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Document type: Article **Language:** English **Cited References:** 57 **Times Cited:** 17**Abstract:**

Tropospheric ozone data measured by ozonesondes during the Transport and Atmospheric Chemistry Near the Equator-**Atlantic** (TRACE A) field mission and the multiyear pre-TRACE A program are analyzed jointly with tropospheric ozone amounts derived from remote satellite data ("residuals"). We present here the first detailed analysis of the entire Ascension Island pre-TRACE A data set. Data from the three pre-TRACE A ozonesonde sites are used to establish a coherent spatial and temporal climatology of ozone in the southern tropical **Atlantic** region. This analysis shows a significant ozone seasonality over the **Atlantic** region, with a period of maximum values that extends from the austral winter through at least October at Natal, Brazil, and Ascension Island. Concentrations begin to decline somewhat earlier at Brazzaville, Congo, especially at lower altitudes. Although Natal exhibits a significantly lower annual average than Ascension Island or Brazzaville by about 4 Dobson Units (DU), the magnitude of the seasonal amplitude at Natal is the largest of the three stations. Additionally, more of the seasonal amplitude at Natal is due to a contribution from ozone in the middle and upper troposphere than at either Ascension Island or Brazzaville. Amplitudes as large as 15 DU are measured at individual sites, and the residuals show an average amplitude over the southern tropical **Atlantic** region of 10-12 DU. Statistical comparison of the residuals to the ozonesonde climatology show that while the residuals tend to underpredict both the means and the seasonal amplitudes compared to the in situ data, they provide a good representation of the variance of ozone in this region and predict the local annual and seasonal means to within better than 10% and seasonal amplitudes to within 15%.

KeyWords Plus:

OBSERVATORY PHOTOCHEMISTRY EXPERIMENT, BIOMASS-BURNING EMISSIONS, TROPOSPHERIC OZONE, SURFACE OZONE, SEASONAL-VARIATIONS, CARBON-MONOXIDE, SATELLITE, O3, CONVECTION, AMAZONIA

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Analysis of the distribution of ozone over the southern Atlantic region

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Abstract. Tropospheric ozone data measured by ozonesondes during the Transport and Atmospheric Chemistry Near the Equator–Atlantic (TRACE A) field mission and the multiyear pre-TRACE A program are analyzed jointly with tropospheric ozone amounts derived from remote satellite data (“residuals”). We present here the first detailed analysis of the entire Ascension Island pre-TRACE A data set. Data from the three pre-TRACE A ozonesonde sites are used to establish a coherent spatial and temporal climatology of ozone in the southern tropical Atlantic region. This analysis shows a significant ozone seasonality over the Atlantic region, with a period of maximum values that extends from the austral winter through at least October at Natal, Brazil, and Ascension Island. Concentrations begin to decline somewhat earlier at Brazzaville, Congo, especially at lower altitudes. Although Natal exhibits a significantly lower annual average than Ascension Island or Brazzaville by about 4 Dobson Units (DU), the magnitude of the seasonal amplitude at Natal is the largest of the three stations. Additionally, more of the seasonal amplitude at Natal is due to a contribution from ozone in the middle and upper troposphere than at either Ascension Island or Brazzaville. Amplitudes as large as 15 DU are measured at individual sites, and the residuals show an average amplitude over the southern tropical Atlantic region of 10–12 DU. Statistical comparison of the residuals to the ozonesonde climatology show that while the residuals tend to underpredict both the means and the seasonal amplitudes compared to the *in situ* data, they provide a good representation of the variance of ozone in this region and predict the local annual and seasonal means to within better than 10% and seasonal amplitudes to within 15%.

1. Introduction

Early measurements of tropospheric ozone (O_3) generally supported the perception that its concentrations reach a minimum in equatorial regions and are larger in the northern hemisphere (NH) than the southern hemisphere (SH) [Chatfield and Harrison, 1977; Fabian and Pruchniewicz, 1977; Fishman *et al.*, 1979; Oltmans, 1981]. Until recently, there was also a general assumption that tropical O_3 concentrations were zonally symmetric. Measurements from interhemispheric aircraft flights during the 1960s and 1970s, however, recorded upper tropospheric concentrations that were between 2.5 and 5 times higher over the tropical Atlantic than over the tropical Pacific [Machta *et al.*, 1969; Pratt and Falconer, 1979; Newell and Wu, 1985]. While these authors argued that the difference was not necessarily a persistent feature based on the observed variability of O_3 in this region, the measurements were perhaps a first hint that the tropical O_3 distribution was more complicated than had been historically perceived. Additional indications of the complex nature of tropical O_3 were given by results

from a NASA aircraft mission flown down the west coast of South America during October and November of 1982 [Gregory *et al.*, 1984] and from ozonesonde data at Natal, Brazil [Kirchhoff, 1984; Logan and Kirchhoff, 1986] which showed evidence of larger O_3 concentrations in the southern tropics than in the northern tropics. Recent analyses of satellite measurements provide a new global-scale perspective on the distribution of O_3 and indicate that, unlike the earlier supposition, tropospheric O_3 exhibits a strong heterogeneity across the tropics on both spatial and temporal scales [Fishman *et al.*, 1990].

