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Enhanced view of the “tropical Atlantic ozone paradox” and “zonal wave one” from the in situ MOZAIC and SHADOZ data

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[1] Ozone vertical profiles from the Measurements of Ozone from Airbus In-service Aircraft (MOZAIC) program over Africa are used to complement pictures of the wave-one pattern and the “tropical Atlantic paradox” identified through soundings in the Southern Hemisphere Additional Ozonesondes (SHADOZ) network. The Atlantic paradox refers to a greater tropospheric ozone column amount over the South Atlantic than the North Atlantic during the West African biomass burning season. SHADOZ and MOZAIC data from 1998–2002 and 1997–2004, respectively, are used to show that these two phenomena are linked. The combined data are used to address the following: Does the (continental) MOZAIC data modify the appearance of the paradox? Do lower tropospheric MOZAIC data lead to new conclusions about ozone in the wave-one maximum region? During December, January, and February (DJF), the lower troposphere over Africa exhibits a higher ozone signal in the burning hemisphere, that is, north of the equator, so the “paradox” does not appear over the African continent. The MOZAIC data set over Africa highlights another component of the wave-one feature when the tropospheric ozone mixing ratio is viewed in zonal cross section. The lower troposphere makes a nonnegligible contribution to the regionally higher ozone column during the biomass burning periods of each hemisphere (DJF) for West Africa and June, July, and August (JJA) for the central Africa region. A southern preference for the wave-one character, previously deduced from satellite data, is confirmed with a stronger maximum in September, October, and November (SON). Both the paradox and wave-one phenomena are consistent with a view that the African continent is a major source of biomass burning and lightning emissions.

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1. Introduction

[2] During the mid-1980s unexpectedly high tropospheric ozone columns were discovered over the South tropical Atlantic with satellite observations [Fishman and Larsen, 1987]. Similar findings were made with shipboard [Smit *et al.*, 1989] and coastal ozone soundings [Logan and Kirchhoff, 1986]. The origins of high ozone in the middle troposphere were ascribed to biomass burning from Africa [Fishman *et al.*, 1991] because the highest Atlantic ozone amount coincides the latter period of the southern Africa

and South America biomass burning season (September–November). The pyrogenic contribution to tropospheric ozone over the South Atlantic and adjacent continents was demonstrated through ground-based and aircraft measurements of ozone and ozone precursors during the 1992 Southern African Fire Atmospheric Research Initiative (SAFARI) and Transport and Atmospheric Chemistry near the Equator-Atlantic (TRACE-A). However, lightning [Thompson *et al.*, 1996; Pickering *et al.*, 1996] and biogenic sources of nitric oxide (NO_x) [Harris *et al.*, 1996; Swap *et al.*, 2003; Jaeglé *et al.*, 2004] can also contribute to ozone formation. Recently, Jaeglé *et al.* [2004] estimated that soils account for 40% of surface NO_x emissions over Africa.

[3] The “Atlantic ozone paradox,” discovered with ozone soundings during the trans-Atlantic Aerosols99 cruise [Weller *et al.*, 1996; Thompson *et al.*, 2000], refers to the persistence of higher tropospheric ozone column amounts over the South Atlantic than over the northern tropical Atlantic, when fire activity is maximum in the Northern Hemisphere, over Africa. The paradoxical differ-

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ence is on the order of 5 Dobson units (DU; $1 \text{ DU} = 2.69 \times 10^{16} \text{ molecules cm}^{-2}$). Satellite records, though somewhat uncertain [Thompson *et al.*, 2000; Martin *et al.*, 2002], indicate that the paradoxical ozone column distribution between the Southern and Northern hemispheres is a persistent occurrence.

[4] A number of papers have attempted to explain the “paradox” phenomenon [Edwards *et al.*, 2003; Jenkins *et al.*, 2003; Jenkins and Ryu, 2004a, 2004b; Chatfield *et al.*, 2004]. Some of them focus on the South Atlantic maximum itself, whereas others investigate why total column is higher in the South Atlantic than in the North. Weller *et al.* [1996] and Thompson *et al.* [2000] noted that aged upper tropospheric or lower stratospheric origins could account for high ozone concentrations measured along the shipboard transect that were extremely dry. Another actor in the “ozone paradox,” though its contribution still needs to be quantified, seems to be the production of ozone precursors by lightning during the Southern Hemisphere rainy season in December, January, and February (DJF), as shown by Thompson *et al.* [2000] and Jenkins and Ryu [2004a, 2004b]. Regional pollution has also been suggested, with cross-equatorial transport of biomass burning products from the Northern Hemisphere during DJF. Upper level distributions of CO pollution plumes south of the ITCZ, as seen by satellite [Edwards *et al.*, 2003], apparently support this hypothesis, as do distributions of Outgoing Longwave Radiation and precipitation rates made by Jenkins and Ryu [2004b]. Jenkins *et al.* [2003] have suggested a westward advection of biomass burning polluted air masses at lower altitudes over the Atlantic, but they did not believe that biomass burning was responsible for elevated ozone in the middle upper troposphere. Satellite tropospheric ozone, TRMM lightning and fire data, along with Ascension sondes, also showed a link between Indian Ocean pollution and the south Atlantic maximum near 300 hPa in January–March 1999 [Chatfield *et al.*, 2004]. No ozone profile measurements except the published marine ones have been used so far to examine the various hypotheses.

[5] On the global scale, Shiotani [1992] observed a zonal wave-one pattern across the entire equatorial zone with a maximum centered over Atlantic and Africa when he examined the total ozone column from the Nimbus7/Total Ozone Mapping Spectrometer (TOMS) satellite. SHADOZ sondes [Thompson *et al.*, 2003b] and TOMS-based products [e.g., Thompson and Hudson [1999] further delineate the tropospheric character of the wave one. The sondes show a zonal pattern with middle upper tropospheric ozone maxima located between 40°E – 60°W . Both SHADOZ and satellite records show that the wave-one pattern appears throughout the year, with a maximum amplitude in September–November (SON) season, where the latter is defined by the difference in column amount between an Atlantic maximum and Pacific minimum. This is demonstrated by Thompson *et al.* [2003b], if we compare the intensity of midtropospheric ozone in their Figures 4a (MAM) and 4b (SON). The satellite record [e.g., Fishman *et al.*, 1991; Thompson *et al.*, 2000] shows a preference of the wave one for the Southern Hemisphere compared to the northern one. However, in situ data over the Northern Hemisphere that could give further insight into the hemispheric contrast have been lacking.

[6] Finally, it is noted that there have been a number of attempts to refine retrievals satellite estimates of tropospheric ozone over Africa and the tropical Atlantic [Ziemke *et al.*, 1998; Thompson *et al.*, 2000; Martin *et al.*, 2002; Fishman *et al.*, 2003; Newchurch *et al.*, 2003; Kim *et al.*, 2005]. They have served to further highlight deficiencies in the in situ observations over equatorial Africa and the Atlantic and in reconciling the ozone record with, for example, Along Track Scanning Radiometer (ATSR) fire counts (<http://dup.esrin.esa.it/ionia/wfa/index.asp>) over the continent.

