**Atmospheric
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Space-borne observations link the tropical atlantic ozone maximum and paradox to lightning

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Abstract. The potential enhancement of tropospheric column ozone values over the Tropical Atlantic Ocean on a seasonal basis by lightning is investigated using satellite derived ozone data, TRMM lightning data, ozonesonde data and NCEP reanalysis during 1998–2001. Our results show that the number of lightning flashes in Africa and South America reach a maximum during September, October and November (SON). The spatial patterns of winds in combination with lightning from West Africa, Central Africa and South America is likely responsible for enriching middle/upper troposphere ozone over the Tropical South Atlantic during SON. Moreover, lightning flashes are high in the hemisphere opposite to biomass burning during December, January, and February (DJF) and June, July and August (JJA). This pattern leads to an enrichment of ozone in the middle/upper troposphere in the Southern Hemisphere Tropics during DJF and the Northern Hemisphere Tropics during JJA. During JJA the largest numbers of lightning flashes are observed in West Africa, enriching tropospheric column ozone to the north of 5° S in the absence of biomass burning. During DJF, lightning is concentrated in South America and Central Africa enriching tropospheric column ozone south of the Equator in the absence of biomass burning.
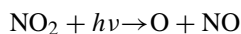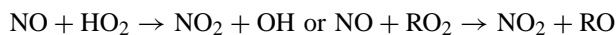
1 Introduction

Fishman et al. (1991) identified trends in total tropospheric column ozone (TCO) over the Tropical Atlantic using the Total Ozone Mapping Spectrometer (TOMS). They found that the highest TCO values (tropical ozone maximum) were found in Northern Hemisphere (NH) autumn (SON) primarily over the Southern Tropical Atlantic Ocean and likely associated with biomass burning in South America and South-

ern Africa. This feature was also found during the Transport and Atmospheric Chemistry Near the Equator-Atlantic (TRACE-A) field experiment (Fishman et al., 1996; Thompson et al., 1996). The TRACE-A observations identified regions of biomass burning and other anthropogenic sources of ozone and ozone precursors, horizontal and vertical transports of ozone/ozone precursors, and limited observations have examined the association of lightning with the production of NO_x and O_3 (Jacob et al., 1996). Weller et al. (1996) suggests that stratospheric intrusions of O_3 may also be responsible for elevated ozone in the upper troposphere.

A feature of interest during the Northern Hemisphere (NH) winter season (DJF), is the relationship between tropospheric ozone and biomass burning. During DJF, biomass burning is confined to the Northern Hemisphere primarily in West Africa. The highest TCO values, however, are still found over the Southern Tropical Atlantic Ocean and denoted as the Tropical Ozone Paradox (Weller et al., 1996; Thompson et al., 2000; Martin et al., 2002; Edwards et al., 2003). In the Aerosols99 Ship campaign, Thompson et al. (2000) found the highest ozone mixing ratios in the lower troposphere downstream of biomass burning over the Northern Tropical Atlantic, but in the middle/upper troposphere over the Southern Tropical Atlantic due most likely to lightning. Edwards et al. (2003) have linked the higher TCO values of the South Tropical Atlantic to lightning from Southern Africa and South America.

Ozone enhancement in the middle/upper troposphere can occur through lightning via NO_x production:

$$NO + HO_2 \rightarrow NO_2 + OH \text{ or } NO + RO_2 \rightarrow NO_2 + RO$$

$$NO_2 + h\nu \rightarrow O + NO$$

$$O + O_2 + M \rightarrow O_3 + M$$

Observations of NO_x and O_3 production associated with deep convection are limited but show that the vertical transport of ozone/ozone precursors, stratospheric intrusions,

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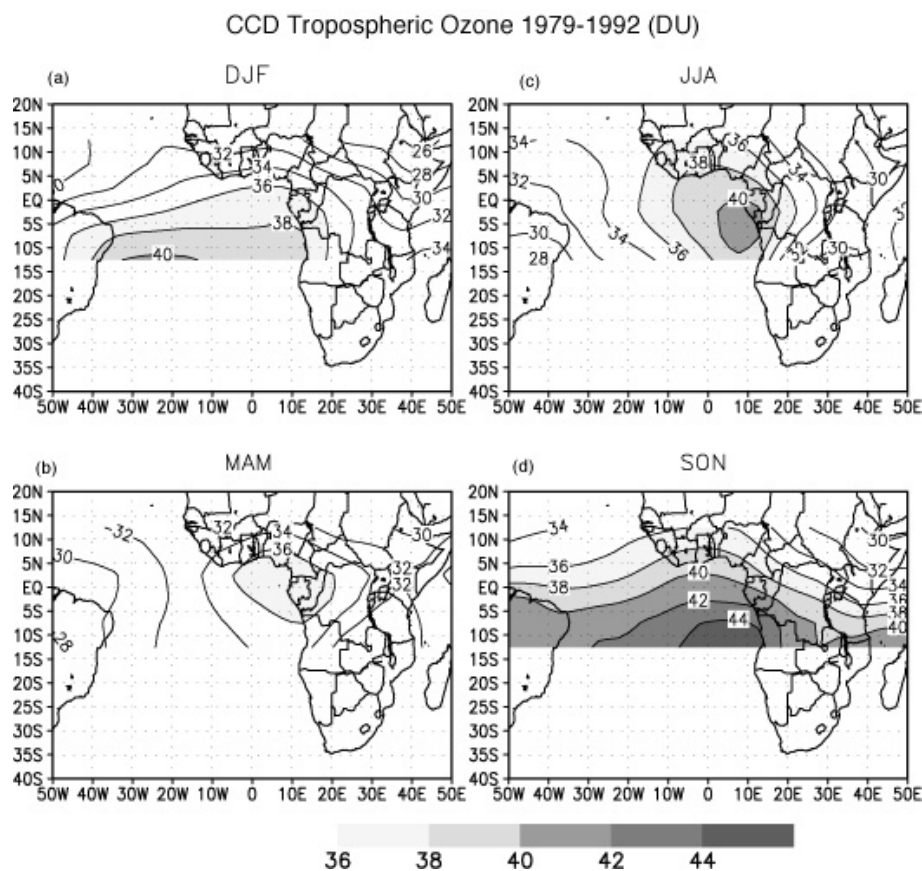
Fig. 1. Climatological (1979–1992) CCD Tropospheric Column Ozone values **(a)** DJF, **(b)** MAM, **(c)** JJA, **(d)** DJF. Units are Dobsons.

NO_x and O_3 production via lightning do occur (Poulida et al., 1996; Pickering et al., 1996; Stith et al., 1999; Dye et al., 1999). The study of DeCaria et al. (2000) found an ozone production of $7\,ppbv\,d^{-1}$ from a simulated thunderstorm in the United States where elevated NO_x was observed.

Thompson et al. (2000) suggests an ozone production of 4–$8\,ppbv\,d^{-1}$ during the Aerosols99 campaign originating from lightning in Central Africa. Martin et al. (2000) using global lightning NO_x emission estimates available from Price et al. (1997) have found that lightning explains about 20% of the variance in tropospheric column ozone values of the Tropical Atlantic using EOF analysis. Using TRMM LIS data Jenkins et al. (2003) and Jenkins and Ryu (2003) have also found that lightning from Central Africa and South America likely contributes to enhancement of ozone in the Southern Hemisphere during DJF. There are also times when ozone in the upper troposphere of the Northern Hemisphere can be enhanced via convection off the coast of West Africa during DJF (Jenkins et al., 2003).

Finding a direct relationship between lightning and enhanced tropospheric ozone is challenging because it requires the concurrent observation of lightning and tropospheric ozone. The introduction of the Optical Transient Detector (OTD) in 1995 (Christian et al., 2003) and more re-

cently the launch of the TRMM satellite (Kummerow et al., 1998) which includes the Lightning Imaging Sensor (LIS) allowed for the mapping of tropical lightning. Techniques have also been developed to derive the tropospheric column ozone from the total column ozone from the Total Ozone Mapping Spectrometer (TOMS) instrument (Fishman et al., 1991; Hudson and Thompson, 1998; Ziemke et al., 2001). Even with these recent advances, there are still problems with the observations. For example, the LIS instrument undersamples lightning in the tropics as it takes 47 days to complete the full diurnal cycle at a particular location (Nesbitt et al., 2000). There are also problems with extracting tropospheric column ozone from total column ozone.

