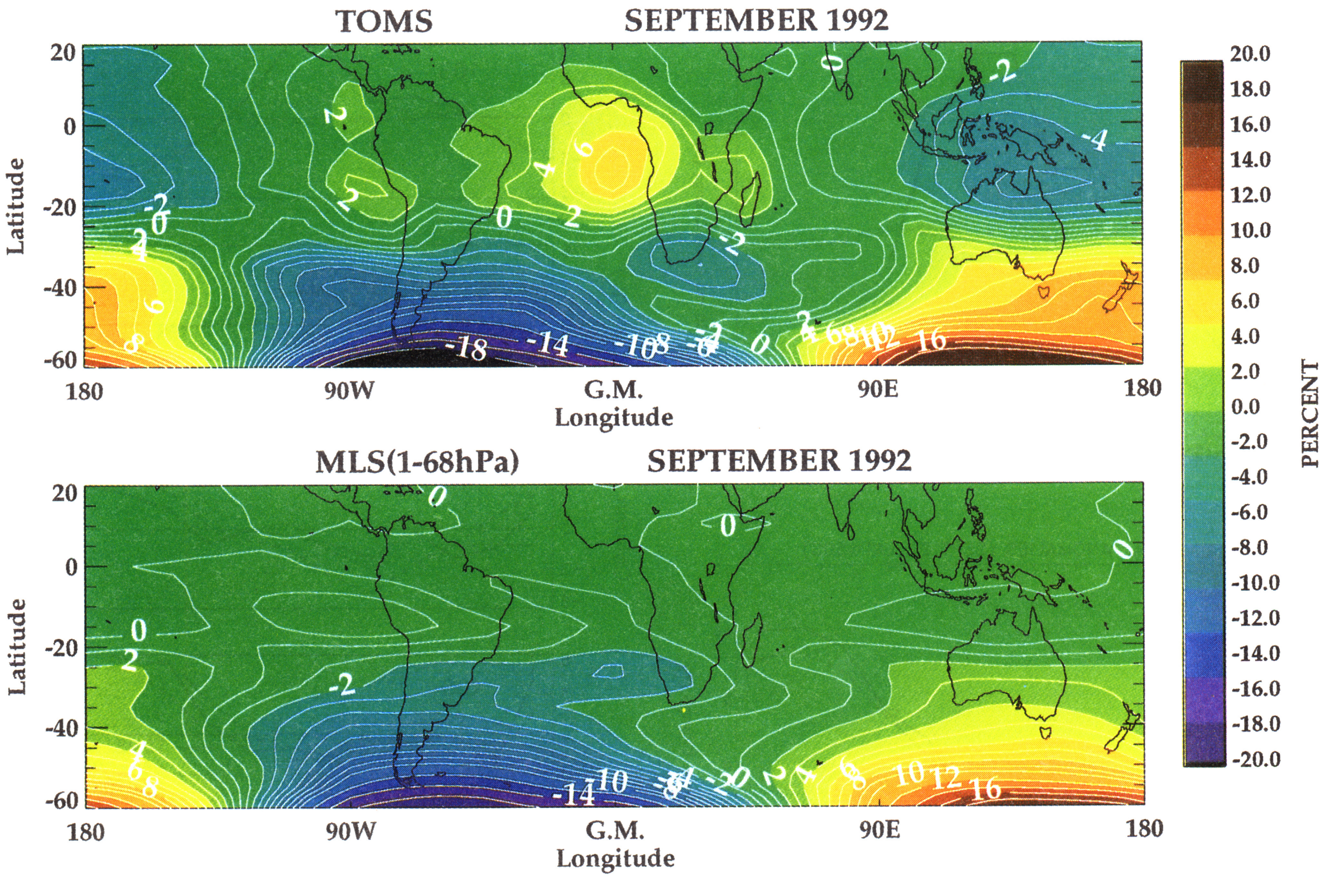

Plate 1. Anomaly fields of version 6 TOMS total ozone (top) and MLS 1 to 68-hPa column ozone (bottom) for September 1992. These monthly averages for both TOMS and MLS were derived by averaging using only MLS measurement days (100% equatorward of 35°, 70% elsewhere). Anomalies were derived by first subtracting the zonal mean from each time series and then dividing the resulting values by the zonal mean. Values shown are in percent. Contour intervals for percentages between -10 and +10 go by 1%, and by 2% for percentages greater than +10% or less than -10%.

anomaly found in stratospheric column ozone. (SAGE stratospheric column ozone was derived by vertically integrating SAGE mixing ratio measurements from 16.5 to 60.5 km.)