[7] The SHADOZ network, providing in situ ozone data throughout the tropics has somewhat limited coverage near the ozone maximum at the longitudes where the paradox appears. Furthermore, the data used to depict the zonal wave one in work by Thompson *et al.* [2003a, 2003b] are all in the southern geographical hemisphere, remote from sources; most are coastal or oceanic. The MOZAIC program, with its aircraft measurements over a number of African countries, gives a unique opportunity to fill the gap in this “window” [Sauvage *et al.*, 2005]. The objective of the present study is to merge the MOZAIC, the Aerosols99 and the SHADOZ data for a more comprehensive view of the wave-one and paradox phenomena. Together these observations allow us to address the following questions: Does the continental data change the characteristic of the paradox over the African-Atlantic region? Does the inclusion of MOZAIC data with SHADOZ give new insights into the wave-one maximum? Does the additional data provide new information to guide models and satellite retrievals in this area of complex dynamical and chemical interaction?

2. Data and Methodology

[8] Data for ozone soundings are taken from the Aerosols99 shipboard campaign [Thompson *et al.*, 2000], and from the SHADOZ network archive for the 1998–2002 period [Thompson *et al.*, 2003a]. Coordinates of each of the SHADOZ stations are summarized by Thompson *et al.* [2003b, Table 2]. Further details can be found on the SHADOZ Web site: <http://croc.gsfc.nasa.gov/shadoz/> and the Aerosols99 Web site <http://croc.gsfc.nasa.gov/shadoz/Aerosols99.html>. Aircraft ozone data are taken from the MOZAIC program [Marenco *et al.*, 1998] (see <http://www.aero.obs-mip.fr/mozaic/> for updated information). The MOZAIC program provides data over continental Africa starting in April 1997 up to mid 2004. A detailed climatology of the three most documented stations has been recently presented by Sauvage *et al.* [2005]. For the purpose of looking at the zonal cross section presented below, we use MOZAIC data over Brazzaville to fill in the gap of the SHADOZ data between Ascension Island and Reunion Island (Figure 1). Data are seasonally averaged during December–February (DJF), March–May (MAM), June–August (JJA), and September–November (SON), and from the ground up to 186 hPa, the maximum of the cruising altitude of the MOZAIC flights.

[9] To investigate the Paradox phenomenon, we use MOZAIC data from Namibia (Windhoek, 22.5°S), Angola (Luanda, 13.2°S), Congo (Brazzaville, 4.2°S), Ivory Coast (Abidjan, 5.4°N), Nigeria (Lagos, 6.6°N) up to Senegal

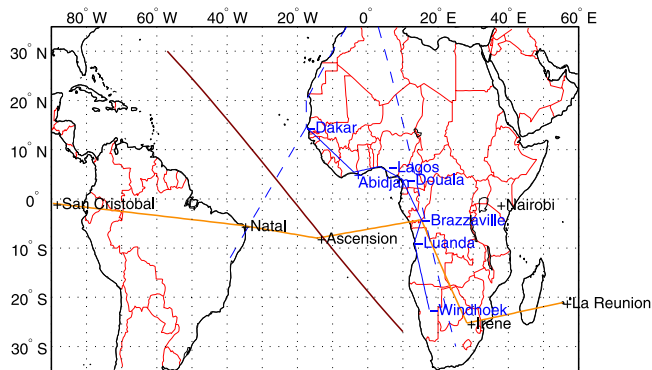


Figure 1. Locations of the MOZAIC (blue) and of the SHADOZ (black) sites in the Atlantic-African region. The brown line represents the locations of soundings during the Aerosols99 campaign. The blue line represents sites used to create the cross section obtained with MOZAIC measurements to investigate the ozone paradox. The orange line symbolizes the cross section made with SHADOZ and MOZAIC measurements to investigate the “zonal wave one.” The dashed blue line represents mean transects of the MOZAIC airplanes from Europe to South America and from Europe to South Africa.

(Dakar, 14.5°N) during the DJF season. This permits us to compute a latitudinal cross section along the Western African coast, parallel to the ship track during the Aerosols99 campaign (Figure 1). We also use the MOZAIC profiles to calculate seasonal tropospheric columns (Lagos, Abidjan and Douala (Cameroon, 4.03°N), merged under the name Gulf of Guinea, and Brazzaville). As the MOZAIC aircraft do not provide measurements above 186 hPa, we use seasonal mean values from the previous TRACE-A soundings between 186 and 100 hPa (the latter considered as the mean tropopause) taken at Brazzaville. For the Gulf of Guinea we use a constant concentration fixed at 70 ppb between 186 and 100 hPa because of the absence of any strong seasonal cycle in the middle and upper troposphere [Sauvage *et al.*, 2005]. This fixed criterion leads only to a 4%–6% difference in the total tropospheric ozone column, if the 70 ppb is substituted with a fixed value of 50 or 100 ppb.

3. Results With In Situ MOZAIC and SHADOZ Observations

3.1. Tropical Ozone Paradox

3.1.1. Continental-Oceanic Contrast

[10] The MOZAIC data allow for the first time a calculation of tropospheric ozone columns over continental Africa, at Lagos, Abidjan, Brazzaville, Luanda, and Windhoek (Figure 2). We also use Ascension (maritime SHADOZ data).

[11] The tropospheric columns averaged over DJF season show 41.5 DU over Lagos, 41.2 DU over Abidjan, 36.7 DU over Brazzaville, 27.2 DU over Luanda, 36.2 DU over Ascension Island and 36.9 DU over Windhoek (1000 hPa to 100 hPa). Thus, for the first time, in situ data regularly recorded north and south of the ITCZ show ozone column

amounts consistent with the location of the emissions sources: greater over the Northern Hemisphere, where emissions from biomass burning maximize at this time of the year. It appears that the paradox does not exist over the African continent. The MOZAIC data highlight the importance of having reliable lower tropospheric ozone amounts to guide ozone column evaluation from models and satellite. Indeed the northern part of the continent, near Gulf of Guinea, is characterized by persistent pollution confined below 700–650 hPa during the fire season [Sauvage *et al.*, 2005]. The ozone column below 700hPa represents respectively 42% (17.3 DU), 41% (17 DU), 32% (12 DU), 26% (7.1 DU), 19.5% (7 DU), and 28% (10 DU) of the tropospheric segments of the aforementioned ozone column amounts. If we omit the lower troposphere (as could occur over a cloud or a dust plume) columns calculated between 700 and 100hPa would be greater over the Southern Hemisphere than over the Northern but not by much (25 DU versus 24 DU). Figure 2 also highlights an asymmetric behavior between the two hemispheres. The weight of the column is definitely in the lower troposphere in the Northern Hemisphere (53% of the total tropospheric ozone column (TTOC) over Lagos because of ozone below 600 hPa) and in the upper troposphere in the Southern Hemisphere (65% of the TTOC over Ascension because of ozone above 550 hPa). Brazzaville appears as a transition zone.

[12] Although tropospheric ozone columns remain higher over Northern Africa (41 DU from MOZAIC data) than in South Atlantic (38 DU calculated with Aerosols99 data), there is still a paradox over the ocean, with tropospheric ozone columns higher in the South than in the North Atlantic (33 DU for the last, calculated with Aerosols99 data).