Kim et al. (2001) suggest that the Modified Residual (MR) technique of Hudson and Thompson (1998) can underestimate the tropospheric ozone column if high ozone mixing ratios are found in the lower troposphere. Martin et al. (2002) have suggested that satellite derived TCO values of the MR technique are underestimated by 3–$5\,DU$ over West Africa during the biomass burning season (DJF) because the highest ozone mixing ratios are confined to the lower troposphere. On the other hand, during the months of June through October, Martin et al. (2002) suggest that MR technique significantly overestimates TCO values for West Africa and the
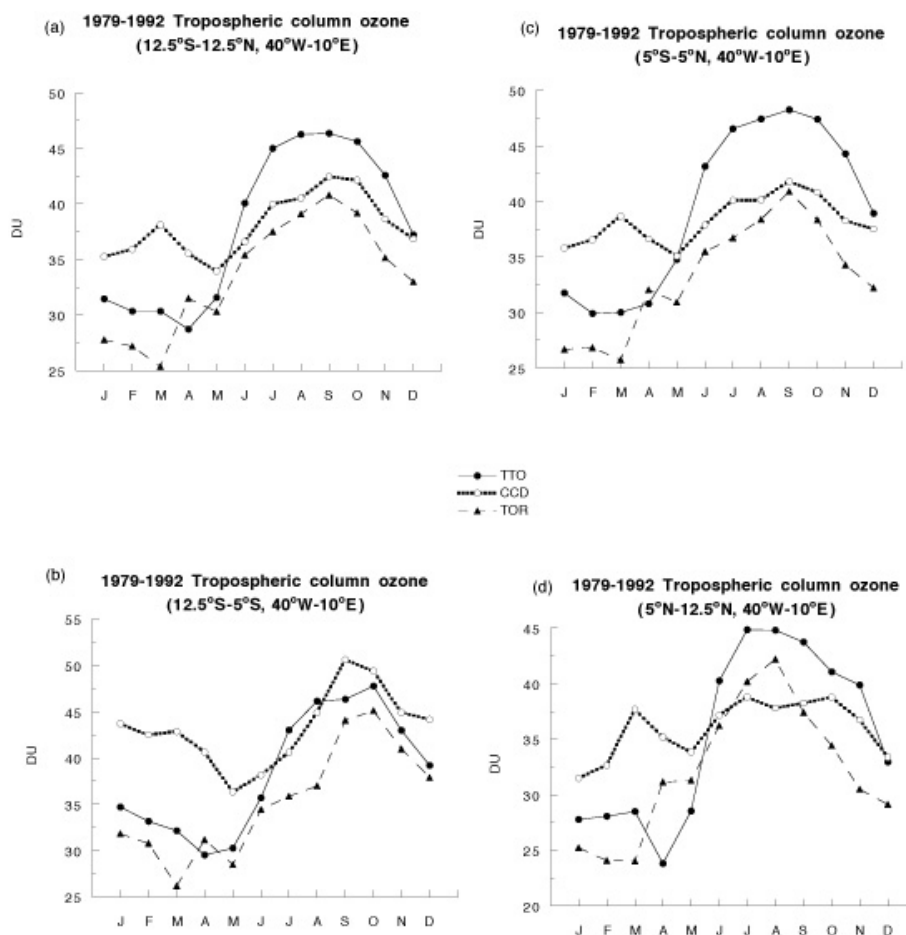
Fig. 2. The annual cycle of the tropospheric column ozone (1979–1992) for TTO, CCD and TOR averaged over **(a)** 12.5° S–12.5° N, 40° W–10° E. **(b)** 12.5° S–5° S, 40° W–10° E, **(c)** 5° S–5° N, 40° W–10° E, **(d)** 5° N–12.5° N, 40° W–10° E. Units are Dobsons.

adjacent Atlantic Ocean when compared to the MOSAIC data. However, Fishman et al. (2003) also show that TCO values are elevated over West Africa during JJA. We suggest in this paper that lightning is responsible for elevated TCO values over West Africa and the adjacent Atlantic during JJA.

The objectives of this paper are: (1) to compare monthly and seasonal TCO values over the Tropical Atlantic using TTO (using the MR technique) data and convective-cloud differential (CCD) data (Ziemke et al., 2000) during 1979–1992. (2) To examine in detail monthly and seasonal data (TCO values, ozonesondes, upper level winds and lightning) during 1998–2001. This is done in order to establish a relationship between enhanced tropospheric ozone and lightning. In particular we identify temporal/spatial distribution of lightning amongst South America, West Africa and Central Africa, upper levels winds and its relationship to ozonesonde measurements and TCO values over the Tropical Atlantic Ocean. (3) To examine if lightning is responsible for elevated TCO values over West Africa and the adjacent waters during JJA as suggested by TTO data. (4) To examine the

relationship between the Tropical Atlantic Ozone Maximum during SON and lightning over Africa and South America. Sinks of ozone such as photolysis, deposition and chemical transformation are not explored and troposphere-stratosphere exchanges of air are also not considered.

2 Data description

The NCEP reanalysis (Kalnay et al., 1996) are used for wind at a horizontal resolution of $2.5° \times 2.5°$. Three datasets that derive TCO values from the TOMS satellite are used for comparison in this study. The Tropical Tropospheric Ozone (TTO) data is based on the MR technique and produce TCO values on a horizontal grid of $1° \times 2°$ (Hudson and Thompson, 1998; Thompson and Hudson, 1999). The cloud-slicing method (CCD) of Ziemke et al. (2001) is produced on a horizontal grid of $5° \times 5°$. The Tropospheric Ozone Residual (TOR) data of Fishman et al. (2003) is produced on a horizontal grid of $1° \times 1.25°$. The TTO, CCD and TOR datasets are compared for the climatological period of 1979–1992.
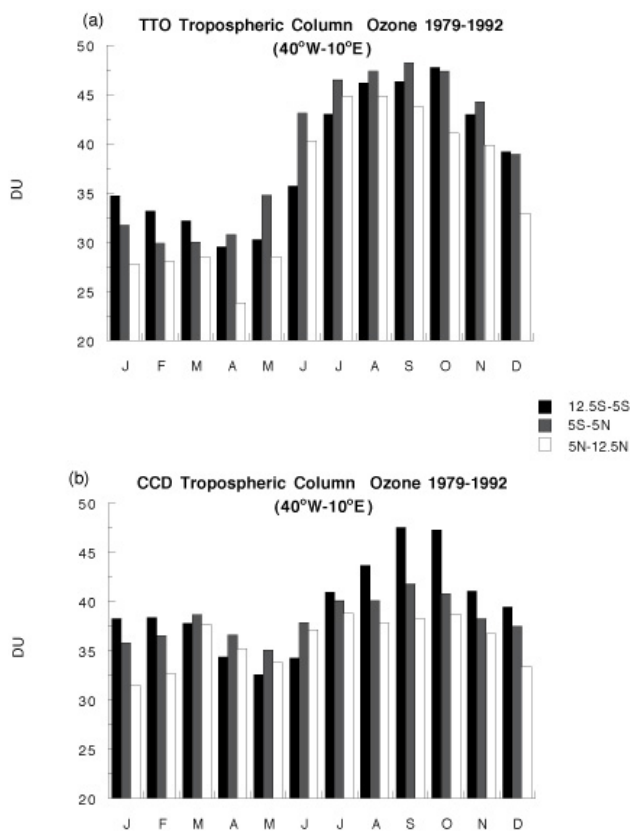
Fig. 3. The annual cycle of the tropospheric column ozone (1997–1992) by latitude zones for **(a)** TTO and **(b)** CCD. Units are Dobsons.

Further, we show MAM and JJA 1998, 1999 TCO using CCD, TTO and the TOR datasets. We did not use the TTO or TOR data for 2000 and 2001 because of possible TOMS instrumentation problems during 2000 and 2001.

The TTO data was available because the MR technique, tropospheric column ozone from ozonesondes near the Atlantic ozone maximum is subtracted from TOMS total ozone to give the stratospheric column ozone values. Stratospheric column ozone is then subtracted from total ozone to yield tropospheric ozone at all other longitudes. In the CCD method, stratospheric ozone is set equal to the TOMS total ozone measurement over high reflecting, high altitude clouds in the western Pacific Ocean. At cloud-free pixels, with a reflectivity <0.2, tropospheric column ozone is obtained by subtracting the above-cloud stratospheric ozone amount from the TOMS total amount. It is assumed that highly reflecting clouds have cloud tops at the tropopause. The TOR data is computed by subtracting the Stratospheric Column Ozone (SCO) from the TOMS total column ozone. The SCO is derived from measurements from the Solar Backscattering Ultraviolet (SBUV) instrument.

The ozone vertical profiles from Southern Hemisphere Additional Ozonesondes (SHADOZ) stations are also used

for 1998–2000 (Thompson et al., 2003). The fire count data is estimated from the Along Tracking Scanning Radiometer (ATSR) instrument, which has a horizontal resolution of 1 km and a swath width of 512 km. Fires are identified at a fixed threshold of 312° K (Arino and Melinotte, 1995). The fire count data are averaged for the period of 1996–2000. Observations of non-gridded monthly lightning flashes from the Lightning Imaging Sensor (LIS) from daily overpasses are used in this study (Christian et al., 2003).

3 Results

3.1 Climatology of the tropical atlantic tropospheric column ozone (TCO)

Figure 1 shows the CCD 1979–1992 time-averaged seasonal TCO values over the Tropical Atlantic Ocean between 12.5° N and 12.5° S. The lowest values (<34 DU) are found during DJF and MAM and increase during the Northern Hemisphere summer and autumn seasons. The highest values (>44 DU) are found during SON over the Tropical South Atlantic Ocean in agreement with Fishman et al. (1991). Figures 2a–d compare the annual cycle of 1979–1992 TCO values for the TTO, CCD and TOR datasets over the Tropical Atlantic Ocean (12.5° N–12.5° S, 40° W–10° E) and its various sub-regions. The annual peak in both datasets for the Tropical Atlantic Ocean occurs in September (Fig. 2a). However, there is a larger range in the annual TTO (∼17–18 DU) and TOR data (∼15–16 DU) relative to the CCD data (∼8–9 DU). Furthermore the TTO and TOR values are lower than CCD values from January through May by 3–5 DU in TTO and up to 10 DU in TOR. During June through November the TTO values are higher than the CCD values by 3–6 DU. This pattern is also found in the sub-regions (Figs. 2b–d), except for the southern latitudinal band (12.5° S–5° S) during the June–November period when the differences between the TTO and CCD are smaller (Fig. 2b). The TOR TCO values are generally smaller than the CCD or TTO values except in the latitudinal zone of 5° N–12.5° N (Fig. 2b).