The SH wave 1 structures in September 1992 are examined further in Figure 1 with line plots comparing 50-hPa lower stratospheric temperatures and SBUV2 total ozone with similar plots of TOMS and MLS column ozone. Data with 5° by 15° grid structures were rebinned to 10° by 30° blocks to match grid spacings of SBUV2. We note that even with the large 10° by 30° gridding, errors induced by low marine stratus clouds in the South Atlantic may still be present in both version 6 TOMS and SBUV2 total ozone because of the ubiquitous nature of these clouds. At 10°S (Figure 1, top), TOMS and SBUV2 total ozone measurements are seen to track each other remarkably well. If the strong tropical wave 1 anomaly in TOMS data originated from stratospheric ozone, we would expect to see at least some evidence of a corresponding wave 1 structure in either lower stratospheric temperature or MLS column ozone, which is not the case. In lower stratospheric temperatures, either radiative coupling with ozone or vertical advection would likely yield a similar TOMS wave 1 pattern. Both 50-hPa temperatures and MLS stratospheric column ozone in Figure 1 show no apparent wave 1 structure.

The same four atmospheric variables from Figure 1 (top) are also plotted at 50°S (Figure 1, bottom). Zonal structures of all quantities are seen to track each other remarkably well at 50°S, indicating that in higher latitudes the wave anomalies in total ozone are primarily of stratospheric origin and are controlled by stratospheric dynamics [*Wirth*, 1993].

We note again that at altitudes below 46 hPa the ozone retrievals of MLS become less reliable; nevertheless, sensitivity of MLS 1 to 68-hPa column ozone should allow detection of major stratospheric wave structures. Figure 2 shows line plots of SH MLS 1 to 68-hPa column ozone alongside TOMS and SBUV2 total ozone for August 1992. At 10°S (top frame), the total ozone wave 1 anomaly in TOMS and SBUV2

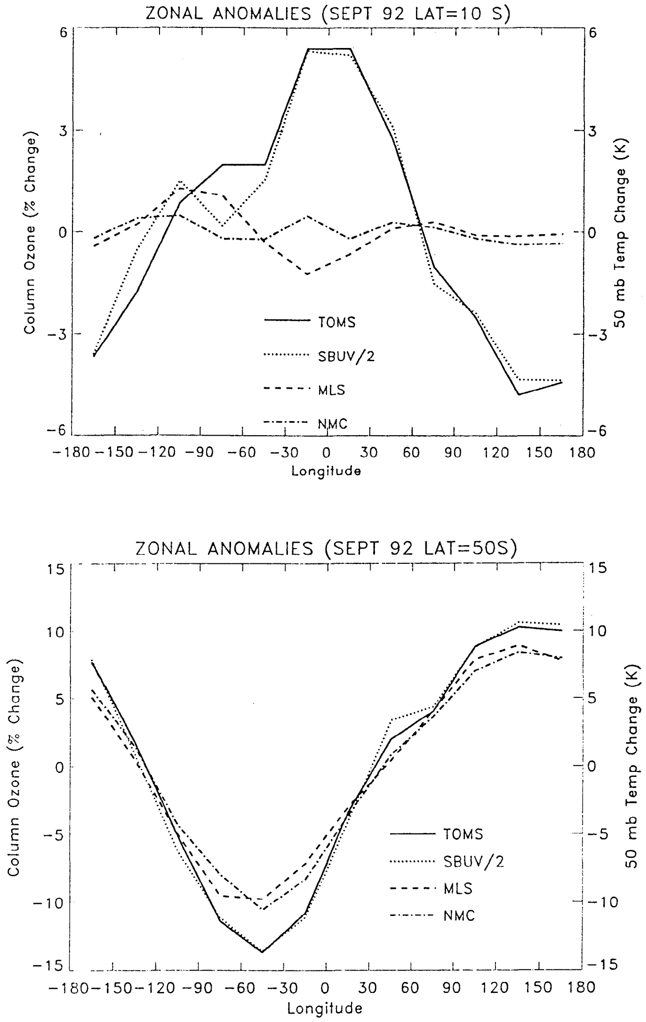

Figure 1. Line plots (longitude series at fixed latitude) of version 6 TOMS and SBUV2 total ozone, MLS 1 to 68-hPa column ozone, and 50-hPa temperature zonal anomalies for September 1992. Anomalies for ozone were derived by first subtracting the zonal mean from each time series and then dividing the resulting values by the zonal mean (values in percent). Anomalies for temperatures were derived by subtracting zonal mean (units in kelvins). Top frame shows 10°S and bottom frame shows 50°S.