3.1.2. South Atlantic Maximum

[13] In this section we compare two latitudinal cross sections over the Atlantic ocean and African continent,

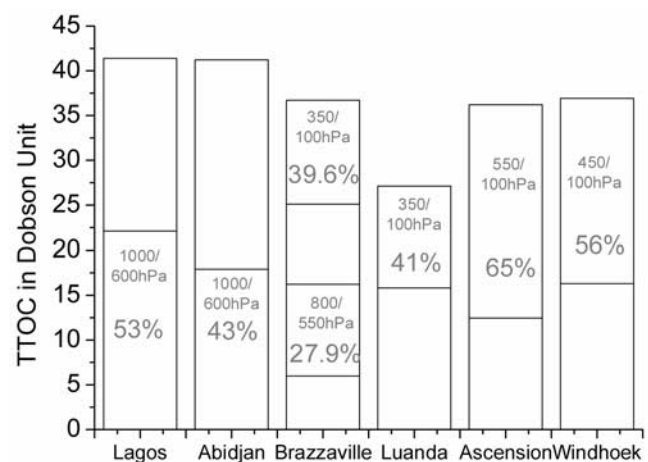


Figure 2. Tropical tropospheric ozone column to 100 hPa in Dobson units (DU) over Lagos, Abidjan, Brazzaville, Luanda, and Ascension Island during December–February (DJF). Between 186 and 100 hPa, for the MOZAIC data, values at Brazzaville are taken from the soundings used in the Trace-A campaign. For the other MOZAIC stations, values are assumed constant at 70 ppbv. The localization of each ozone maxima and its contribution to the total tropospheric ozone column are given in gray.

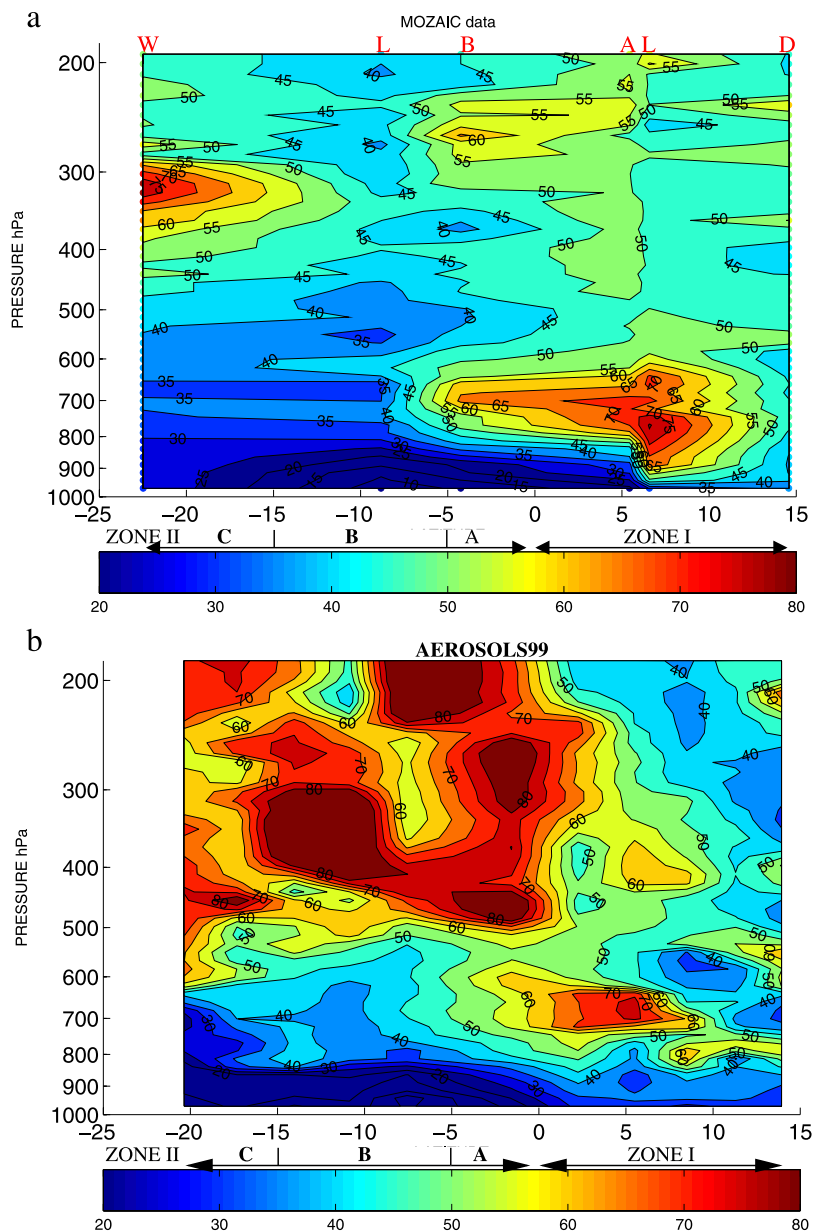


Figure 3. Meridional (22°S – 15°N) cross section (a) as seen by the continental MOZAIC data (D, Dakar; L, Lagos; A, Abidjan; B, Brazzaville; L, Luanda; and W, Windhoek) from the surface up to 200 hPa during DJF and (b) as seen over the Atlantic during the Aerosols99 cruise (soundings from 23 January to 3 February 1999).

addressing these questions: Why does the paradox remain over the ocean? What can explain the contrasting ocean-continental ozone distributions? How do the published interpretations or hypotheses about the paradox apply to the combined SHADOZ-MOZAIC view?

[14] Figure 3a shows for the first time a continental view of the latitude window corresponding to the “Atlantic ozone paradox” on the basis of high-resolution profiles from in situ instruments during DJF season (MOZAIC stations). For comparison convenience, we also use measurements made over the Atlantic during Aerosols99 shipboard campaign [Thompson *et al.*, 2000, Figure 1] represented in Figure 3b. Note that the January–February 1993 trans-Atlantic Polarstern cruise of Weller *et al.* [1996] was nearly identical in

ozone latitudinal distribution to the Aerosols99 cruise [Thompson *et al.*, 2000] even though the Polarstern crossed in the opposite direction, southwest (from South America) to northeast (Europe). Similar results were observed by Polarstern researchers in months of April–May, suggesting that the paradox is a common Atlantic feature, not an anomaly. We look more closely at the latitudes referred to as zones 2 and 3 (I and II in our terminology) by Thompson *et al.* [2000, Figure 1].