The TTO and TOR data suggests that some factor (anthropogenic or natural) leads to a significant rise in TCO values over the Tropical Atlantic after the month of May. This is also implied in the CCD dataset but the amplitude of the increase is smaller relative to TTO (Fig. 2a). While the increase is seen at all latitude zones it is largest in the zone between 5° N and 12.5° N (Fig. 2d). For example there is a relative increase of 16 DU and 9 DU between May and July in the TTO and TOR datasets respectively. A reduction in TCO values begins after the month of October. Given that the high TCO values occur over the ocean in a region of sinking motions and that there are no natural (lightning is negligible over the ocean), or anthropogenic sources located there, the ozone must be transported from remote locations.
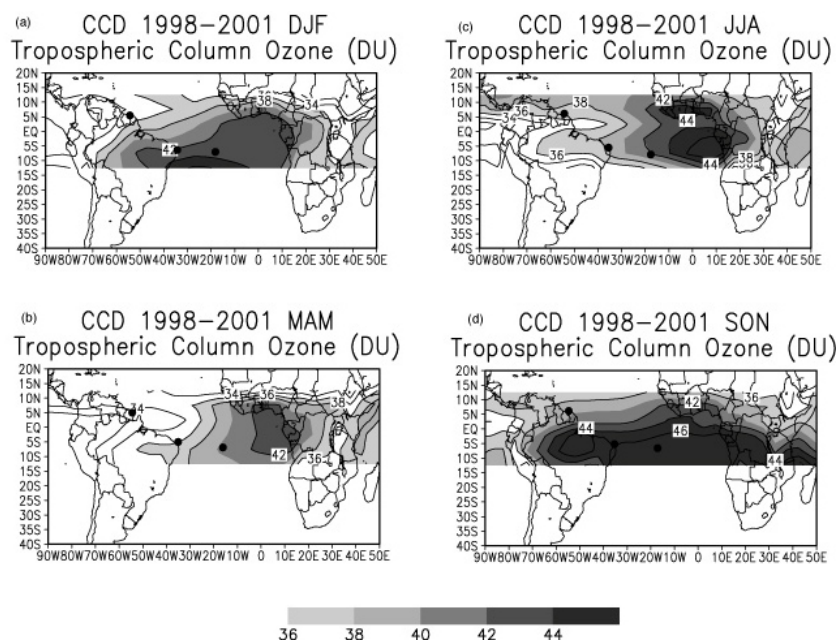
Fig. 4. 1998–2001 CCD Tropospheric Column Ozone values **(a)** DJF, **(b)** MAM, **(c)** JJA, **(d)** DJF. Units are Dobsons.
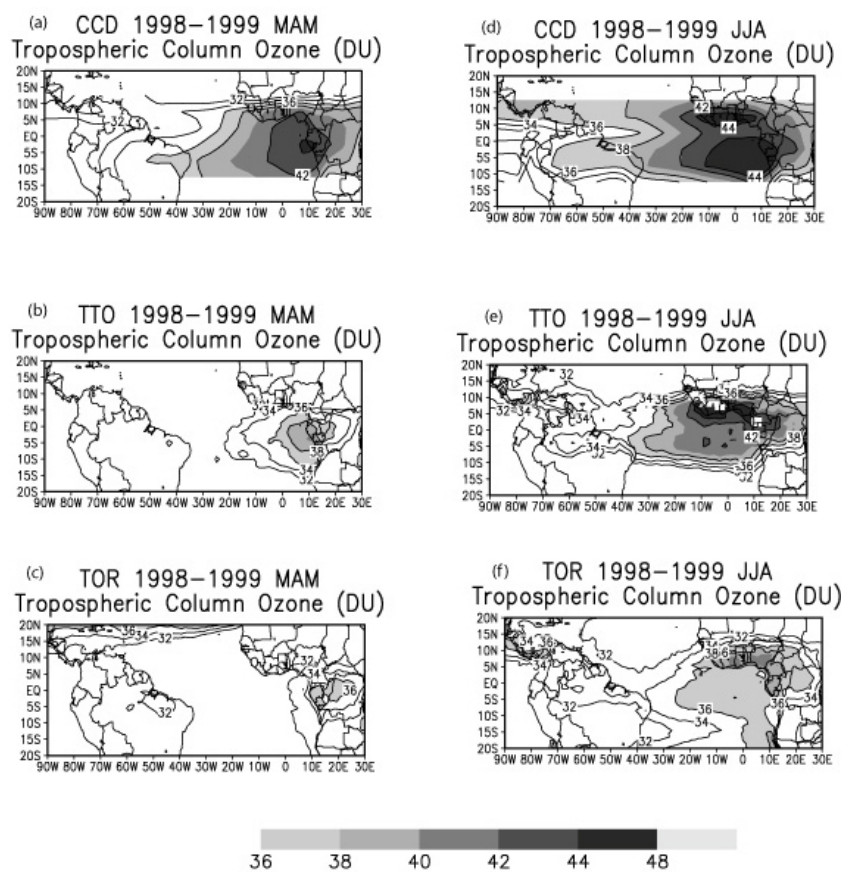
Fig. 5. Average 1998–1999 Tropospheric Column Ozone values **(a)** CCD-MAM, **(b)** TTO-MAM, **(c)** TOR-MAM, **(d)** CCD-JJA, **(e)** TTO-JJA, **(f)** TOR-JJA. Units are Dobsons.
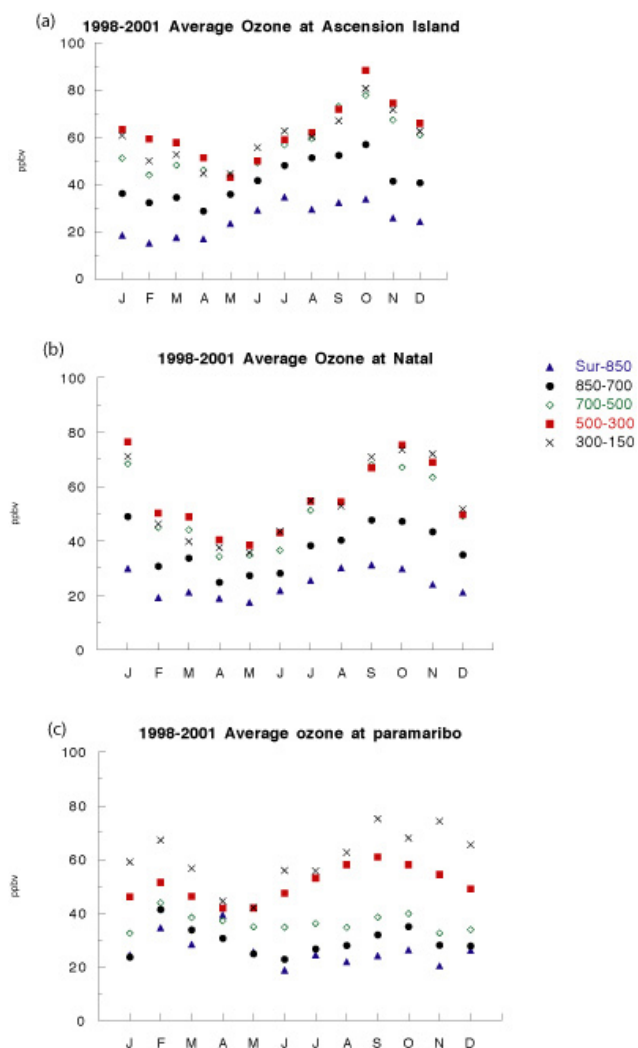
Fig. 6. 1998–2001 average ozone mixing ratios in various tropospheric layers. **(a)** Ascension Island, **(b)** Natal, **(c)** Paramaribo. Units are ppbv.

Figures 3a, b, which is follows from Figs. 2a–d shows the TCO values in specific latitudinal zones of the Tropical Atlantic from 12.5° S to 12.5° N for the TTO and CCD ozone datasets. During the months of December through February the tropospheric column ozone values are highest between 15° S and 5° S, which is probably not due to anthropogenic sources given that biomass burning is confined to the NH. The TTO data show that ozone in the latitudes between 5° S and 5° N have the highest values from the months of April through September, while in the CCD data this latitudinal zone has the highest values from April through June. The latitudinal zone of 5° N–15° N shows a 12 DU increase in the tropospheric ozone column for TTO between May and June and the maximum values are found in July. In the CCD data the increase between May and June is less than 5 DU and the maximum value is found during July, but is approximately

6–8 DU less than the TTO value. The TOR TCO latitudinal variations are also similar, but the highest TCO values are found in the latitude band of 5–15° N from May through August (not shown).

3.2 1998–2001 tropical atlantic tropospheric column ozone (TCO), ozonesonde measurements and lightning

We use 1998–2001 as the period to examine Tropical Atlantic TCO values when satellite and ozonesonde data are available. Lightning, wind and fire data are used to identify sources of O_3 and their transport from the source regions. Figures 4a–d show seasonal CCD TCO distribution during 1998–2001 over the Tropical Atlantic Ocean and adjacent land areas. The ozonesonde stations of Ascension Island (7.98° S, 14.42° W), Natal (5.42° S, 35.38° W) and Paramaribo (5.81° N, 55.21° W) are shown for reference. During NH winter (DJF), TCO values are higher over the Southern Tropical Atlantic Ocean and there is a relative minimum between 5–10° N, which is downstream of biomass burning in West Africa (Fig. 4a). The high TCO values in the Southern Hemisphere have been linked to lightning in Central Africa and South America (Thompson et al., 2000; Martin et al., 2002; Edwards et al., 2003; Jenkins et al., 2003).