The satellite-derived distribution of the column amount of tropospheric O_3 (termed the tropospheric residual) shows very large amounts of column O_3 over the tropical Atlantic during the austral spring (Figure 1). Measurements of tropospheric carbon monoxide (CO) from the Measurement of Air Pollution From Satellite (MAPS) instrument on board the space shuttle during October also show enhanced concentrations of this pollutant over the tropical Atlantic [Reichle *et al.*, 1986, 1990; V.S. Connors, NASA Langley, personal communication, 1995]. Various studies have suggested that large-scale biomass burning contributes to the pollution of this region with concomitant high concentrations of O_3 [Seiler and Crutzen, 1980; Logan and Kirchhoff, 1986; Fishman *et al.*, 1990; 1991].

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Figure 1. Seasonally averaged tropospheric ozone residuals (TOMS/SAGE method) in Dobson units: (a) December–February, (b) March–May, (c) June–August, and (d) September–November. Also shown are the locations of the pre-TRACE A ozonesonde sites. “A” is Ascension Island, “B” is Brazzaville, Congo, and “N” is Natal, Brazil.

In response to this finding, several recent field measurement campaigns were held to assess the role of biomass burning in the chemistry of the tropical atmosphere [e.g., Setzer *et al.*, 1991; Fontan *et al.*, 1992]. Additionally, although the NASA Arctic Boundary Layer Expedition (ABLE) 2A and 2B missions were not designed to focus on the various impacts from biomass burning, data obtained during those missions have proven to be valuable in assessment of this issue [Andreae *et al.*, 1988; Browell *et al.*, 1988; Richardson *et al.*, 1991]. The TRACE A (Transport and Atmospheric Chemistry near the Equator–Atlantic) field mission was sponsored by NASA and was held during part of the southern hemisphere burning season in September and October of 1992 [Fishman *et al.*, this issue (b)]. In an effort to characterize the background atmosphere of the Atlantic Ocean and adjacent areas prior to the mission, a pre-TRACE A program provided for several years of ozonesonde measurements at three stations within and bordering the tropical southern Atlantic Ocean: Ascension Island (8° S, 15° W), Brazzaville, Congo (4° S, 15° E), and Natal, Brazil (6° S, 35° W), which was coordinated with the Brazilian space agency Instituto Nacional de Pesquisas Espaciais (INPE). The locations of these sites are marked in Figure 1d. Measurements were obtained from July 1990 through October 1992 at Ascension Island and from June 1990 through October 1992 at Brazzaville and Natal. A preexisting INPE/NASA program has provided for ozonesonde launches at Natal, Brazil, since 1978 [Kirchhoff *et al.*, 1991], which are used here in addition to the TRACE A and pre-TRACE A measurements. While Fishman *et al.* [1992] presented the first year of data at Ascension Island (June 1990 to June 1991), we present here the complete data set obtained at that site (June 1990 to October 1992). Kirchhoff

et al. [this issue] and Nganga *et al.* [this issue] present analyses of ozonesonde data primarily focused on the few weeks during the TRACE A mission.

We investigate the ozonesonde data at these three sites jointly with the satellite residuals to derive a coherent picture of the large-scale spatial and temporal climatology of O₃ across the tropical Atlantic. This analysis allows for the measurements of various trace species obtained during TRACE A (detailed by several papers in this issue) to be viewed in the context of this climatology. Additionally, we discuss possible influences on O₃ seasonality due to tropical biomass burning emissions.

2. Ozonesonde Data

All the pre-TRACE A ozonesonde sites used an electrochemical concentration cell (ECC) instrument on the ozonesondes, which is based on the oxidation reaction of O₃ with potassium iodide. An electrical current proportional to the concentration of O₃ in air pulled through the instrument is generated between the cathode/anode assembly [Komhyr, 1969; Komhyr and Harris, 1971]. All instruments were calibrated at Wallops Island, Virginia, before being shipped, and were checked in the field against a Dasibi UV instrument prior to flight. An ozone instrument intercomparison has shown that electrochemical cell (ECC) sondes are accurate to about 10% compared to a National Bureau of Standards ultraviolet photometer in the laboratory, and in the troposphere, a precision of about 7% to 10% can be expected [Barnes *et al.*, 1985; Hilsenrath *et al.*, 1986].