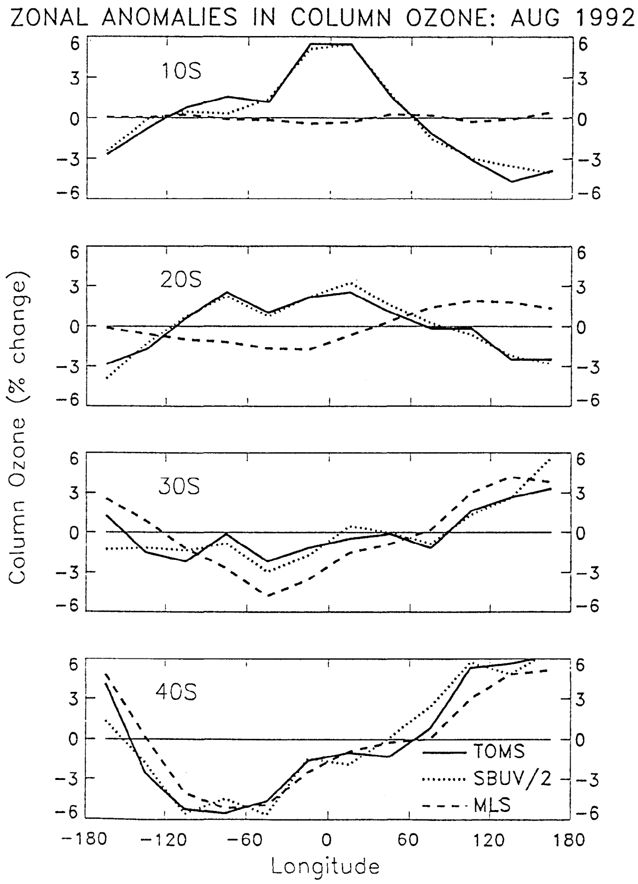

Figure 2. Line plots of zonal anomalies in southern hemisphere (latitudes shown) version 6 TOMS and SBUV2 total ozone and MLS 1 to 68-hPa column ozone zonal anomalies for August 1992. Anomalies are computed as in Figure 1 and are in percent.

is 5–6% of the zonal mean, while MLS column ozone shows amplitudes close to zero. At higher latitudes, MLS shows a gradually developing wave pattern that at 40°S, approximately matches TOMS and SBUV2 anomalies at around 5–6%. The MLS wave pattern in Figure 2 from low to higher latitudes is probably a real feature, implying that zonal wave sensitivity of stratospheric MLS column ozone is conservatively ~2–3% (seen around 20°–30°S). Assuming similar value in the tropics, MLS should easily detect the strong 5–6% total ozone anomaly if it were present in the stratosphere. Thus the indication from Figure 2 is that the tropical wave 1 structure in total ozone originates from ozone below the levels measured by MLS.

The Stationary Tropical Wave 1 in Ozone

The tropical wave 1 anomaly is observed in all seasons in total ozone, but it is never seen in MLS stratospheric column ozone. In TOMS and SBUV2 total ozone, the wave appears to be a permanent structure modulated by seasonal increases in September–October–November (SON) associated with intense biomass burning in Africa and South America around July–October each year.

Plate 2 shows seasonal (3-month) partitions of low-latitude version 7 TOMS and 1 to 68-hPa MLS column ozone anomalies (zonal means removed) using data from September 16, 1991, through April 30, 1993. Version 7 TOMS data include correction for low marine stratus cloud errors and were used in Plate 2 in an effort to provide better comparisons with ozonesonde measurements (discussed in the next section) in the tropics. For conciseness, only equinox seasons March–April–May (MAM) and SON are shown in Plate 2. (Observed amplitudes of the tropical wave 1 in total ozone in the South Atlantic region are largest (smallest) during SON (MAM).)