During NH spring (MAM), TCO values are reduced over much of the SH tropics (Fig. 4b) but remain relatively high (42 DU) off the coast of Central Africa (5° S, 9° E). However, by JJA this area has expanded into the NH with the largest tropospheric column ozone values (>42 DU) found over land areas in West Africa (7° N) and off the coast of Central Africa (7.5° S) (Fig. 4c). During SON the highest TCO values are found in the South Tropical Atlantic and extend westward over South America and eastward over Africa toward the Indian Ocean (Fig. 4d). The 1998–2001 CCD data predicts that the highest TCO values would be found at Ascension Island, followed by Natal and Paramaribo.

Figure 5 compares three satellite derived ozone datasets (CCD, TTO, TOR) during MAM and JJA 1998–1999. All three datasets show a maximum in TCO values off the coast of Central Africa during MAM (Figs. 5a–c). However, the TTO and TOR values are considerably smaller (>4 DU) than the CCD values. The source of the elevated TCO values are associated with land areas of Central Africa, but biomass burning is minimal during this season. The elevated TCO values off the coast of Central Africa are likely associated with lightning in Central Africa during MAM (see below). During JJA, the area of elevated TCO values has expanded both westward and northward (Figs. 5d–f). A maximum in TCO values are found over West Africa and the adjacent waters during this period in the 3 datasets. The TTO and TOR datasets place the highest TCO values over West Africa rather than the Southern Hemisphere. This maximum occurs in spite of the fact that is it wet season in West Africa and biomass burning is confined to the Southern Hemisphere. We
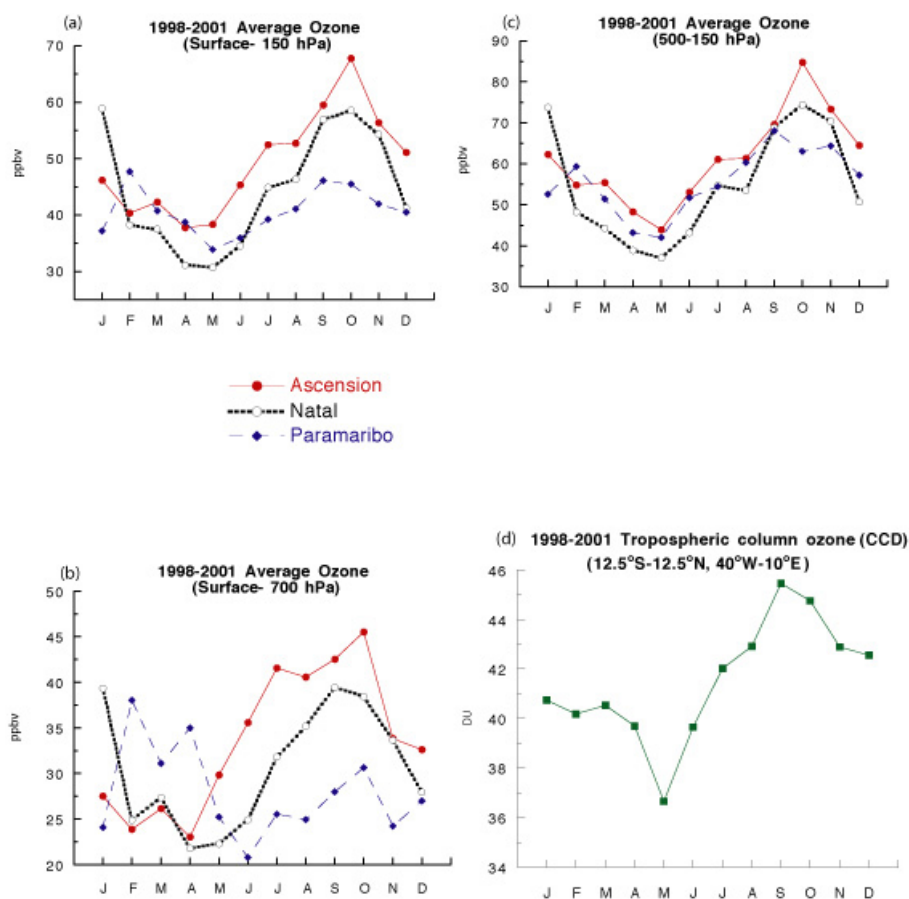
Fig. 7. 1998–2001 average ozone mixing ratios (ppbv) at Ascension Island, Natal and Paramaribo from **(a)** surface to 150 hPa, **(b)** Surface to 700 hPa, **(c)** 500 to 150 hPa. **(d)** 1998–2001 CCD tropospheric column ozone for 12.5° S–12.5° N, 40° W–10° E.

show below that the most likely cause of the elevated TCO values is associated with lightning in West Africa.

Figures 6a–c show the 1998–2001 annual cycle of average ozone mixing ratios for specific layers at Ascension Island, Natal and Paramaribo based on SHADOZ ozonesonde launches. The three sites show: (a) the lowest ozone mixing are found in April or May followed by an increase during NH summer and autumn seasons. (b) The highest ozone values are found in the upper troposphere. (c) The ozone values in the lower troposphere at Paramaribo are relatively constant, while relatively large variations are found at Ascension Island and Natal.

The surface-150 hPa ozone mixing ratio are highest at Ascension Island followed by Natal and Paramaribo, consistent with the 1998–2001 CCD satellite estimates (Fig. 7a). The ozonesondes also show a minimum in ozone mixing ratios during the month of May followed by a significant increase, which reaches its maximum value during September. There is approximately a 25–30 ppbv increase in the surface-150 hPa layer between May and September at Ascension Island. In the lower troposphere (surface-700 hPa), ozone mix-

ing ratios have a minimum during April at Ascension Island and Natal and rises steeply at Ascension Island from April through July (Fig. 7b). The rise in lower tropospheric ozone is delayed slightly at Natal with the largest increase found between July and September. At Paramaribo, the ozone mixing ratios in the lower troposphere are highest from February through April (likely associated with biomass burning transported from Africa) and remain relatively low during the NH summer and autumn seasons.

Ozone in the middle/upper troposphere (500–150 hPa) shows a relative minimum during the month of May at each site (Fig. 7c). The ozone then increases steadily at each site reaching a maximum value in September or October. The ozone mixing ratios increase by 25–40 ppbv in the upper troposphere between May and September even at Paramaribo when biomass burning is found in the Southern Hemisphere and the values in the lower troposphere are relatively constant. The annual pattern in the upper troposphere ozonesonde data is similar to the CCD 1998–2001 TCO values when averaged over the Tropical Atlantic (12.5° S–12.5° N, 40° W–10° E) (Fig. 7d). The CCD shows a
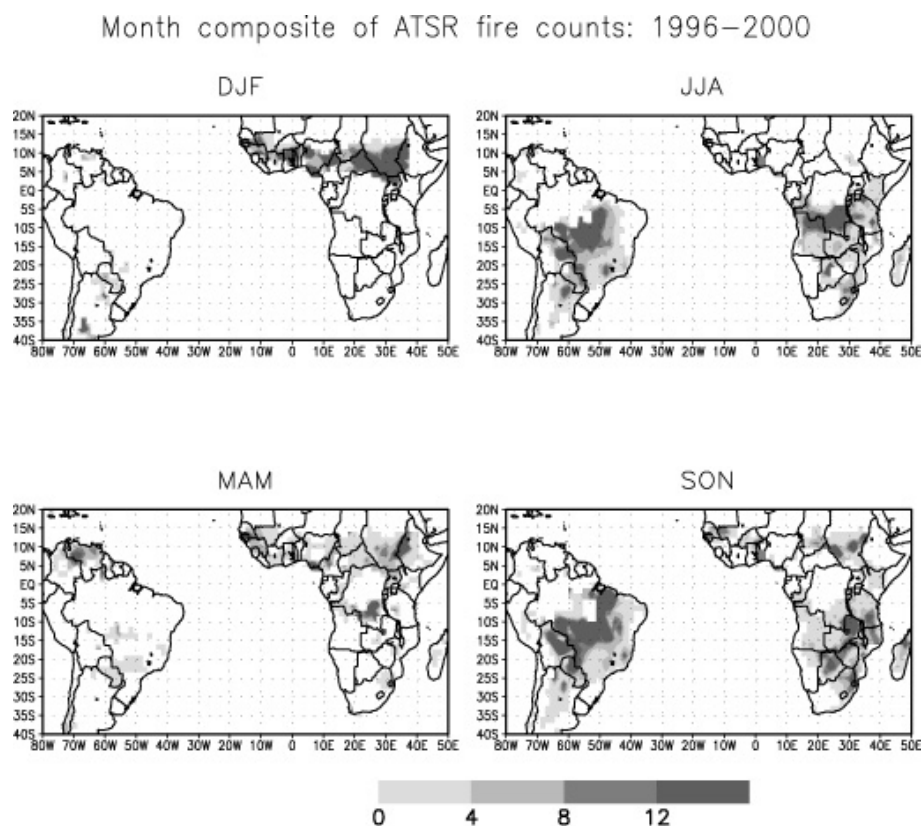
Fig. 8. ATSR fire count during 1996–2000 for: **(a)** DJF, **(b)** MAM, **(c)** JJA, **(d)** DJF. Number of fires per pixel.

minimum in tropospheric column ozone values during May and a maximum value during September with an increase of 10 DU during this time period.