[15] The paradox in the oceanic view can be partly explained by the dynamic features over the African continent. The continental-oceanic contrast of ozone in the lower troposphere, between 0° and 8°N (zone I, Figures 3a and 3b) reflects divergent transport features of biomass burning

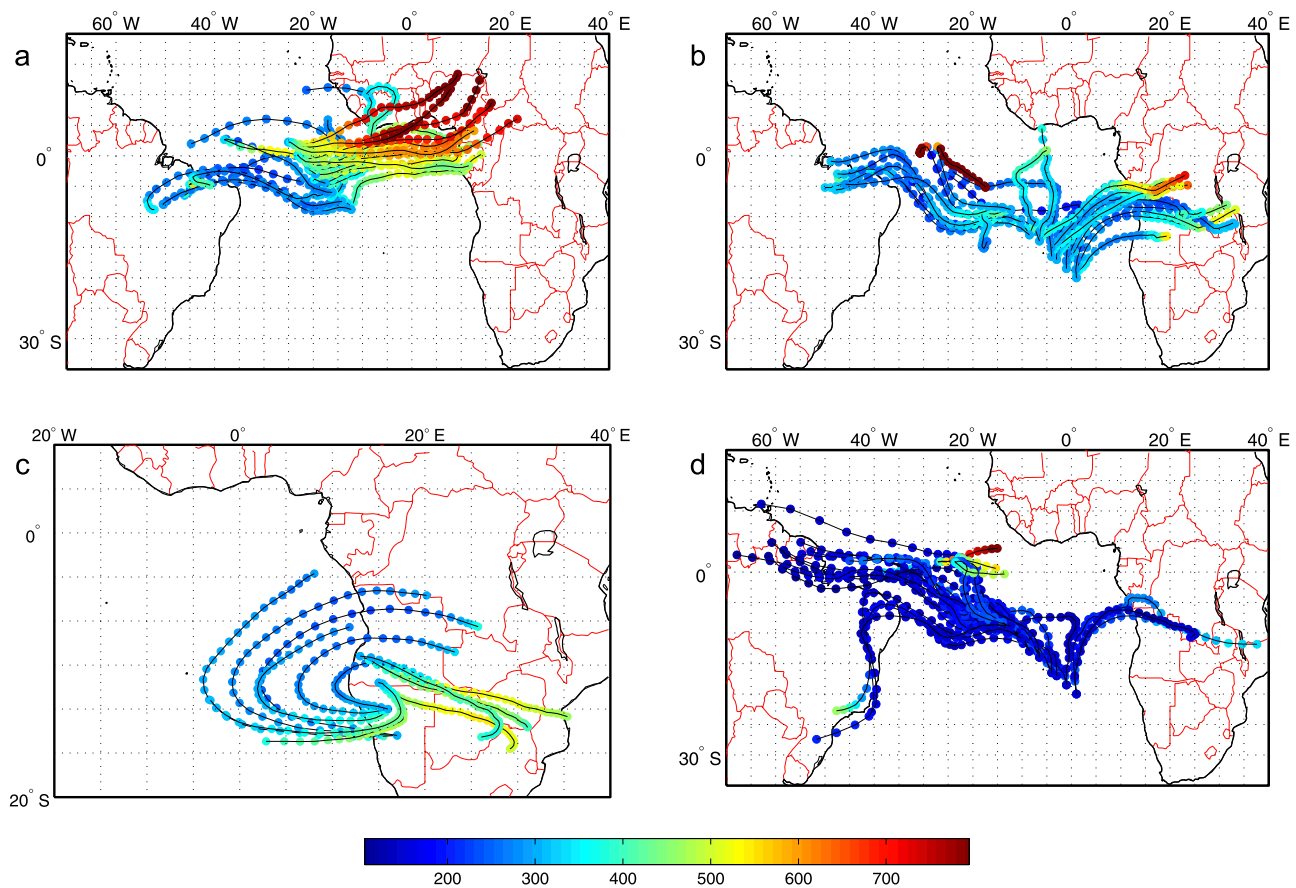


Figure 4. Six-day trajectories in the Aerosols99 transect, computed with the Lagrangian LAGRANTO model [Wernli and Davies, 1997], calculated with European Centre for Medium-Range Weather Forecasts (ECMWF) analysis, in February 1999. Trajectories are color coded with pressure (in hPa).

emissions. Whereas both the southwesterly Harmattan flow and the African Easterly Jet (AEJ) bring high ozone from the ground up to 600hPa over the African continent [Sauvage *et al.*, 2005], only AEJ advection exports high ozone over the north Atlantic, to 35°W, 750–650 hPa (Figure 3b). The Harmattan only advects emissions in a southwesterly flow, with no export toward the North Atlantic so the North Atlantic lower troposphere may be deficient in up to 14 DU ozone.

[16] In addition to differences between lower tropospheric ozone over Africa and the Atlantic, it is striking that an ozone maximum persists over the middle upper troposphere of the South Atlantic and not over the African continent.

[17] Zone II A (0°–5°S, Figure 3b), displays higher ozone concentrations above the South Atlantic middle troposphere near 400–150 hPa than over the continent (Figure 3a) and the North Atlantic. The 6-day backtrajectories initialized in these oceanic maxima show a West equatorial Africa origin in the lower levels ($P = 800$ hPa, Figure 4a), that is, partial Harmattan contribution as the southwesterly Harmattan flow encounters the Intertropical Convergence Zone (ITCZ) just south of the Gulf of Guinea coast. Convection in the vicinity of the ITCZ allows vertical transport of biomass burning emissions through detrainment of polluted air in cloud anvils to reach the South Atlantic middle upper troposphere. Thus 10 DU or more ozone may

be introduced into the South Atlantic in contrast to the North Atlantic. Over the continent, near the Congo (Brazzaville), high ozone concentrations, 60 ppbv at 300–250 hPa (Figure 3a), probably correspond to the detrainment of polluted air masses from the ITCZ that had previously been advected through the Harmattan flow. Figure 4b shows this upward motion over the Brazzaville area. However, a comparison of Figures 3a and 3b (zone II A) shows that the continental maxima is not as strong as over the South Atlantic. One reason is that Brazzaville is in a transition zone at the edge of the ITCZ. Previous studies have implicated lightning influence from South America and/or Central Africa for this part of the South Atlantic maximum [Thompson *et al.*, 2000; Jenkins *et al.*, 2003]. Indeed, lightning activity maximizes over southern African continent at this time of the year (see LIS data, <http://thunder.msfc.nasa.gov/lis/>).

[18] The middle upper troposphere of zone II B depicts the major difference between the Atlantic and continental latitudinal cross sections. The continental area, in the Luanda region, exhibits lower ozone concentrations (40–45 ppbv, Figure 3a). Deep convection outflow in the ITCZ region could explain vertical transport of low ozone, for example, the marine boundary layer is typically less than 20 ppbv. Brazzaville (4°S) seems to be the southern limit of the Harmattan influence [Sauvage *et al.*, 2005]. Although

the Atlantic observations show the low surface ozone mixing ratios expected in a clean marine boundary layer (less than 20 ppbv), the oceanic middle upper troposphere shows ozone mixing ratios more than 80 ppbv. This oceanic vs. continental difference (30–40 ppbv) may reflect the influence of lightning on ozone formation far from continental sources. *Pickering et al.* [1996] show that 20 ppbv of ozone per day can be created during transport. Persistent southeasterly circulation shown by *Thompson et al.* [2000] may favor ozone photochemical creation during transport and explain high ozone values over the South Atlantic. Flow from the southeast may also suppress ozone buildup over the continent if ozone precursors are exported. In summary, in zone IIB, lightning seems to play a prominent role in the Atlantic-Africa upper tropospheric ozone difference.