Table 1 shows the total number of launches each month that

Table 1. Number of Ozone-sonde Launches Used in This Analysis

	Total	Jan.	Feb.	March	April	May	June	July	Aug.	Sept.	Oct.	Nov.	Dec.
Natal, Brazil Nov 1978–Dec 1992	289	23	19	20	22	16	14	30	29	32	34	30	20
Ascension Island July 1990–Oct 1992	57	6	5	5	7	4	—	2	5	8	7	2	6
Brazzaville, Congo June 1990–Oct 1992	78	3	5	2	3	5	7	5	11	9	16	7	5

are used here. At Ascension Island, the number of observations throughout the year is fairly evenly distributed with the exception of the period June through July. The number of launches per week was enhanced during the TRACE A mission, resulting in the relatively large number of measurements at Brazzaville and Natal during the austral spring. Most of the ozone-sonde measurements were made in the local middle to late morning hours (0800 to 1200 LT).

2.1. Integrated Tropospheric Ozone

The column O₃ amount integrated from the surface to the tropopause for each launch is shown in Figure 2. Monthly averages are marked by the horizontal lines, and ± 1 standard deviation is shown by the vertical bars. The tropopause is determined as the altitude where the temperature lapse rate

measured by attached radiosondes decreases to $\leq 2 \text{ K km}^{-1}$ for at least two signals (about 0.5 km). Cases flagged by unusually high tropospheric O₃ amounts greater than 50 Dobson Units (DU, $1 \text{ DU} \approx 2.69 \times 10^{16} \text{ mol cm}^{-2}$) and cases where the calculated tropopause height is significantly different from 100 mbar are individually analyzed and appropriately adjusted to ensure that a proper representation of the tropospheric component of O₃ is achieved. The average tropopause heights for the stations are near 100 mbar and are listed in Table 2. A visual interpretation of the data in Figure 2 shows a trend of decreasing O₃ from the austral spring (roughly August–October) to the fall (March–May) at all three sites.

The ozone-sonde total means and standard deviations are listed in Table 2. The data show a good deal of variability, with standard deviations equal to 20% to 30% of the total means. An annual mean at each station, also listed in Table 2, is calculated by equally weighting the monthly means, using a three-point linear interpolation from the two surrounding months for months with less than two measurements. The annual means at Brazzaville and Natal are lower than the total means by about 5% due to the disproportionately large number of observations during the austral spring when O₃ concentrations are greatest, while differences at Ascension Island are less than 1%. The annual means at these stations are quite large, especially at Ascension Island and Brazzaville, where values are about 40 DU. In contrast, the annual average of tropospheric O₃ from 2 years of ozone-sonde data at American Samoa (14° S, 171° W) is 23 DU, and the tropospheric O₃ content is about 30 DU at Hilo, Hawaii (20° N, 155° W) [Komhyr et al., 1989; Oltmans et al., 1989].

Analysis of these data using a standard student T test to determine the significance of differences between means of two independent sample populations shows that a characterization of temporal behavior on a monthly scale is not appropriate, largely due to the high variance in the data. To determine if a significant seasonal signal can be detected, the data must be grouped into larger populations. March through May (MAM) data at each site are designated as the seasonal minimum period, for which the means and standard deviations are shown in Table 3.

A definition of a consistent period of seasonal maximum concentrations is less obvious because there appears to be a phase shift in the maximum season from station to station (see Figure 2). Figure 3 shows the percentage of observations per month that are greater than the annual mean at each station. At Natal, the percentage is greater than 60% from August through December. Analogous periods are July through October at Ascension Island and June through September at Brazzaville. In all cases, these months also represent the highest monthly means at the stations. This criterion is used to determine the period defined as the station-dependent seasonal maximum. The means and standard deviations during

Figure 2. Integrated ozone from the surface to the tropopause in Dobson units for each ozone-sonde launch at (a) Natal, (b) Ascension Island, and (c) Brazzaville. The "X" shows individual measurements. Horizontal lines and vertical bars connected with the dashed lines show monthly means and the envelope of plus/minus one standard deviation. The horizontal axis shows months of the year from June through May. All years of data are superimposed.

Table 2. Comparison of Total and Annual Means From Ozonesonde Data and Satellite Residuals

	Tropopause Height	Ozonesondes (Integrated to Tropopause)	Residuals All Sage Data 1979–1981; 1985–1991	Residuals Sage II Data 1985–1991
<i>Total Mean, (s.d.), DU</i>				
Natal	100 mbar 16.5 km	38, (11)	34, (9)	35, (9)
Ascension Island	98 mbar 16.9 km	40, (9)	37, (10)	40, (7)
Brazzaville	100 mbar 16.7 km	42, (8)	40, (10)	41, (10)
<i>Annual Mean, DU</i>				
Natal	99 mbar 16.5 km	36	34	36
Ascension Island	97 mbar 17.0 km	40	37	40
Brazzaville	98 mbar 16.8 km	40	40	42

the maximum season are listed in Table 3. At all three sites, the student T evaluation shows that to a greater than 99% confidence level, the minimum and maximum seasonal populations are statistically distinct.