Since there is likely a anthropogenic contribution to ozone especially in the lower troposphere, Figs. 8a–d identifies fires associated with biomass burning during 1996–2000. During DJF, the fires are concentrated in the NH, reaching a minimum in MAM and then are confined to the SH during JJA. During SON, fires are found in Eastern Africa and in some parts of the Northern Hemisphere near 10° N (Fig. 8d). The pattern of fires here are consistent with the study of Duncan et al. (2003). During DJF, fires are confined primarily to West Africa, while during JJA fires are found over Africa (Central and Eastern) and South America (Brazil). However, during JJA the fires are not located near the Equator or Northern Hemisphere Tropics where TCO values are increasing (Fig. 4). This is especially true near the coast of West Africa where high TCO values are found during JJA.

Biomass burning from Central Africa is a likely candidate for the increase in lower tropospheric ozone mixing ratios that are found at Ascension Island and Natal from JJA through SON (Fig. 7b). Easterly winds can transport biomass burning emission and aerosols from Central Africa towards the Tropical Atlantic and is likely the cause for increasing the lower troposphere ozone mixing ratios in Natal from June through September (Kirchhoff et al., 1996). During SON, fires are confined primarily to eastern parts of Africa and South America (Brazil) (Fig. 8d) and consequently, any significant contributions of biomass burning from these regions to the Tropical Atlantic Ocean must require long-range transport (>1000 km).

Figure 9a shows that the total number of flashes for land areas (South America and Africa) in latitudes between 15° S–15° N and longitudes 80° W–30° E for the period of 1998–2001 as detected from LIS overpasses. The smallest and largest numbers of flashes occur during DJF and SON respectively. Figure 8b shows that during DJF the majority of lightning flashes are from the latitudinal zones of 15° S–5° S and 5° S–5° N. During MAM there is a poleward shift in lightning flashes with majority occurring in the zones of 5° S–5° N and 5° S–15° N. This is just upstream of where elevated TCO values are found during MAM (Figs. 4, 5). During JJA, the period when TOC values begin to increase substantially, the largest numbers of flashes are found in the latitude band of 5° N–15° N. During SON, when TOC values are highest over the Tropical Atlantic Ocean a large number of flashes are found at all three latitude zones. Over the annual cycle, the latitudes of 15° S–5° S produce the largest number of flashes from October through January (NH autumn/winter) (Fig. 9c). The latitude band of
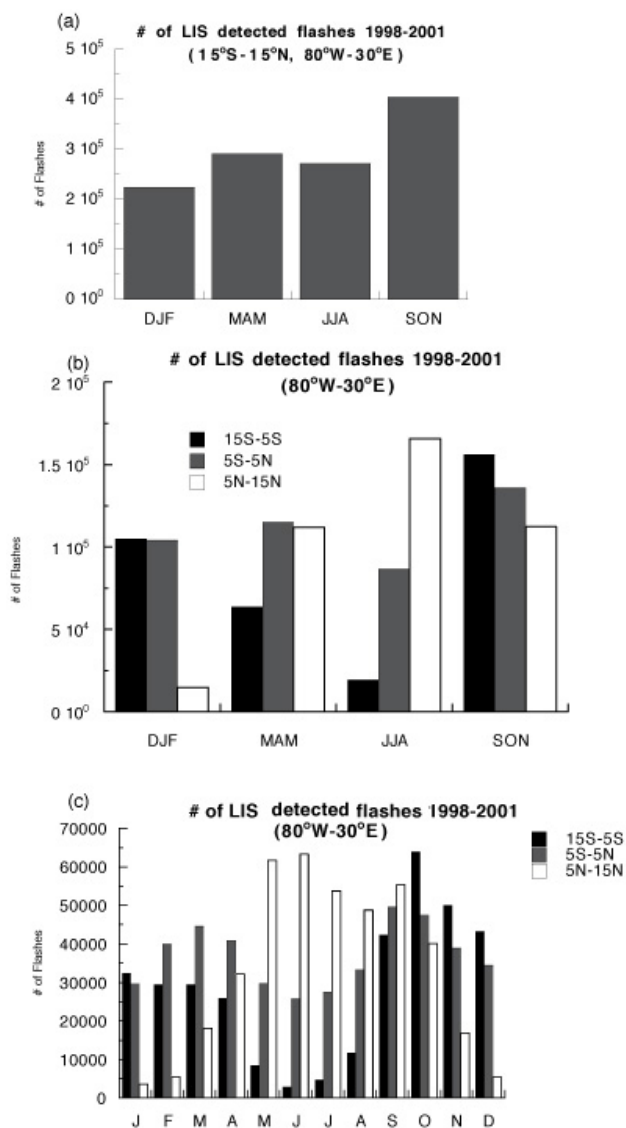
Fig. 9. 1998–2001 LIS seasonal lightning statistics: **(a)** total number of seasonal flashes **(b)** distribution of seasonal flashes for latitude zones of 15° S–5° S, 5° S–5° N, 5° N–15° N, **(c)** distribution of monthly flashes for latitude zones of 15° S–5° S, 5° S–5° N, 5° N–15° N.

5° S–5° S produces the largest number of flashes from February through April (NH winter/spring) and the latitude band of 5° N–15° N produces the largest number of flashes from May through September (NH late spring/summer/early autumn). This time period overlaps with the significant increase in TOC values from the TOMS satellite measurements.

During DJF, MAM and JJA the majority (>60%) of lightning flashes are associated with the African continent (Fig. 10a). During SON the distribution of lightning flashes between Africa and South America is nearly equal. Figures 10b–d show the number of lightning flashes in the three latitude zones for the continents of Africa and South Amer-

ica during 1998–2001. In the most southern latitudes (15° S–5° S), there are more flashes in South America in three of the four seasons with the exception of MAM. The opposite pattern is found in the other two latitude zones (5° S–5° N and 5° N–15° N) where lightning in Africa is dominant throughout the year when compared to South America (Figs. 10c, d). These results are consistent with lightning distribution by the Optical Transient Detector (OTD) (Christian et al., 2003). They show that the highest flash densities are found in Central Africa followed by regions in South America. The number of lightning flashes is lower by a factor of 4–5 during MAM relative to SON for South America in the latitude zones of 15° S–5° S (Fig. 10b). This implies that there is either less frequent convective activity or that convective intensity is reduced during MAM relative to SON in South America. It is also possible that aerosols from biomass burning during SON can alter the microphysical properties (possibly serving as ice nuclei) of deep convection leading to more lightning.

Figures 11–14 show the DJF, MAM, JJA and SON middle/upper tropospheric zonal winds and streamlines at 500, 400, 300, 200 hPa during 1998–2001. The ozonesonde stations of Ascension Island, Natal, and Paramaribo are shown for reference. Easterly winds are persistent over the Tropical Atlantic at 500, 400 hPa during all seasons with a migration toward the Northern Hemisphere from DJF through JJA (Figs. 11–14a,b). This wind pattern would be conducive to the westward transport of NO_x and O_3 associated with lightning from Central Africa (15° S–5° N) during DJF, MAM and SON and from West Africa (5° N–15° N) during JJA. At lower pressure levels (300, 200 hPa), westerly winds become more important during DJF, MAM and SON making South America a potential source of ozone enrichment via lightning in the upper troposphere (Figs. 11–14c, d).

During DJF, the ozonesonde stations are under the influence of 200 hPa westerly winds, which originate over South America and have been linked to elevated tropical Atlantic ozone via lightning (Edwards et al., 2003). A similar wind pattern also exists during MAM at 200 hPa when lightning in South America is less frequent and could help to explain why upper troposphere ozone mixing ratios are significantly reduced at the three ozonesonde stations during this period (Fig. 6). Moreover, it would explain why TCO values in the western Atlantic near South America are significantly reduced between DJF and MAM (Figs. 4a, b). During SON the ozonesonde stations are under the influence of westerly winds from South America when lightning is the most frequent and should contribute to elevated ozone mixing ratios in the upper troposphere.

During JJA, however, easterly flow is dominant in the upper troposphere and is associated with the 200 hPa Tropical Easterly Jet (TEJ), which crosses much of Africa from India (Fig. 13d). Consequently, O_3 and NO_x enrichment over the Tropical Atlantic and at the ozonesonde stations is likely associated with lightning activity over West Africa.
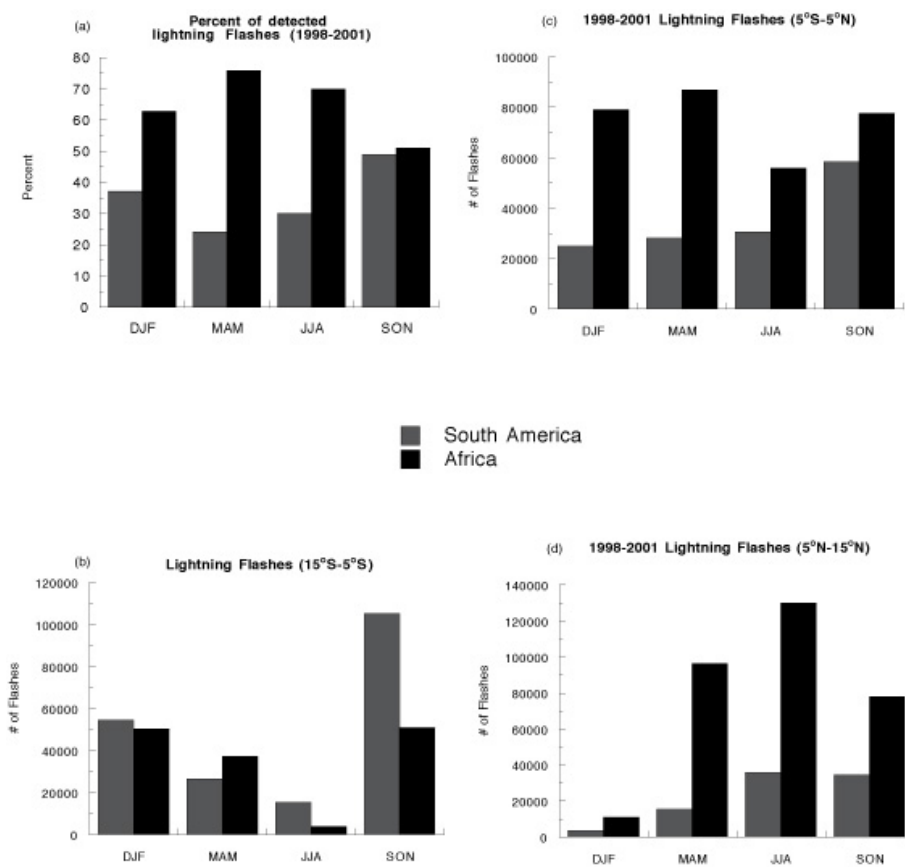
Fig. 10. (a) Percentage of seasonal flashes in South America and Africa, (b) number of detected lightning flashes in 15° S–5° S for Africa and South America, (c) number of detected lightning flashes in 5° S–5° N for Africa and South America, (d) number of detected lightning flashes in 5° N–15° N for Africa and South America.