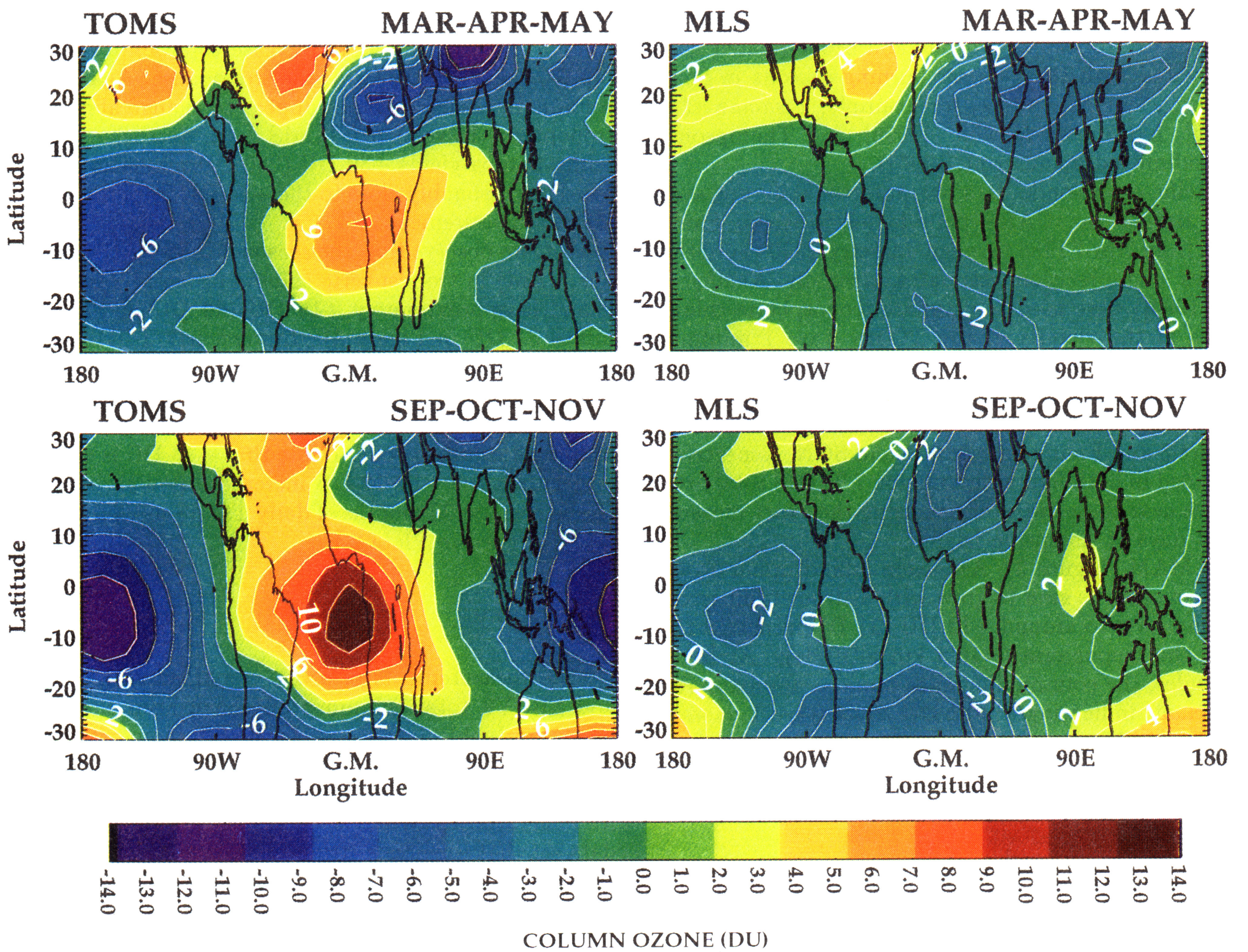

Plate 2. Seasonal (3-month) partitions of low-latitude preliminary version 7 TOMS total ozone (left-column frames) and 1 to 68-hPa MLS column ozone (right-column frames) anomalies. Anomalies represent ozone fields following subtraction of zonal means. Data used are from September 16, 1991, through April 30, 1993. Units are Dobson units. Contour intervals for TOMS are 2 Dobson units. Contour intervals for MLS are one half the values for TOMS.

The persistent tropical wave 1 anomaly in TOMS data in Plate 2 (left column frames) maximizes around 0° longitude and 5°–10°S in the South Atlantic with similar zonal structures in both seasons. Corresponding minima are seen to occur in the tropical South Pacific sector near the date line. Even during MAM, this structure exhibits a clear wave 1 pattern with extreme minimum and maximum values ~-7 and +8 DU, respectively. In SON, extreme values of the wave grow to around -12 and +13 DU. The stronger wave structure in southern spring coincides with intense (potentially ozone-producing) biomass burning over Africa and Brazil [*Fishman et al.*, 1991, 1992].