A seasonal amplitude is defined here as the difference between the seasonal means. Although both the annual and the seasonal means at Natal are significantly lower than at Brazzaville, the amplitude is larger at Natal (15 DU compared to 12 DU at Brazzaville). Possible explanations for this are discussed in section 3.

2.2 Seasonality of O₃ as a Function of Height

Figure 4a shows the seasonal amplitude at each station calculated within 100 mbar pressure slabs throughout the troposphere. The cumulative percentage contribution of these altitude slabs to the total amplitude is shown in Figure 4b. Because there is a negative tendency at the highest level at Brazzaville (200 mbar to tropopause), the calculations are done for O₃ between the surface and 200 mbar to remove the

bias toward greater contributions of the lower altitudes at Brazzaville. At Natal, only 39% of the surface to 200 mbar seasonal amplitude is due to O₃ below 600 mbar, while at Ascension Island and Brazzaville, the fractions are 56% and 60%, respectively. Thus the contribution of upper tropospheric O₃ to the total seasonal change is more pronounced at Natal than at the other two stations.

For the purpose of the following discussion, the data are divided into three altitude levels: the surface layer (surface to 900 mbar), the lower troposphere (900 mbar to 600 mbar), and the middle/upper troposphere (600 mbar to 200 mbar).

Surface layer. Throughout middle and high latitudes in the SH, surface data from remote sites show that O₃ reaches an annual maximum during the austral winter (June–August) and a minimum during the summer [Olmans, 1981; Olmans and Komhyr, 1986; Komhyr et al., 1989; Scheel et al., 1990; Ayers et al., 1992; Olmans and Levy, 1994]. The cause of this seasonality is still not completely understood. Several investigations

Table 3. Comparison of Seasonal Means From Ozonesonde Data and Satellite Residuals

Station	Ozonesondes (Integrated to Tropopause)	Residuals All Sage Data 1979–1981; 1985–1991	Residuals Sage II Data 1985–1991
<i>Seasonal Minimum, (s.d.), DU</i>			
Natal ^a	28, (9)	27, (10)	28, (9)
Ascension Island ^a	32, (6)	30, (9)	33, (5)
Brazzaville ^a	36, (7)	34, (7)	36, (5)
<i>Seasonal Maximum, (s.d.), DU</i>			
Natal ^b	43, (9)	41, (8)	43, (6)
Ascension Island ^c	47, (8)	43, (7)	44, (6)
Brazzaville ^d	48, (7)	46, (10)	48, (10)
<i>Seasonal Amplitude, DU</i>			
Natal	15	14	15
Ascension Island	15	13	11
Brazzaville	12	12	12

^aThree-month average: March–May (MAM)

^bFive-month average: August–December (ASOND)

^cFour-month average: July–October (JASO)

^dFour-month average: June–September (JJAS)

Figure 3. Percentage of monthly observations that are greater than the annual mean. Horizontal axis shows months of the year from January through December. Results are shown for Natal (heavy line, marked by "N"), Ascension Island (thin solid line, marked by "A"), and Brazzaville (dashed line, marked by "B"). The location of 50% is indicated.

have noted the similarity of surface O₃ behavior to that of lower stratospheric O₃ at SH middle latitudes and suggested that the driving mechanism is stratospheric injection at middle latitudes, followed by horizontal transport throughout the hemisphere [e.g., Oltmans, 1981; Oltmans and Komhyr, 1986; Oltmans et al., 1989].

Other studies have investigated photochemical destruction as an additional source of surface O₃ seasonality [Liu et al., 1983; Ayers et al., 1992; Oltmans and Levy, 1992; 1994]. For example, Ayers et al. [1992] concluded from photochemical

analysis of surface O₃ and other reactive species at the middle latitude site of Cape Grim, Tasmania (41° S), that maximum O₃ concentrations from June through August are likely due to annual minima in both water vapor and the radiation required to initiate photochemical O₃ destruction within the marine boundary layer. Oltmans and Levy [1992] suggest that even at the tropical location of American Samoa, variations in cloudiness, solar zenith angle, and total column O₃ indicate a seasonal minimum in solar radiation during the winter (July) which should contribute to a seasonal variation in surface O₃ photochemical destruction with maximum concentrations during the austral winter months (June–August).

The ozonesonde data at Ascension Island examined here show this same seasonality of surface layer O₃, with maximum concentrations during the austral winter and minima during the summer (Figure 5b). Surface layer water vapor measured by the radiosondes at this tropical location is highest during the austral summer (January through March) (3.0–3.6% mixing ratio) and lowest during the winter and early spring (July through October) (1.8 to 2.2%). This general seasonality is reproduced in the 10-year climatology compiled by Oort [1983].