4   Summary and discussion

In this paper we have examined the annual evolution of tropospheric ozone over the Tropical Atlantic in connection to biomass burning, lightning and large-scale transport with an emphasis on 1998–2001. Our findings show:

- The highest TCO values are found in the hemisphere opposite of biomass burning during DJF and JJA (Tropical Ozone Paradox). During DJF, biomass burning is concentrated in the Northern Hemisphere (West Africa), but elevated ozone levels over the South Tropical Atlantic are due to lightning (NO_x production) in South America and Central Africa. During JJA, biomass burning is concentrated in the Southern Hemisphere (southern sector of Central Africa, South America), but elevated ozone levels over the Equatorial Atlantic (5° S–10° N) are probably due to lightning (NO_x production) in West Africa and Central Africa to a lesser extent.

- During MAM, the highest tropospheric column ozone values are found off the western coast of Central Africa and are caused by lightning and the production of NO_x

over Central Africa. Biomass burning is at a minimum in Africa and South America during MAM.

- In the latitude zone of 15° S–15° N, the majority of lightning flashes are from Africa except during SON. During SON, the majority of lightning flashes occurring in the latitudinal zone of 15° S–5° S are from South America.

- During SON, lightning from South America, West and Central Africa combine to produce the Tropical Atlantic Ozone Maximum. However, the conditions leading up to the SON Tropical Atlantic Ozone Maximum develop during JJA. Biomass burning in South America and Eastern Africa and its subsequent transport to the Tropical Atlantic Ocean is also an important factor (Jenkins et al., 1997).

A number of observational studies have suggested that the high ozone in the troposphere primarily during Northern Hemisphere Autumn (SON) is associated with biomass burning in Africa and South America. This may be true in the lower troposphere but does not necessarily explain the
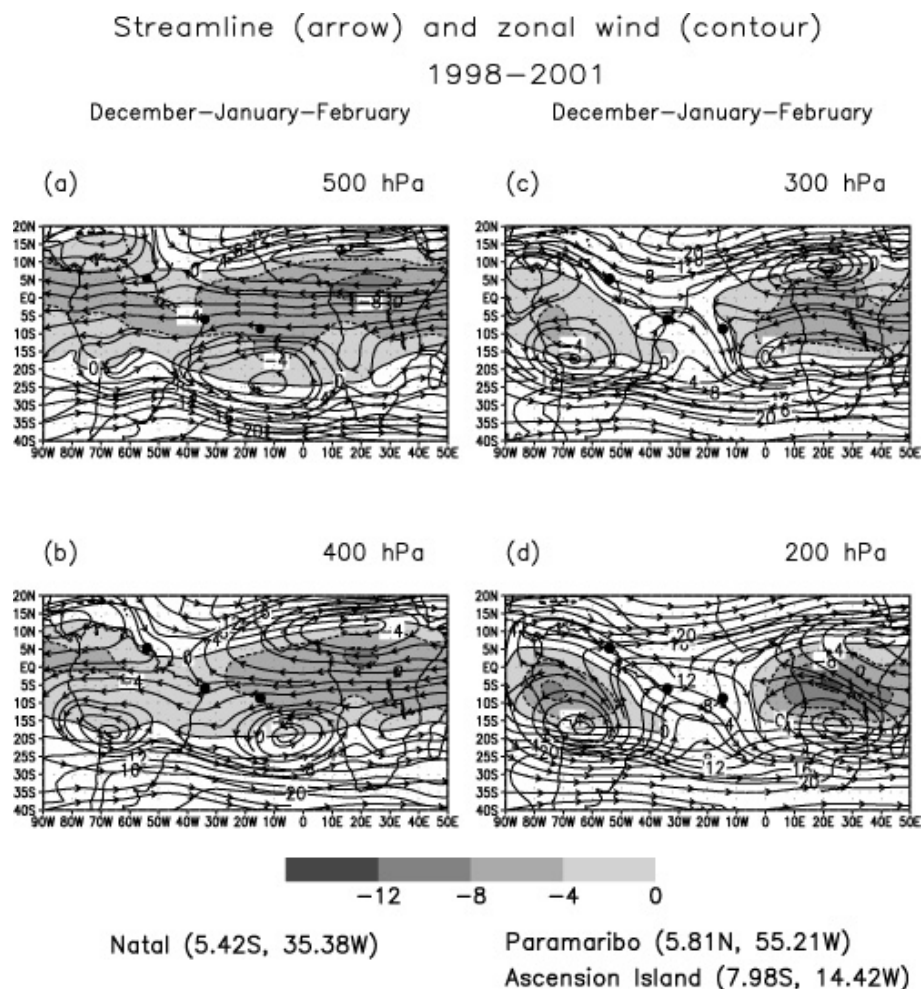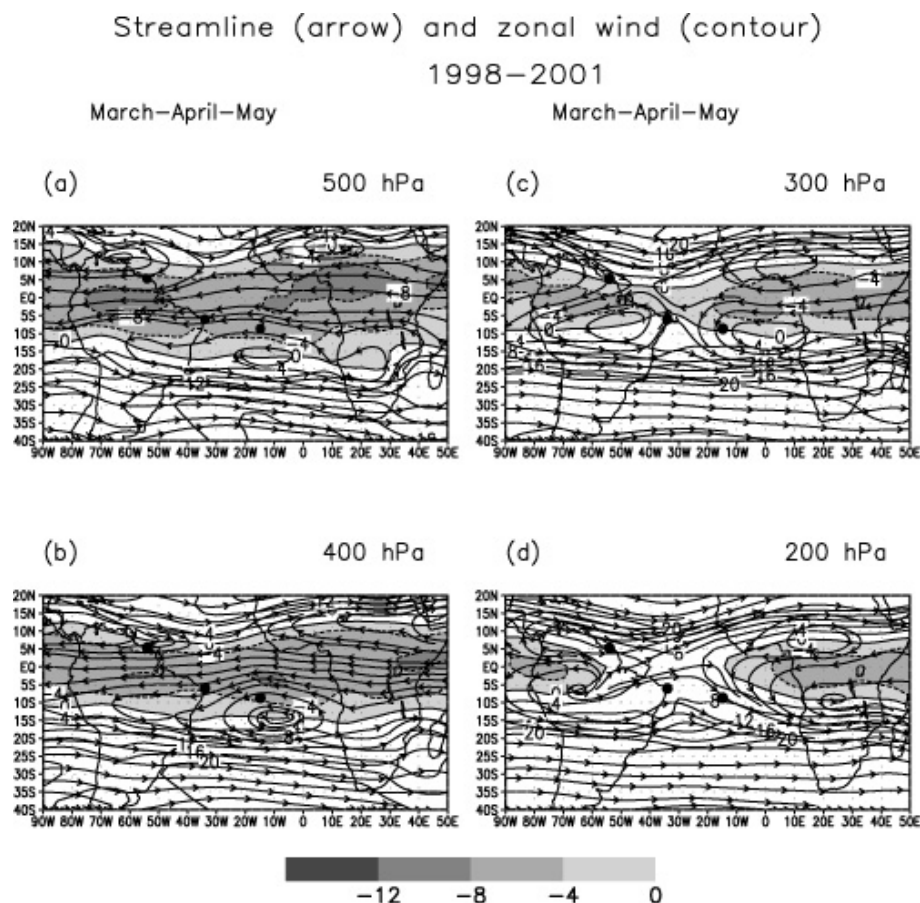
Fig. 11. 1998–2001 DJF middle/upper troposphere zonal winds and streamlines for **(a)** 500 hPa, **(b)** 400 hPa, **(c)** 300 hPa, **(d)** 200 hPa. Shaded regions are easterly winds. Units are m-s^{-1}.

high ozone mixing ratios that are typically found in the upper troposphere (Weller et al., 1996). One can explain these high values through biomass burning if there is deep convection, which can vent pollutants from the Planetary Boundary Layer into the upper troposphere. This may be the case for South America and Southeastern Africa where low level winds in South America and Central Africa can transport pollutants westward into regions of deep convection (Kirchhoff et al., 1996; Jenkins et al., 1997; Jenkins, 2000). Weller et al. (1996) suggests that stratospheric intrusions of air could also explain high values in the upper troposphere. Such intrusions could take place through the entrainment of stratospheric air associated with deep convection.