[19] Zone II C (15°S–25°S) shows that a broad maximum (60–75 ppbv) appears in the upper levels (400–250 hPa) over the continent (Figure 3a), similar to the distribution over the ocean at the same latitude and altitude. Windhoek is under the influence of the St Helena anticyclone (Figure 4c) that allows recirculation of lightning emissions situated over the continent. This recirculation allows ozone formation in that region, in contrast to the Luanda area to the north. An additional mechanism may play a role in the South Atlantic maximum (zone II C, Figure 3b), through Rossby wave breaking, a process that is visible with 6-day backtrajectories flowing around an upper level trough over northeast Brazil (Figure 4d, particles starting near 20°S–30°S; 50°W). A case study of Rossby wave breaking and associated stratosphere-troposphere exchanges over the same region has been documented with MOZAIC data [*Scott et al.*, 2001]. Intrusions of high stratospheric ozone into the tropical Atlantic upper troposphere occur as a consequence of filamentary structures penetrating the upper troposphere of zone II C, and reaching the middle troposphere of zone II B (up to Ascension).

[20] To summarize, comparing oceanic and continental ozone cross sections (15°N to 22°S) gives further insight into a middle upper tropospheric ozone maximum over South Atlantic, that contributes to the paradox. The south Atlantic seems to be influenced by a combination of local and imported ozone, including biomass burning from west equatorial Africa that is transported through interaction of the Harmattan and ITCZ convection. Upper level circulation allows South American and African lightning to influence the south Atlantic. North Atlantic influences on ozone appear to be limited to easterly advection between 750–650 hPa. Stratospheric intrusions may also participate in the South Atlantic maximum.

3.2. Wave One: Longitudinal View of Ozone Mixing Ratio Cross Section

[21] SHADOZ stations permitted the first detailed characterization of the zonal wave one, showing an ozone maximum localized in the middle upper troposphere of the Atlantic–African–western Indian Ocean region [*Thompson et al.*, 2003b, 2004]. The climatology presented by *Thompson et al.* [2003b, 2004] was deficient in having only one near-equatorial African station (Nairobi). More generally, SHADOZ stations are relatively remote from biomass burning emissions, a disadvantage for evaluating photochemical and radiative properties of various types of

ozone profiles. Merging MOZAIC profiles with SHADOZ observations allows a gap in the African window to be filled. It also permits insight into the zonal character of northern tropical tropospheric ozone. When MOZAIC stations are separated by positions (north/south) relative to the ITCZ, the satellite view [*Kim et al.*, 1996] that the wave one is more pronounced over the Southern Hemisphere than over the Northern Hemisphere is confirmed. Figure 5 gives the Southern Hemisphere zonal wave-one view, with MOZAIC profiles integrated with the mean SHADOZ to 200 hPa. In contrast to *Thompson et al.* [2003b, Figure 4], Nairobi is omitted but Brazzaville, Congo, and Irene, South Africa are included in Figure 5.

[22] The prominent results are as follows.

[23] 1. In SON, the amplitude of the wave one appears to be stronger (larger column ozone amount over Africa) with the extra stations than implied by the *Thompson et al.* [2003b] climatology. Emissions (seen from satellite fire counts and lightning flashes) in SON are shifting from Tanzania to South Africa, adding to middle and lower tropospheric ozone over Brazzaville, Irene and Reunion (Figure 5a). Brazzaville and Irene ozone (>70 ppbv above 700 hPa) appears to be influenced by fire emissions (ATSR fire maps, <http://dup.esrin.esa.it/ionia/wfa/index.asp>, SON season) both from Brazil and southern Africa, predominantly from Mozambique and Tanzania. This is consistent with an analysis of *Thompson et al.* [1996], who used 1992 campaign sondes to estimate that 1/3 of south Atlantic free tropospheric ozone above a 'background amount' of 30 DU originated South America; the remainder was mostly from Africa. The SON wave intensity can also be explained by the observation that lightning in SON ramps up in frequency with the onset of the Southern Hemisphere wet season [*Moxim and Levy*, 2000]. Monthly wind streamlines at 500hPa from European Centre for Medium-Range Weather Forecasts (ECMWF) analyses (not shown), show persistent flow from the southeast on the northern branch of the St Helena anticyclone, centered over Namibia and Angola. This promotes advection of ozone and ozone precursors into the middle troposphere over the region around Brazzaville. Besides biomass burning and lightning, subsidence of upper tropospheric ozone in SON may also contribute to upper tropospheric ozone maximizing over the Atlantic at this time of the year [*Krishnamurti et al.*, 1996; *Jacob et al.*, 1996; *Martin et al.*, 2002; *Thompson et al.*, 2003b]. The ozone has a lifetime of weeks to a months or more. It may be photochemically formed with emissions interacting with convective systems [*Thompson et al.*, 1997] or lightning; it may even have some stratospheric character.

[24] 2. During DJF (Figure 5b) a new contribution to the wave one appears, with a lower tropospheric ozone maximum over Brazzaville from northern African biomass burning emissions (750–650 hPa, 55 ppbv). There is an upper tropospheric maximum over Irene (300–200 hPa, 75 ppbv), under the influence of continental anticyclonic recirculation [*Sauvage et al.*, 2005] monthly wind streamlines) and lightning activity (see section 3). *Diab et al.* [2004] noticed an intense convective activity which could give rise to the rapid vertical redistribution of surface-generated pollutants.

[25] 3. During boreal spring (MAM, Figure 5c) augmenting SHADOZ data with African profiles gives a similar