The seasonality of the total photochemical loss is estimated using a simple photochemical box model calculation [Richardson, 1988]. Measured meteorological variables at 900 mbar and species representative of clean, surface level remote air were used as input. For conditions representative of the austral summer (February), the calculated net O₃ lifetime is 7.5 days and during August, it is about 40% longer (10.6 days) due to a decrease in the photolysis rate of O₃ to O(¹D) (about 17% lower) and a decrease in the water vapor mixing ratio (about 35% lower) from February conditions. In contrast, if a mixing depth of ~1 km is assumed, an O₃ deposition rate of 0.05 cm s⁻¹ over the ocean [Aldaz, 1969; Galbally and Roy, 1980; Lenschow et al., 1982] gives it a lifetime against surface destruction of 23 days, more than twice the lifetime suggested by photochemistry.

Data from Ascension Island show an anticorrelation (r) between O₃ and water vapor mixing ratios in the mixed layer equal to -0.58 . Coupled with this significant anticorrelation (to the 99% confidence level), the calculations described above indicate the potential importance of photochemical destruction in determining surface O₃ seasonality in the vicinity of Ascension Island. However, because biomass burning is prevalent during this same period of the late austral winter, especially in the southern tropics of Africa, an influence from transport of tropical biomass burning emissions into the southern Atlantic cannot be completely ruled out. Analysis of surface measurements of species which can be used as combusive tracers (such as CO) are needed to determine the relative source of the seasonality of surface level O₃ at Ascension Island.

At Brazzaville (Figure 5c), a clear statistical description of the seasonality of surface O₃ is less evident, although the maximum season June, July, August, and September (JJAS) average is slightly higher than the minimum season average. Surface O₃ at Natal maximizes several months later during the austral spring (September/October) than at Ascension Island (Figure 5a). This springtime maximum in surface O₃ has also been observed at the tropical continental site of Cuiaba, Brazil (16° S) [Kirchhoff et al., 1989] and over the southern African tropics at Sa da Banderia (15° S) and Windhoek (23° S) [Fabian and Pruchniewicz, 1977]. The anticorrelations between surface layer O₃ and water vapor mixing ratios at Brazzaville

Figure 4. Seasonal amplitudes of ozone in Dobson units within altitude slabs at the three pre-TRACE A ozonesonde stations. The amplitude is defined as the seasonal maximum minus minimum, as defined in the text: (a) amplitude integrated within 100 mbar pressure slabs, and (b) cumulative percentage of tropospheric seasonal amplitude from the surface to 200 mbar. Vertical lines mark the cumulative percentages at 600 mbar.

Figure 5. Ozone data integrated within the surface layer (between the surface and 900 mbar) in Dobson units at (a) Natal, (b) Ascension Island, and (c) Brazzaville. The "X" shows individual measurements. Horizontal lines and vertical bars connected by dashed lines show monthly means and the envelope of plus/minus one standard deviation. The horizontal axis shows months of the year from June through May. All years of data are superimposed.

and Natal are -0.12 and -0.18 , respectively, both of which are not significant. This lack of correlation suggests that processes other than photochemical destruction drive the O₃ behavior at these sites.

Lower troposphere. Figure 6 shows the observed O₃ seasonality within the lower troposphere at the three sites. O₃ concentrations at Brazzaville (Figure 6c) exhibit a pronounced seasonal cycle with maximum values during the austral winter (June–August) and a decrease throughout the spring (September–November) of about 10 DU. From July through September, 100% of the observations are greater than the annual mean, while from November through May that number is less than 15%. Within this altitude slab, the large decreases in the monthly means between August and October are statistically significant to the 99% level of confidence. Data from Natal show that the gradual increase in O₃ concentrations throughout the austral winter and into the spring seen at the surface level (Figure 5a) also occurs in the lower troposphere (Figure 6a).

While local urban pollution may be assumed to be insignificant at these sites due to relatively low population densities and/or dominant flow regimes [e.g., Logan and Kirchhoff, 1986; Cros et al., 1987], the seasonality of lower tropospheric O₃ at the two continental sites is consistent with a seasonal source due to generation of O₃ from emissions from biomass burning. Fires across southwest Africa are prevalent during the austral

Figure 6. Ozone data integrated within the lower troposphere (between 900 mbar and 600 mbar) in Dobson units at (a) Natal, (b) Ascension Island, and (c) Brazzaville. Data are plotted as in Figure 5.

winter from late June–August [Cahoon et al., 1992; Justice et al., this issue] and across the Brazilian cerrado, fires normally maximize from August through October [Kirchhoff et al., 1989; Coutinho, 1990].

Figure 6b indicates that concentrations at Ascension Island are uniformly elevated from July through October in the lower troposphere, unlike the winter maximum at the surface level. Wind fields in the lower to middle troposphere show likely transport of air masses from both Africa and South America into the tropical Atlantic during this period, depending on the latitudinal location of the subtropical anticyclone [Krishnamurti et al. 1993; Thompson et al., this issue]. It is speculated here that this broad period of high O₃ concentrations in the lower troposphere at Ascension Island could be indicative of input of enhanced O₃ from a combination of both African and South American source regions.