Our analysis presents a slightly different version of the Tropical Atlantic Ozone Maximum during SON. We suggest that lightning, based on LIS observations, is to a first order approximation responsible for the SON tropospheric ozone maximum with long-range transport of pollutants and

stratospheric intrusions being secondary. Even if these other processes are important to enriching tropospheric ozone they are associated with deep convective processes, which includes lightning. Ozone production during SON adjacent to the Tropical Atlantic may be considerably higher given three regional sources (South America, Central Africa, West Africa) for lightning. The modeling study of Moxin and Levy (2000), suggests that lightning is responsible for the Tropical South Atlantic tropospheric ozone maximum during SON. The modeling study of Martin et al. (2002) suggests a 10–14 DU enhancement of ozone from lightning.

Some challenges to the modeling community are simulating deep convective processes and lightning events. In order to examine deep convective processes associated with vertical transports of ozone/ozone precursors, global climate models or mesoscale models with fine horizontal resolutions (10–20 km for non-hydrostatic conditions) will be necessary. In Jenkins et al. (1997), deep convective processes were
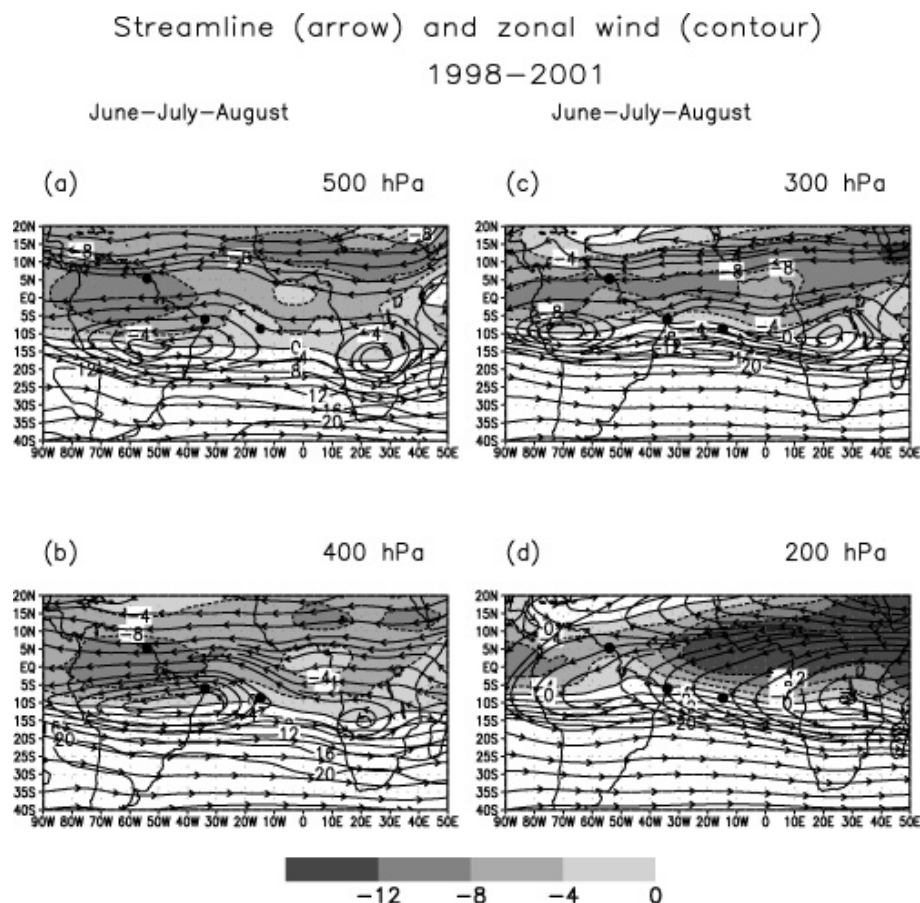
Fig. 12. 1998–2001 MAM middle/upper troposphere zonal winds and streamlines for **(a)** 500 hPa, **(b)** 400 hPa, **(c)** 300 hPa, **(d)** 200 hPa. Shaded regions are easterly winds. Units are m-s^{-1}.

examined at 99, 33 and 11 km grid spacing. The most vigorous convection with vertical velocities greater than 10 m/sec was found using the 11 km grid. Strong updrafts are also a requirement for diagnostic or prognostic modeling of lightning. The simulation of mixed phased precipitation, however, is also a significant requirement for lightning production. Very fine GCM simulations are unlikely in the near future and mesoscale/cloud resolving models will only give a limited view of modeled deep convection and lightning. Mixed phase physics, however, can be incorporated into GCMs, which will provide some diagnostic or prognostic measures of lightning processes although the vertical velocities will tend to be underestimated.

Martin et al. (2002) suggests that TTO derived tropospheric column ozone values are significantly overestimates when compared with MOSAIC data during JJA. However, the ozonesonde data shows a pattern that is consistent with the TTO satellite measurement using the MR technique with a significant rise in ozone values after the month of May. The TOR satellite measurements also show enhanced TCO values over West Africa during JJA. The rise in ozonesonde mixing ratios is most pronounced in the middle/upper tro-

posphere making lightning a likely cause. Drummond et al. (1988) also found enhanced upper troposphere NO_x values, indirectly supporting the TTO satellite TOC estimates, during the month of June over the Tropical Atlantic Ocean and suggested that lightning processes over continents may have been responsible. During JJA, we have found that lightning over West Africa is likely a significant contributor to NO_x and the subsequent production of O_3. Recent studies (Mohr and Zipser, 1996; Nesbitt et al., 2000; Toracinta and Zipser, 2001; Sealy et al., 2003) using the data from the polar orbiting satellites and the TRMM satellite (Kummerrow et al., 1998) have examined precipitation and lightning properties, using the Optical Transient Detector (OTD) and (LIS) data, in the tropics and West Africa.

Nesbitt et al. (2000), show that lightning is typically associated with precipitation features that contain a significant ice content. Mesoscale convective systems (MCSs), which include squall line, mesoscale convective complexes and non-squall tropical clusters, have low polarization temperatures because of their high ice content in Africa. Moreover, MCSs were identified with the largest number of lighting flashes in Africa and South America (Nesbitt et al., 2000).
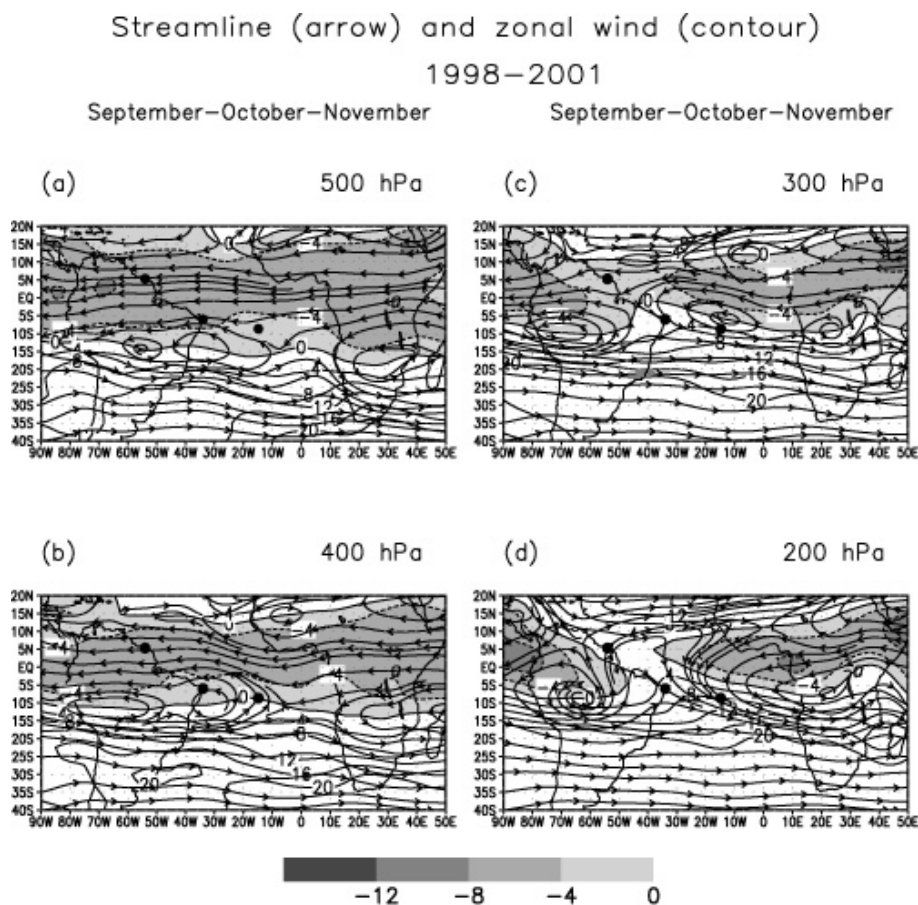
Fig. 13. 1998–2001 JJA middle/upper troposphere zonal winds and streamlines for **(a)** 500 hPa, **(b)** 400 hPa, **(c)** 300 hPa, **(d)** 200 hPa. Shaded regions are easterly winds. Units are m-s^{-1}.