Ozonesonde Measurements and the Tropical Wave 1

Our study has indicated that the tropical wave 1 anomaly is caused by ozone at altitudes below the levels measured by MLS. We now look to ozonesonde data to further determine contributions from ozone lying below the lowest MLS level (~68 hPa).

After zonal mean measurements are added to the TOMS wave fields in Plate 2, the tropical wave 1 maximum near 0° longitude is found to be approximately +10–12 DU larger in SON than in the MAM season. We compared this seasonal increase with tropospheric ozonesonde data from Ascension Island (8°S, 15°W), which lies near the TOMS maximum. In addition, we examined the intriguing zonal wave 1 structure in MAM and SON seasons by comparing ozonesonde data from Samoa (14°S, 170°W) with Ascension Island data.

Tropospheric column ozone at Ascension Island for the MAM season was estimated by averaging 16 days of vertically integrated (from ground level) ozonesonde ozone mixing ratio data from 1991 through 1992. For SON, tropospheric column ozone was derived from 20 ozonesonde measurements during the TRACE A (Transport and Atmospheric Chemistry near the Equator–Atlantic) campaign in September–October 1992 and 6 days in 1990 and 1991. TRACE A was designed to investigate tropospheric ozone and ozone-related gases associated with the biomass burning seasons in South America and Africa (see, for example, *Fishman et al.* [1996], *Krishnamurti et al.* [1996], and Thompson et al. (submitted manuscript, 1996)).

Pacific ozonesonde data are unfortunately not available during the UARS period; nevertheless, Samoa data from April 1986 through January 1990 (published means and precision estimates given by *Komhyr et al.* [1994]) were used to estimate tropospheric column ozone in the vicinity of the TOMS wave 1 minimum (around 5°–10°S and 170°W in Plate 2).

Comparisons between tropospheric column ozone measurements at Samoa and Ascension Island are shown in Figure 3 and Figure 4 for MAM and SON, respectively. Included in both figures are version 7 TOMS total ozone values near these two stations using the exact measurement days at Ascension Island.

In MAM (Figure 3), Ascension Island minus Samoa column ozone is seen to grow with altitude from zero at ground level to around +12 DU (31 DU minus 19 DU) near the tropopause (~16.5 km altitude). At higher altitudes, the column difference remains approximately invariant, indicating a +12 DU zonal wave 1 anomaly during MAM caused by tropospheric ozone. In comparison, TOMS data show approximately +13 DU (264 DU minus 251 DU) in total column.

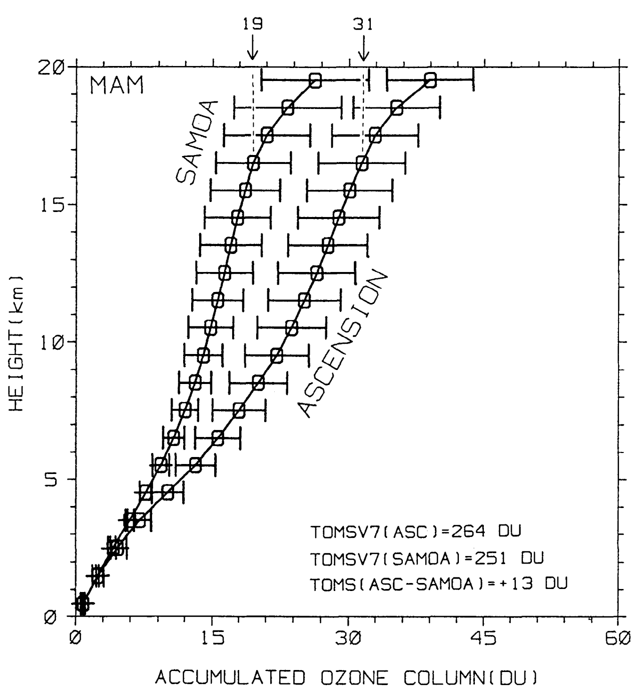

Figure 3. Vertical profiles of accumulated column ozone (Dobson units) measured upward from ground level for Ascension Island (8°S, 15°W) and Samoa (14°S, 170°W) during the March-April-May season (see text). Horizontal bars indicate ±one standard deviation about the mean. Concurrent TOMS version 7 values (and their difference) are indicated near Ascension Island and Samoa using ozonesonde measurement days at Ascension Island.