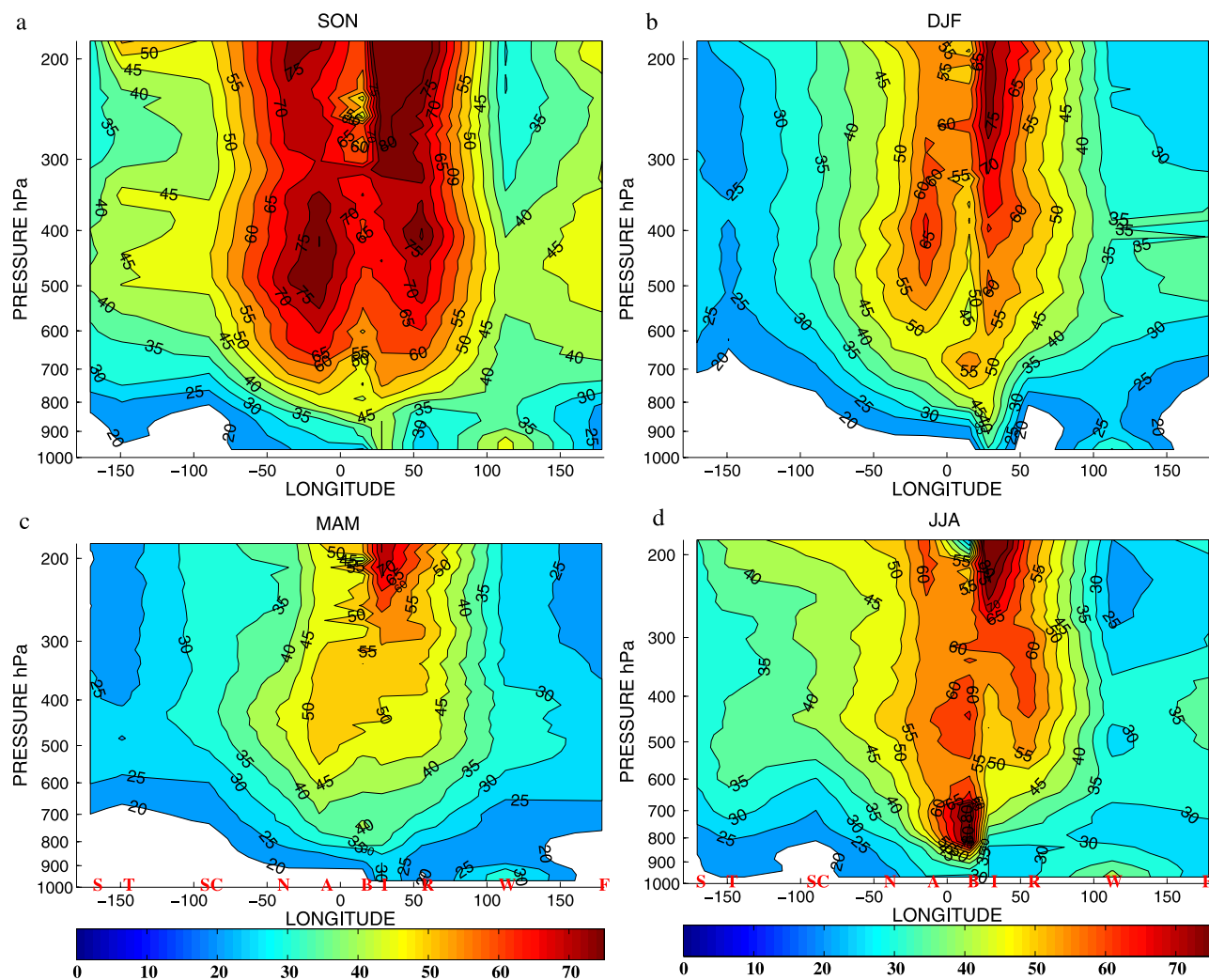


Figure 5. Zonal cross section of the ozone mixing ratio (in ppbv), as seen by the SHADOZ network and the MOZAIC data south of the ITCZ during (a) September–November, (b) December–February, (c) March–May, and (d) June–August. Letters in red indicate the positions of the different sites of measurements: S, Samoa; T, Tahiti; SC, San Cristobal; N, Natal; A, Ascension; B, Brazzaville; I, Irene; R, Reunion Island; W, Watukosek; and F, Fiji.

pattern to the SHADOZ-only data [cf. *Thompson et al.*, 2003b, Figure 4a]. The ozone wave one has its maximum at 50–55 ppbv in the lower middle troposphere, the lowest values in the annual cycle. This is the least polluted season over Africa.

[26] 4. The lower tropospheric contribution to the zonal wave-one maximum, not captured well in SHADOZ data alone, is highlighted during JJA (Figure 5d). Brazzaville appears to be constantly supplied with ozone-enriched air from the flow of the southeasterly trades as well as from regional biomass burning [*Sauvage et al.*, 2005]. Note 60–80 ppbv ozone from 0°–35°E near 900–650 hPa. Irene is south of the maximum fire activity and less under the influence of the polluted trades. The high ozone concentrations in the upper troposphere over Irene may be partly from stratospheric intrusions between August and November. The higher ozone reflects a lower tropopause [*Thompson et al.*, 2003b; *Diab et al.*, 2003].

[27] The merged SHADOZ-MOZAIC data point to Africa as the predominant factor in the ozone maximum that gives rise to the zonal wave one. Multiple sources over Africa are probably responsible. In SON the wave one is partly enhanced through increased subsidence and an ozone transported from South American sources.

[28] Figure 6 displays a latitudinal view of upper tropospheric ozone taken over the broad region in which the maximum of the zonal wave one is found. Shown are seasonal variations of mean ozone along the MOZAIC flight transects over the Atlantic (on Europe–Brazil flights) and Africa (along flights between Europe and South Africa). The 60–70 ppbv ozone isocontour viewed in a 5°S–10°S latitude band (Figure 5a), is seen to extend from 0° up to 20°S (Figures 6a and 6b), at least for the 300–200 hPa level. This confirms the overall ozone maximum observed by satellite over the south tropical Atlantic–African region and by individual soundings by *Weller et al.* [1996] and *Thompson et al.* [2000]. Ozone

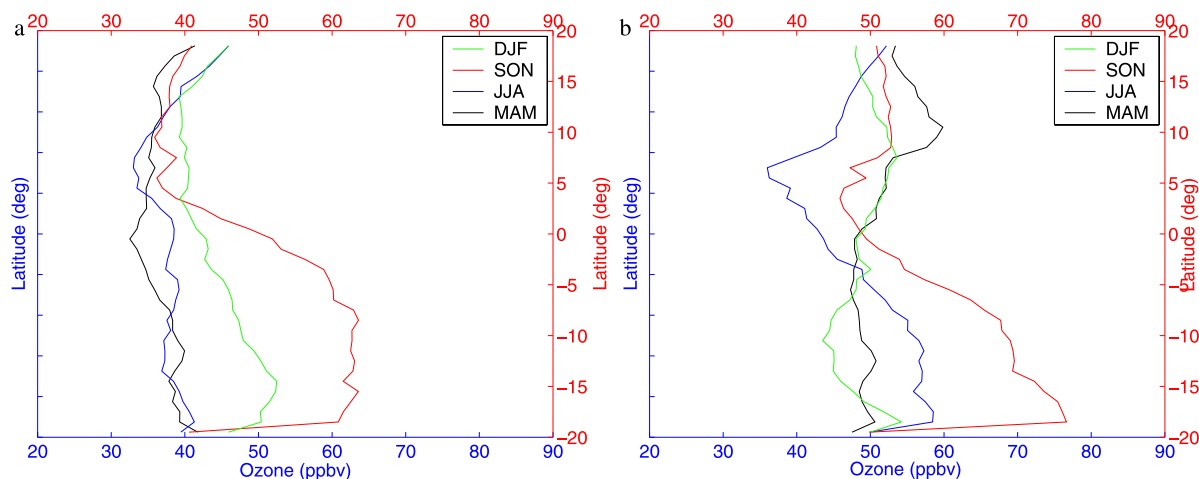


Figure 6. Ozone seasonal mean of the MOZAIC transect (1994–2002) over (a) the Atlantic and (b) Africa. Mean altitude flight is around 257 hPa.

concentrations along the transect are indeed greater south than north of the ITCZ. The difference between the hemispheres is more than 20 ppbv over the Atlantic and African regions, consistent with the southern character of the zonal wave one. From 10°N to 20°S, the transits show that ozone is higher over Africa than over Atlantic in all seasons except DJF (see section 3). During DJF the maximum over Africa remains in the Northern Hemisphere. At that time, in the Southern Hemisphere, the maximum is higher over the Atlantic than over Africa. This South Atlantic maximum with 53 ppbv on average near 15°S is much lower than the ozone soundings at the corresponding altitude during the Aerosols99 campaign [Thompson *et al.*, 2000, Figure 1]. Note, however, that at this latitude, most MOZAIC data are recorded near 40°W whereas a mean longitude for the tropical segments of the Aerosols99 cruise was 10°W. Individual soundings during the Polarstern January–February 1993 [Weller *et al.*, 1996], their Plate 1b) situated near 30°W also display greater values, from 60 up to 80 ppbv near 10–12 km. Some 60–80 ppbv values occur in MOZAIC data when looking at individual cases, but the maxima are still lower than the strong ozone concentrations measured further east during the Aerosols99 campaign. This implies that there are some longitudinal gradients affecting ozone across the Atlantic south of the equator (Figure 6a). For example, Figure 3a highlights the high ozone in the lower troposphere north of the ITCZ over Africa. This is not found over the Atlantic. Conversely, the high ozone in the middle upper troposphere over the Atlantic (compare Figures 6a and 6b) may reflect multiple sources, as inferred from the satellite data invoked to explain the Aerosols99 vertical cross section (Figure 3b).