Sealy et al. (2003) have found that the region closest to the Sahara desert–the Sahel (10–18° N), produced the highest number of flashes relative to the wetter Guinea region (5–10° N) during May–September 1998–2000. A higher frequency of lightning flashes in the Sahel occurred at night, which is consistent with studies of MCSs showing a distinct diurnal pattern, with the largest mean radius occurring between 18:00 and 22:00 LST (Mathon and Laurent, 2000). Consequently, nighttime lightning may lead to an accumulation of NO_2, NO_3 and N2O5 until sunrise of the next day thereby serving as a source of ozone in the absence of deep convection. Moreover, there may be a direct link between lightning, ice content and aerosols. In the case of West Africa, the aerosols come from Saharan dust and would serve as ice nuclei in updrafts of developing convective cells. Strong vertical motions, ice particles, graupel and super-cooled water would be associated with the high flash rates found in the Sahel, thereby enriching upper troposphere NO_x and ozone mixing ratios. Demott et al. (2003) have found high concentrations of ice nuclei in Saharan dust and show that the ice nuclei concentrations exceeded the typical values by a factor of 20 to 100.

The results presented in this paper imply that increases in TOC values during JJA as implied by TTO and TOR satellite estimates are real and caused by lightning. There is an opportunity to determine the cause for the discrepancies between satellite estimated TOCs and the MOSAIC data during the Northern Hemisphere summer season. The proposed African Monsoon Multidisciplinary Experiment (AMMA) in West Africa, which has a summer special observing period (http://medias.obs-mip.fr/amma, http://www.joss.ucar.edu/amma) during JJA of 2006, could help to resolve this issue. This can be achieved through the launch of ozonesondes, ship and aircraft measurements of O_3 and NO_x and concurrent satellite measurements.

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Fig. 14. 1998–2001 SON middle/upper troposphere zonal winds and streamlines for **(a)** 500 hPa, **(b)** 400 hPa, **(c)** 300 hPa, **(d)** 200 hPa. Shaded regions are easterly winds. Units are m-s^{-1}.

References

Arino, O. and Melinotte, J.-M.: The Fire Atlas, Earth Observation Quarterly, 50, 1995.

Christian, H. J., Blakeslee, R. J., Boccippio, D. J., et al.: Global frequency and distribution of lightning as observed from space by the Optical Transient Detector, J. Geophys. Res., 108, doi:10.1029/2002JD002347, 2003.

DeCaria, A. J., Pickering, K. E., Stenchikov, G. L., Scala, J. R., Stith, J. L., Dye, J. E., Ridley, B. A., and Laroche, P.: A cloud-scale model study of lightning-generated NO$_x$ in an individual thunderstorm during STERAO-A, J. Geophys. Res., 105, 11 601–11 616, 2000.

Demott, P. J., Sassen, K., Peollot, M. R., et al.: African dust aerosols as atmospheric ice nuclei, Geophys. Res. Lett., 30, doi:10.1029/2003/GL017410, 2003.

Drummond, J. W., Ehhalt, D. H., and Volz, A.: Measurements of Nitric Oxide between 0–12 km altitude and 67° N to 60° S latitude obtained during STRATOZ III, J. Geophys. Res., 93, 15 831–15 849, 1988.

Duncan, B. N., Martin, R. V., Staudt, A. C., Yevich, R., and Logan, J. A.: Interannual and seasonal variability of biomass burning emissions constrained by satellite observations, J. Geophys. Res.,

108, dio:10.1029/2002JD002378, 2003.

Dye, J. E., Ridley, B. A., Skamarock, W., et al.: An Overview of the STERAO-Deep Convection Experiment with Results for the 10 July Storm, J. Geophys. Res., 105, 10 023–10 045, 1999.

Edwards, D. P., Lamarque, J.-F., Attie, J.-L., et al.: Tropospheric Ozone over the Tropical Atlantic: A satellite perspective, J. Geophys. Res., 108, doi:10.1029/2002JD002927, 2003.

Fishman, J., Fakhruzzaman, K., Cros, B., and Nganga, D.: Identification of Widspread Pollution in the Southern Hemisphere Deduced from Satellite Analyses, Science, 252, 1693–1693, 1991.

Fishman, J., Hoell Jr., J. M., Bendura, R. D., McNeal, R. J., and Kirchhoff, V. W. J. H.: NASA GTE TRACE A Experiment (September–October 1992): Overview, J. Geophys. Res., 101, 23 865–23 879, 1996.

Fishman, J., Wozniak, A. E., and Creilson, J. K.: 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, 2003.

Hudson, R. D. and Thompson, A. M.: Tropical tropospheric ozone from total mapping spectrometer by a modified-residual method, J. Geophys. Res., 103, 22 129–22 145, 1998.

Jacob, D. A., Heikes, B. G., Fan, S.-M., et al.: 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, 24 069–24 082, 1996.

Jenkins, G. S., Mohr, K., Morris, V. R., Arino, O.: The Role of Convective Processes over the Zaire and Congo Basins to the Southern Hemisphere Ozone Maximum, J. Geophys. Res., 102, 18 963–18 980, 1997.

Jenkins, G. S.: TRMM satellite estimates of convective processes in Central Africa during September, October, November 1998: implications for elevated Atlantic tropospheric ozone, Geophys. Res. Lett., 27, 1711–1714, 2000.

Jenkins, G. S., Ryu, J.-H., Thompson, A., and Witte, J.: 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. Dis., 3, 5061–5098, 2003.

Jenkins, G. S., Ryu, J.-H., Thompson, A., and Witte, J.: 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., doi.10.1029/JD2002003297, 2003.

Kalnay, E. M., Kanamitsu, M., Kistler, R., et al.: The NCEP/NCAR 40-Year Reanalysis Project, Bulletin of the American Meteorological Society: Vol. 77, No. 3, pp. 437–472, 1996.

Kim, J. H., Newchurch, M. J., and Han, K.: Distribution of Tropical Ozone Determined by the Scale-Angle Method Applied to TOMS Measurements, J. Atmos. Sci., 58, 2699–2708, 2001.

Kirchhoff, V. W. J. H, Alves, J. R., Da Silva, F. R., and Fishman, J: Observations of ozone concentrations in the Brazilian cerrado during the TRACE A field expedition, J. Geophys. Res., 101, 24 029–24 042, 1996.

Kummerow, C., Barnes, W., Kozu, T., Shiue, J., and Simpson, J.: The Tropical Rainfall Measuring Mission (TRMM) sensor package, J. Atmosp. Oc., 15, 809–817, 1998.

Martin, R. V, Jacob, D. J, Logan, J. A., et al.: Detection of a lightning influence on tropical tropospheric ozone, Geophys. Res. Lett., 27, 1639–1642, 2000.

Martin, R. V., Jacob, D. J., Logan, J. A., et al.: Interpretation of TOMS observations of tropical tropospheric ozone with a global model and in situ observations, J. Geophys. Res., 107, doi:10.1029/2001JD001480, 2002.

Mathon, V. and Laurent, H.: Life cycle of Sahelian mesoscale convective cloud systems, Q. J. R. Meteorol. Soc., 127, 377–406, 2001.

Mohr, K. I. and Zipser, E. J.: Defining mesoscale convective systems by the 85-GHz ice scattering signatures, BAMS, 77, 1179–1188, 1996.

Moxim, W. J. and Levy II, H.: A model analysis of the tropical South Atlantic Ocean tropospheric ozone maximum: The interaction of transport and chemistry, J. Geophys. Res., 105, 17 393–17 415, 2000.

Nesbitt, S. W., Zipser, E. J., and Cecil, D. J.: A census of precipitation features in the tropics using TRMM: Radar, ice scattering and lightning observations, J. Clim., 13, 4087–4106, 2000.

Pickering, K. E., Thompson, A. M., Wang, Y., et al.: Convective transport of biomass burning emissions over Brazil during TRACE-A, J. Geophys. Res., 101, 23 993–24 012, 1996.

Poulida, O., Dickerson, R. R., and Heymsfield, A.: Stratosphere-troposphere exchange in a mid-latitude mesoscale convective complex, 1, Observations, J. Geophys. Res., 101, 6823–6836, 1996.

Price, C., Penner, J., and Prather, M.: NO$_x$ from lightning 1. Global distribution based on lightning physics, J. Geophys. Res., 102, 5929–5941, 1997.

Rosenfeld, D.: TRMM observed direct evidence of Smoke from Forest Fire Inhibiting Rainfall, Geophys. Res. Lett., 26, 3105–3108, 1999.

Sealy, A., Jenkins, G. S., and Walford, S. C.: Seasonal/regional comparisons of rain rates and rain characteristics in West Africa using TRMM observations, J. Geophys. Res., 108, doi:10.1029/2002JD002667, 2003.

Stith, J., Dye, J. E., Ridley, B., et al.: NO signatures from lightning flashes, J. Geophys. Res., 104, 16 081–16 089, 1999.

Thompson, A. M., Pickering, K. E., McNamara, D. P., 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 1992, J. Geophys. Res., 101, 24 251–24 278, 1996.

Thompson, A. M. and Hudson, R. D.: 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, 1999.

Thompson, A. M., Doddridge, B. G., Witte, J. C., et al.: 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, 2000.

Thompson, A. M., Witte, J. C., McPeters, R. D., et al.: Southern Hemisphere Additional Ozonesondes (SHADOZ) 1998–2000 tropical ozone climatology. 1. Comparison with Total Ozone Mapping Spectrometer (TOMS) and ground-based measurements, J. Geophys. Res., 108, doi:10.1029/2001JD000967, 2003.

Toracinta, E. R. and Zipser, E. J.: Lightning and SSM/I ice-scattering mesoscale convective systems in the global tropics, J. Appl. Met., 40, 983–1002, 2001.

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

Ziemke, J. R., Chandra, S., and Bhartia, P. K.: Cloud slicing: A new technique to derive upper tropospheric ozone from satellite measurements, J. Geophys. Res., 106, 9853–9867, 2001.