Upper/middle troposphere. In contrast to the lower troposphere, the seasonality of O₃ in the upper troposphere is similar at all three sites, with O₃ increasing throughout the austral winter and maximizing during the spring (Figure 7). While this seasonality is relatively coherent with respect to altitude at Natal, there seems to be less of a coupling of the lower and upper tropospheric O₃ seasonality at Brazzaville and Ascension Island. A correlation (r) between the O₃ column amount contained within the lower troposphere to that within the middle/upper troposphere is calculated for each site. This correlation at Natal is 0.80, which is higher than (but not strictly statistically significant from) similar calculations for Ascension Island ($r = 0.63$) and Brazzaville ($r = 0.51$).

Figure 7. Ozone data integrated within the middle/upper troposphere (between 600 mbar and 200 mbar) in Dobson units at (a) Natal, (b) Ascension Island, and (c) Brazzaville. Data are plotted as in Figure. 5.

Previous investigations have suggested that seasonal transport of O₃-rich air from the stratosphere or from altitudes near the tropopause through planetary circulation regimes may impact the upper tropospheric O₃ distribution over the tropical Atlantic [Levy *et al.*, 1985; Krishnamurti *et al.*, 1993]. There has also been speculation within the literature that as convective activity in the troposphere over the equatorial regions begins to migrate slightly southward during the late austral spring, biomass burning emissions advected into regions of this convective activity may be transported into the upper troposphere where photochemical O₃ production potential is increased [Chatfield and Delany, 1990; Pickering *et al.*, 1991; this issue]. This speculation is supported by observations of CO recorded by the MAPS instrument during October of 1984 and again in 1994 which show that high concentrations of this combusive pollutant are widespread across the continents of South America and Africa and over the tropical Atlantic in the free troposphere (about 400 mbar) [Reichle *et al.*, 1986; 1990; V.S. Connors, NASA Langley, personal communication, 1995].

This O₃ analysis alone does not include sufficient information to draw robust conclusions as to the source of the observed upper tropospheric seasonality. However, examination of the ozonesonde data from 200 mbar to the tropopause shows insignificant seasonality at Ascension Island and Natal, and only a slight increase (< 1 DU) from the minimum season value to the August–October average at Brazzaville. This suggests only minimal, or insignificant, effects from transport from the stratosphere or from altitudes near the tropopause due to planetary-scale circulations. Photochemical box model calcu-

lations using data from TRACE A by Jacob *et al.* [this issue] support a dominating effect from in situ photochemical production of O₃ at high altitudes over the Atlantic region and adjacent continents during the austral spring.

Additional mechanistic studies are required to address this issue, and in particular, measurements within other remote tropical areas (e.g., Pacific Ocean) should assist in better clarification of the magnitude of effects from natural processes which are not limited to the Atlantic region (such as stratospheric influence or the effect from Walker-type planetary circulation regimes).

3. Satellite Residual Calculations

The method for deriving the tropospheric O₃ residual from two sets of satellite data is described in detail by Fishman *et al.* [1990]. The residual is calculated by subtracting the stratospheric component of O₃ measured by the satelliteborne Stratospheric Aerosol and Gas Experiment (SAGE) or SAGE II instrument from a near-concurrent total atmospheric amount measured by the Total Ozone Mapping Spectrometer (TOMS) instrument on the Nimbus 7 satellite. The resolution of the calculated O₃ residuals is limited by that of SAGE, which obtains approximately 30 vertical profiles of O₃ from cloud top to 65 km each day through a solar occultation technique each orbital sunrise and sunset. Current research is directed toward refining the residual calculation by using solar backscattered ultraviolet (SBUV) 2 data which provide higher temporal resolution stratospheric O₃ data than that from SAGE [Fishman *et al.*, this issue (a)].

Some recent studies point out potential errors in the TOMS data which may result in artificially elevated total ozone amounts where marine stratus are present [Thompson *et al.*, 1993] or where TOMS may underestimate how much total ozone is present over land under the influence of a heavy pollution event generating very large amounts of ozone [Fishman *et al.*, this issue (a); Kim *et al.*, this issue]. Some of these potential deficiencies in the TOMS data have been addressed in an ongoing effort to reprocess the data for Version 7 of the TOMS archive [McPeters *et al.*, 1994]. During the time of the present analysis, however, Version 7 has not been released, so we utilize derived climatological residual fields using the most recent available TOMS data (Version 6) and information from SAGE and SAGE II. We present here a detailed statistical comparison of these climatological residuals to the complete, multiyear pre-TRACE A ozonesonde data set.