[29] To summarize, the wave-one pattern deduced from the SHADOZ data [Thompson *et al.*, 2003a, 2003b] has been enhanced through finer-scale observations provided by MOZAIC. The resulting view shows higher ozone over lower altitudes, especially over the African continent. This ozone can be explained by influences from lightning, biomass burning and/or biofuel fires over Africa and a

tendency for ozone to build up preferentially over the Southern Hemisphere (except in DJF).

4. Discussion and Conclusions

[30] The MOZAIC data over equatorial Africa, with the majority of profiles taken over that part of the continent, fill an African “gap” when merged with the SHADOZ and Aerosols99 soundings, giving an updated and more detailed description of tropical tropospheric ozone in this region. The result is a broader understanding of both the tropical Atlantic ozone paradox and a more persistent feature: the South Atlantic maximum in ozone. The ozone paradox is evidently limited to the Atlantic and does not hold over the African continent. Furthermore, the paradox, initially identified during January and February, is seen to be a component of the zonal wave one. Through the amplified view provided by the combined data the wave one appears to be, as initially inferred [Kim *et al.*, 1996], a predominantly Southern Hemisphere phenomenon.

[31] By merging SHADOZ and MOZAIC data over Africa a number of results and some new insights are obtained. The South Atlantic maximum, a component of both a ‘paradox’ and the zonal wave one in tropospheric ozone, is better defined, thanks to additional continental observations in western Africa over the Atlantic. This is the best view of the zonal wave one presently available. The in situ view, especially the finding that lower tropospheric ozone from pollution sources makes a sizable contribution to the Northern Hemisphere African ozone column, helps define requirements for models and satellite ozone retrievals.

[32] Completion of a southern African window with data from Luanda, Windhoek, Brazzaville and Irene, shows that the seasonal SON maximum is more pronounced than in the published views, largely because of new lower-altitude features. Within the wave one, previously delineated with profiles from relatively unpolluted sites [Thompson *et al.*, 2003a, 2003b, 2004], the intensity of low-altitude ozone is very strong in JJA and DJF, the maximum of Southern and

Northern hemisphere biomass fire activity over Africa. These lower tropospheric ozone maxima have the same intensity, 65–75 ppbv, as those measured in the upper levels [e.g., *Thompson et al.*, 2003a, Figure 4b]. Mechanisms relating to biogenic activity and precipitation may also play a role in lower tropospheric ozone seasonality [*Harris et al.*, 1996; *Swap et al.*, 2003; *Jaeglé et al.*, 2004]. Middle and upper troposphere ozone distributions are also better documented with the combined MOZAIC-SHADOZ view. Depending on the season, regional and/or long-scale transport processes “load” the South Atlantic and equatorial Africa with anthropogenic and/or natural ozone.

[33] A number of origins for ozone emerge.

[34] 1. The upper troposphere over South America may influence upper levels with high ozone concentrations over the region of the South Atlantic ozone maximum. This would be consistent with the estimate from SAFARI-92/TRACE-A (September–October season) that 20–30% of pollution ozone over the South Atlantic ozone maximum originated from South America [*Thompson et al.*, 1996]. Deep convection with associated lightning activity may also contribute to ozone production in this area [*Pickering et al.*, 1996].

[35] 2. The arguments for an African lightning influence on the middle troposphere over the South Atlantic [*Jenkins et al.*, 2003] are consistent with the observations here. Easterly flow maintains a connection between continental sources and the Atlantic. This flow pattern and convective ventilation of dilute emissions suppress ozone production to some degree, over the continent. However, ozone can form during transport and supply the oceanic areas where ozone builds up. The St Helena anticyclone, leads to redistribution of ozone from farther north in Africa around Namibia [*Diab et al.*, 2003, 2004].

[36] 3. Biomass burning (and mostly likely anthropogenic pollution) connected with the African continent play a major role in enhancing ozone in the lower troposphere over the Gulf of Guinea. These high ozone concentrations (and presumably some of the ozone precursors) may be vertically redistributed via convection in the region of the ITCZ into the middle upper troposphere over the Congo and South Atlantic. This transport may be combined with advection within the Harmattan as the winds transport pollution from the continent to the southwest.

[37] Other mechanisms may be at work, for example, stratospheric intrusion induced by Rossby wave breaking, a hypothesis of *Weller et al.* [1996] regarding air masses of high ozone and low water vapor in the free troposphere of the Southern Hemisphere.

[38] The aforementioned transport processes have been assessed using monthly averaged data, for instance, by looking at 3D streamlines based on ECMWF analysis. To study episodes in detail, a global model that incorporates various sources of ozone precursors is needed. Such a model also would allow us to evaluate the regional African-Atlantic contribution to the global ozone budget. The most challenging aspect presented by the ozone data is discriminating among major contributions from biomass burning emissions, lightning and biogenic activity. There seems to be no doubt each of these influences is operative.