In SON (Figure 4), Ascension Island minus Samoa column ozone station data increase to around +24 DU (49 DU minus 25 DU) near the tropopause. Again, this column difference does not appear to change above the tropopause. In version 7 TOMS data, Ascension Island minus Samoa yields +30 DU (280 DU minus 250 DU), which is reasonably close to the +24 DU from the sonde station measurements.

Last, we observe that the seasonal increase (SON minus MAM) in tropospheric column ozone at Ascension Island is approximately +18 DU (49 DU in Figure 4 minus 31 DU in Figure 3). In version 7 TOMS data, SON minus MAM total ozone at Ascension Island yields around +16 DU (280 DU minus 264 DU). Again, there is close agreement between TOMS total ozone and tropospheric column ozone. (Note that +16 DU is slightly larger than the +10–12 DU increase stated earlier, but this is because 20 months of daily TOMS measurements were included in Plate 2.)

Summary

MLS measurements of ozone combined with TOMS and SBUV2 total column ozone have been shown to be an invaluable data source for determining properties of global stratospheric (and potentially tropospheric) ozone, giving nearly daily coverage throughout the tropics. This study provides the first evidence from a UARS data set that the South Atlantic tropical wave 1 peak in total ozone is caused primarily by tropospheric effects (dynamics coupled with biomass burning).

MLS 1 to 68-hPa stratospheric column ozone in middle and high latitudes during times of large plane-

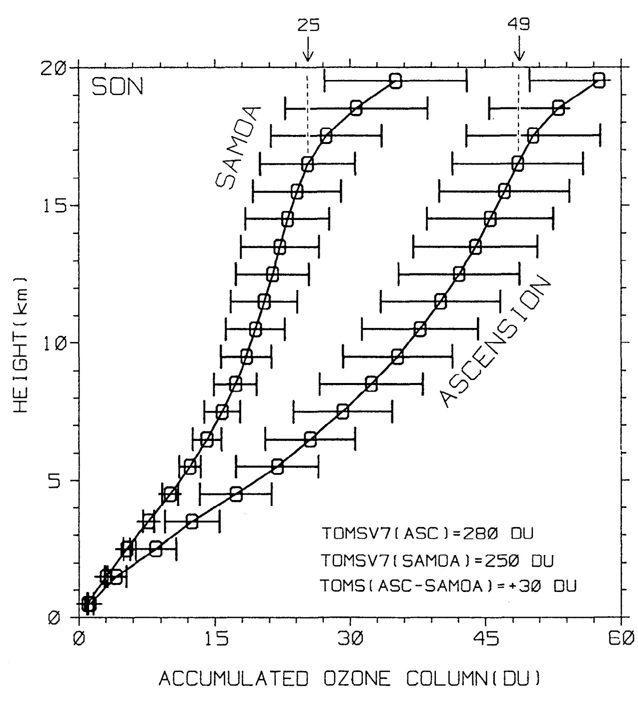

Figure 4. Same as Figure 3 but for the September-October-November season.

tary wave events (southern spring shown) explains most (around 50–80%) of TOMS and SBUV2 wave anomalies. Zonal signatures of TOMS, SBUV2, MLS column ozone, and lower stratospheric temperatures in the middle to high latitudes all showed coherent zonal structures, indicating the dominance of stratospheric effects.

Ozonesonde data from tropical stations in the South Atlantic (Ascension Island) and western Pacific (Samoa) provide additional evidence that the persistent tropical wave 1 in total column ozone is formed largely by the horizontal distribution of tropospheric ozone. In MAM (SON), the Ascension Island minus Samoa tropospheric column ozone difference was determined to be +12 DU (+24 DU). The residual increase (+12 DU) from MAM to SON may be associated largely with the biomass burning seasons in Africa and South America, but further study is necessary to establish the relative role of dynamics in these regions.

Persistence of the tropical wave 1 structure in total ozone in all seasons suggests dynamical processes that need further investigation. In order to understand the relative role of biomass burning, it will be neccessary to monitor tropospheric column ozone from satellite. However, ground-based ozone measurements (currently very sparse) are necessary to determine vertical profiles of tropospheric ozone.

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S. Chandra, D. P. McNamara, A. M. Thompson, and J. R. Ziemke, NASA Goddard Space Flight Center, Mail Code 916, Greenbelt, MD 20771. (e-mail: chandra@chapman.gsfc.nasa.gov; mcnamara@caiman.gsfc.nasa.gov; thompson@gator1.gsfc.nasa.gov; ziemke@jwocky.gsfc.nasa.gov)

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