3.1 Station Comparisons

Pre-TRACE A stations. There are no residuals available after June 1991 (using the TOMS/SAGE method) due to the eruption of Mount Pinatubo, so the years of data obtained by the ozonesondes and the residuals do not coincide at Ascension Island and Brazzaville. A comparison between this derived data set and the pre-TRACE A ozonesonde data set described in section 2 is, therefore, dependent on the assumption that the residuals and the ozonesonde data both represent a climatological depiction of O₃. Proceeding under this assumption, Figure 8 shows the calculated monthly mean residuals and ± 1 standard deviation from data obtained over a $6^\circ \times 10^\circ$ box centered over each of the pre-TRACE A sites. The ± 1 standard deviation boundaries from the ozonesonde data are overlaid for comparison. The residual calculations qualitatively predict the seasonality and the variance of O₃

Figure 8. Calculated tropospheric residuals in Dobson units within $6^\circ \times 10^\circ$ boxes centered over (a) Natal, (b) Ascension Island, and (c) Brazzaville. The "R" indicates individual residual calculations. Horizontal lines and vertical bars are residual monthly means and plus/minus one standard deviation. The heavy lines show the plus/minus one standard deviation envelope for the ozonesonde data in Figure 2. The horizontal axis is for months of the year from June through May. All years of data are superimposed.

over these sites. Like the ozonesonde data, however, a monthly analysis using the residuals does not provide statistically significant results and a seasonal grouping is required for time-dependent analysis.

For a more quantitative comparison, Tables 2 and 3 show the annual and seasonal residual means and standard deviations along with those from the ozonesonde data described in section 2. Table 2 shows that other than for the annual mean at Brazzaville which shows good quantitative agreement, the total and annual means calculated from all of the available residual values are consistently less than those from the ozonesonde data by between 5 and 10%. The standard deviations are similar to those from the ozonesonde data, with values between 20 and 30% of the means. The total and annual means calculated from residuals using the SAGE II data only (1985–1991) are in somewhat better agreement with the ozonesonde means (within 6%).

Fishman *et al.* [1993] have shown that the residual technique indicates a temporal increase in tropospheric ozone at southern tropical latitudes. This secular increase is apparent in both the residuals derived from SAGE II between 1985 and 1990, and with the combined SAGE and SAGE II residual database dating back to 1979. As pointed out in Fishman *et al.* [1993], the uncertainty in the measurements precludes the unambiguous identification of a trend in the tropospheric residuals, but

this analysis does provide a possible explanation as to why there appears to be a better agreement between the more recent residuals and the ozonesondes than when the entire residual data set is compared with the Ascension Island and Brazzaville soundings.

The agreement between seasonal means (Table 3) is similar to that of the total mean; in general, the residuals agree with the ozonesonde data to within better than 10% using all residual data and better than 5% using residuals from SAGE II data only. The amplitudes calculated from the residuals and the ozonesonde data (Table 3) are similar at Brazzaville, but at Natal and Ascension Island, the residual calculations underpredict the amplitudes (up to 15% using the complete set of residuals). For all cases, however, the amplitude at Natal is larger than that at Brazzaville.

Examination of Table 3 shows that the relatively greater seasonal amplitude over Natal is primarily attributable to lower values during the minimum season. The lower values could be accentuated by a large wet season sink over the Brazilian rain forest [Fan *et al.*, 1990; Gregory *et al.*, 1990; Jacob and Wofsy, 1990]. Fan *et al.* [1990] estimated that the mean daytime deposition velocity for O₃ just at the forest canopy level was 1.8 cm s^{-1} ($2.3 \times 10^{11} \text{ mol cm}^{-2} \text{ s}^{-1}$) and Jacob and Wofsy [1990] estimated an additional daytime boundary layer photochemical loss of $7 \times 10^{10} \text{ mol cm}^{-2} \text{ s}^{-1}$ during the wet season. Assuming ~ 10 hours of daylight per day, this translates to an O₃ sink equal to 0.4 DU d^{-1} during the wet season. In addition, the minimum season value of O₃ over Brazzaville may be impacted by the occasional transport of relatively high concentrations of O₃ in the harmattan flow toward the southwest from areas of biomass burning in northeast Africa during the late austral summer [Andreae *et al.*, 1992; Lopez *et al.*, 1992].

Pretoria ozonesondes. Figure 9 shows a comparison of the residuals to a 3-year record of ozonesonde data (1990–1993) at a subtropical continental site in Pretoria (Irene), South Africa (25° S , 27° E) [Diab *et al.*, this issue; Thompson *et al.*, this issue]. Like the tropical pre-TRACE A stations, the climatological residuals qualitatively reproduce the seasonality and variance of tropospheric O₃ measured by the ozonesondes at this continental southeast African location. The annual average (35 DU) and seasonal minimum (MAM) average of 30 DU are predicted to within a few percentage points by the residuals. The seasonal maximum period (September through November) is determined here using the same criterion described in section 2.1. The residuals underpredict this seasonal average (40 DU) by about 3 DU (8%).