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References

- Chatfield, R. B., H. Guan, A. M. Thompson, and J. C. Witte (2004), Convective lofting links Indian Ocean air pollution to paradoxical South Atlantic ozone maxima, *Geophys. Res. Lett.*, *31*, L06103, doi:10.1029/2003GL018866.
- Diab, R. D., A. Raghunandan, A. M. Thompson, and V. Thouret (2003), Classification of tropospheric ozone profiles over Johannesburg based on MOZAIC aircraft data, *Atmos. Chem. Phys.*, *3*, 713–723.
- Diab, R. D., A. M. Thompson, K. Mari, L. Ramsay, and G. J. R. Coetzee (2004), Tropospheric ozone climatology over Irene, South Africa, from 1990 to 1994 and 1998 to 2002, *J. Geophys. Res.*, *109*, D20301, doi:10.1029/2004JD004793.
- Edwards, D. P., et al. (2003), Tropospheric ozone over the tropical Atlantic: A satellite perspective, *J. Geophys. Res.*, *108*(D8), 4237, doi:10.1029/2002JD002927.
- Fishman, J., and J. C. Larsen (1987), Distribution of total ozone and stratospheric ozone in the tropics: Implications for the distribution of tropospheric ozone, *J. Geophys. Res.*, *92*, 6627–6634.
- Fishman, J., K. Fakhruzzaman, B. Cros, and D. Nganga (1991), Identification of widespread pollution in the Southern Hemisphere deduced from satellite analyses, *Science*, *252*, 1696–1963.
- Fishman, J., A. E. Wozniak, and J. K. Creilson (2003), Global distribution of tropospheric ozone from satellite measurements using the empirically corrected tropospheric ozone residual technique: Identification of the regional aspects of air pollution, *Atmos. Chem. Phys.*, *3*, 893–907.
- Harris, G. W., F. G. Wienhold, and T. Zenker (1996), Airborne observations of strong biogenic NO_x emissions from the Namibian savanna at the end of the dry season, *J. Geophys. Res.*, *101*, 23,707–23,712.
- Jacob, D. J., et al. (1996), Origin of ozone and NO_x in the tropical troposphere: A photochemical analysis of aircraft observations over the South Atlantic basin, *J. Geophys. Res.*, *101*, 24,235–24,250.
- Jaeglé, L., R. V. Martin, K. Chance, L. Steinberger, T. P. Kurosu, D. J. Jacob, A. I. Modi, V. Yoboué, L. Sigha-Nkamdjou, and C. Galy-Lacaux (2004), Satellite mapping of rain-induced nitric oxide emissions from soils, *J. Geophys. Res.*, *109*, D21310, doi:10.1029/2004JD004787.
- Jenkins, G. S., and J.-H. Ryu (2004a), Space-borne observations link the tropical Atlantic ozone maximum and paradox to lightning, *Atmos. Chem. Phys.*, *4*, 361–375.
- Jenkins, G. S., and J.-H. Ryu (2004b), Linking horizontal and vertical transports of biomass fire emissions to the tropical Atlantic ozone paradox during the Northern Hemisphere winter season: Climatology, *Atmos. Chem. Phys.*, *4*, 449–469.
- Jenkins, G. S., J. Ryu, A. M. Thompson, and J. C. Witte (2003), Linking horizontal and vertical transports of biomass fire emissions to the tropical Atlantic ozone paradox during the Northern Hemisphere winter season: 1999, *J. Geophys. Res.*, *108*(D23), 4745, doi:10.1029/2002JD003297.
- Kim, J. H., R. D. Hudson, and A. M. Thompson (1996), A new method of deriving time-averaged tropospheric column ozone over the tropics using TOMS radiances: Intercomparison and analysis, *J. Geophys. Res.*, *101*, 24,317–24,330.
- Kim, J. H., S. Na, M. J. Newchurch, and R. V. Martin (2005), Tropical tropospheric ozone morphology and seasonality seen in satellite and in situ measurements and model calculations, *J. Geophys. Res.*, *110*, D02303, doi:10.1029/2003JD004332.
- Krishnamurti, T. N., M. C. Sinha, M. Kanamitsu, D. Oosterhof, H. Fuelberg, R. Chatfield, D. J. Jacob, and J. Logan (1996), Passive tracer transport relevant to the TRACE A experiment, *J. Geophys. Res.*, *101*, 23,889–23,908.
- Logan, J. A., and V. W. J. H. Kirchhoff (1986), Seasonal variations of tropospheric ozone at Natal, Brazil, *J. Geophys. Res.*, *91*, 7875–7888.
- Marengo, A., et al. (1998), Measurement of ozone and water vapor by Airbus in-service aircraft: The MOZAIC airborne program, An overview, *J. Geophys. Res.*, *103*, 25,631–25,642.
- Martin, R. V., et al. (2002), Interpretation of TOMS observations of tropical tropospheric ozone with a global model and in situ observations, *J. Geophys. Res.*, *107*(D18), 4351, doi:10.1029/2001JD001480.
- Moxim, W. J., and H. Levy II (2000), A model analysis of the tropical South Atlantic Ocean tropospheric ozone maximum: The interaction of transport and chemistry, *J. Geophys. Res.*, *105*, 17,393–17,416.

- Newchurch, M. J., D. Sun, J. H. Kim, and X. Liu (2003), Tropical tropospheric ozone derived using clear-cloudy pairs (CCP) of TOMS measurements, *Atmos. Chem. Phys.*, *3*, 683–695.
- Pickering, K. E., et al. (1996), Convective transport of biomass burning emissions over Brazil during TRACE-A, *J. Geophys. Res.*, *101*, 23,993–24,012.
- Sauvage, B., V. Thouret, J.-P. Cammas, F. Gueusi, G. Athier, and P. Nédélec (2005), Tropospheric ozone over equatorial Africa: Regional aspects from the MOZAIC data, *Atmos. Chem. Phys.*, *5*, 311–335.
- Scott, R. K., J.-P. Cammas, P. Mascart, and C. Stolle (2001), Stratospheric filamentation into the upper tropical troposphere, *J. Geophys. Res.*, *106*, 11,835–11,848.
- Shiotani, M. (1992), Annual, quasi-biennial, and El Niño–Southern Oscillation (ENSO) time-scale variations in equatorial total ozone, *J. Geophys. Res.*, *97*, 7625–7633.
- Smit, H., D. Kley, S. McKeen, A. Voltz, and S. Gilge (1989), The latitudinal and vertical distribution of tropospheric ozone over the Atlantic Ocean in the Southern and Northern hemispheres, in *Ozone in the Atmosphere, Stud. Geophys. Opt. Remote Sens.*, edited by R. D. Bojkov and P. Fabian, pp. 419–422, A. Deepak, Hampton, Va.
- Swap, R. J., H. J. Annegarn, J. T. Suttles, M. D. King, S. Platnick, J. L. Privette, and R. J. Scholes (2003), Africa burning: A thematic analysis of the Southern African Regional Science Initiative (SAFARI 2000), *J. Geophys. Res.*, *108*(D13), 8465, doi:10.1029/2003JD003747.
- Thompson, A. M., and R. D. Hudson (1999), 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*, 26,961–26,975.
- 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 (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., W.-K. Tao, K. E. Pickering, J. R. Scala, and J. Simpson (1997), Tropical deep convection and ozone formation, *Bull. Am. Meteorol. Soc.*, *78*, 1043–1054.
- Thompson, A. M., B. G. Doddridge, J. C. Witte, R. D. Hudson, W. T. Luke, J. E. Johnson, B. J. Johnson, S. J. Oltmans, and R. Weller (2000), A tropical Atlantic paradox: Shipboard and satellite views of a tropospheric ozone maximum and wave-one in January–February 1999, *Geophys. Res. Lett.*, *27*, 3317–3320.
- Thompson, A. M., et al. (2003a), Southern Hemisphere Additional Ozone-sondes (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 Ozone-sondes (SHADOZ) 1998–2000 tropical ozone climatology: 2. Tropospheric variability and the zonal wave-one, *J. Geophys. Res.*, *108*(D2), 8241, doi:10.1029/2002JD002241.
- Thompson, A. M., J. C. Witte, S. J. Oltmans, and F. J. Schmidlin (2004), SHADOZ (Southern Hemisphere Additional Ozone-sondes): A tropical ozone-sonde-radiosonde network for the atmospheric community, *Bull. Am. Meteorol. Soc.*, *85*, 1549–1564.
- Weller, R., R. Lilisckis, O. Schrems, R. Neuber, and S. Wessel (1996), Vertical ozone distribution in the marine atmosphere over the central Atlantic Ocean (56°S–50°N), *J. Geophys. Res.*, *101*, 1387–1400.
- Wernli, H., and H. C. Davies (1997), A Lagrangian-based analysis of extratropical cyclones, I: The method and some applications, *Q. J. R. Meteorol. Soc.*, *123*, 467–489.
- Ziemke, J. R., S. Chandra, and P. K. Bhartia (1998), Two new methods for deriving tropospheric column ozone from TOMS measurements: Assimilated UARS MLS/HALOE and convective-cloud differential techniques, *J. Geophys. Res.*, *103*, 22,115–22,128.

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