It is noteworthy that the maximum season at this station is several months later than it is at the west coast site of Brazzaville. Slightly more than half of this seasonal amplitude at Irene is due to O₃ below 5 km. This behavior is likely to be at least partially due to the migration of the large-scale fires from the western coast of Africa during June–August to the east coast maximum during September [Cahoon *et al.*, 1992; Justice *et al.*, 1995].

3.2 Regional Evaluation

Use of the satellite residuals enables a two-dimensional picture of the behavior of tropospheric O₃ as opposed to a piecemeal analysis of individual ozonesonde sites. Figure 10 shows the seasonal evolution of smoothed residuals from the March–May (MAM) average to the June–August (JJA) average, and from JJA to the September–November (SON) average. The

Figure 9. (a) Ozonesonde data at Pretoria (Irene), South Africa integrated from the surface to the tropopause in Dobson units. The "X" shows individual measurements. Horizontal lines and vertical bars connected with the dashed lines show monthly means and the envelope of plus/minus one standard deviation. The horizontal axis is for months of the year from June through May. All years of data are superimposed. (b) Calculated tropospheric residuals within a $6^\circ \times 10^\circ$ boxes centered over Pretoria (Irene) in Dobson units. The "R" indicates individual residual calculations. Horizontal lines and vertical bars are residual monthly means and plus/minus one standard deviation. The heavy lines show the plus/minus one standard deviation envelope for the ozonesonde data in Figure 9a.

residual data used to generate these plots required the application of time and space smoothing in order to ensure global coverage of all grid boxes. The net effect of the smoothing is a slight suppression of seasonal amplitudes, but the large-scale features remain unchanged.

Across the northwestern subtropical Pacific from the March–May average to the June–August average, there is a decrease in tropospheric O₃. Several studies have suggested that tropospheric O₃ in this region is primarily controlled by photochemical destruction [Oltmans, 1981; Liu *et al.*, 1983; 1992; Ridley *et al.*, 1992; Oltmans and Levy, 1994]. The decrease in the residual coincident with the movement of the sun toward northern latitudes is consistent with an increase in photochemical destruction in this area. At the same time, a "plume" of large increase is apparent off the west coast of Africa in the southern Atlantic. A similar increase, although slightly lower in magnitude, is shown off the west coast of South America. From the June–August average to the September–November average, virtually the entire northern tropics is characterized by decreasing O₃, while the southern hemisphere shows consistently increasing O₃. A region of maximum increase is centered over Brazil.

When the period of seasonal maximum that was defined for Ascension Island (July–October) is taken as representative of the entire southern Atlantic Ocean, the unsmoothed residual data averaged over this area (from the equator to 30° S, and from 45° W to 15° E) show a seasonal change of 10 DU from

Figure 10. Seasonal changes in the satellite residuals shown with contours of 2 Dobson units: (a) the change from the March through May average to the June through August average, and (b) the change from the June through August average to the September through November average.

the MAM average. If the estimated up to 15% underprediction of the amplitude by the residuals as suggested by ozonesonde data at Natal and Ascension Island is considered, the seasonal amplitude averaged over the southern Atlantic could be as high as 12 DU.

4. Summary

The complete record of ozonesonde data from the pre-TRACE A sites has been presented, and a unified analysis of this data set has been performed jointly with satellite ozone residuals. Analysis of the statistics between the climatological residuals and the multiyear in situ data show agreement to within better than 10% for seasonal and annual means, and to within better than 15% for amplitudes. This indicates that the residuals may be used to assess the general behavior of O₃ on a regional scale, which is, for the most part, beyond the scope of in situ data.

A significant seasonal variation of tropospheric O₃ over the southern Atlantic of about 10–15 DU was calculated, and the period of maximum values occurs somewhat earlier at Brazzaville than at Natal or Ascension Island. O₃ in the upper troposphere contributes more to the tropospheric amplitude at Natal than is the case for Brazzaville or Ascension Island. Surface water vapor and O₃ data from Ascension Island suggest that photochemical destruction of O₃ may play a significant role in determining surface seasonality there.

The climatological residual fields show that the springtime southern Atlantic O₃ increase is fed by processes increasing O₃ off the west coast of Africa in JJA and then by processes

increasing O₃ over South America in SON. Further, ozone-sonde data indicate that the increase near Africa and into the eastern tropical Atlantic during JJA is likely concentrated at lower altitudes, while the increase which spreads from South America into the tropical Atlantic later in the season is distributed more uniformly throughout the troposphere.

Within the context of this analysis, we determine that the TRACE A mission occurred during the climatological maximum O₃ season over South America (late September), and toward the end, or just after, the climatological maximum O₃ season over western Africa (October). Additionally, enhanced concentrations of O₃ measured in the middle and upper troposphere measured during TRACE A aircraft flights [Browell *et al.*, this issue] are most likely not unique to 1992 and appear to occur on an annual basis.

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