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Tropospheric ozone over Equatorial Africa: regional aspects from the MOZAIC data

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Abstract

We analyze MOZAIC ozone observations recorded over Equatorial Africa, from April 1997 to March 2003 to give the first ozone climatology of this region. The monthly mean vertical profiles have been systematically analyzed with monthly mean ECMWF data using a Lagrangian-model (LAGRANTO). We assess the roles played by the dynamical features of Equatorial Africa and the intense biomass burning sources within the region in defining the ozone distribution. The lower troposphere exhibits layers of enhanced ozone during the biomass burning season in each hemisphere (boreal winter in the northern tropics and boreal summer in the southern tropics). The monthly mean vertical profiles of ozone are clearly influenced by the local dynamical situation. Over the Gulf of Guinea during boreal winter, the ozone profile is characterized by systematically high ozone below 650 hPa. This is due to the high stability caused by the Harmattan winds in the lower troposphere and the blocking Saharan anticyclone in the middle troposphere that prevents any efficient vertical mixing. In contrast, Central African enhancements are not only found in the lower troposphere but throughout the troposphere. The boreal summer ozone maximum in the lower troposphere of Central Africa continues up to November in the middle troposphere due to the influx of air masses laden with biomass burning products from Brazil and Southern Africa. Despite its southern latitude, Central Africa during the boreal winter is also under the influence of the northern tropical fires. This phenomenon is known as the "ozone paradox". However, the tropospheric ozone columns calculated from the MOZAIC data give evidence that the Tropical Tropospheric Ozone Column (TTOC) maximum over Africa swings from West Africa in DJF to Central Africa in JJA. This contrasts with studies based on TOMS satellite data. A rough assessment of the regional ozone budget shows that the northern tropics fires in boreal winter might contribute up to 20% of the global photochemical ozone production. This study gives the first detailed picture of the ozone distribution over Equatorial Africa that should be used to validate both global models over this region and future satellite products

ACPD

4, 3285–3332, 2004

1. Introduction

Tropospheric ozone acts both to control the oxidizing capacity of the atmosphere and as a greenhouse gas (Logan and Kirchhoff, 1986; Crutzen, 1987). Most of the oxidation of long-lived gases by hydroxyl radicals (OH) takes place in the tropics, where high UV and humidity promote the formation of OH from the photolysis of ozone (Logan et al., 1981; Thompson, 1992). Ozone is produced within the troposphere by the photochemical oxidation of hydrocarbons, methane (CH_4) and carbon monoxide (CO) in the presence of nitrogen oxides (NO_x=NO+NO_2) (Crutzen, 1974, 1988; Fishman et al., 1979; Marenco et al., 1989) and is also transported down from the stratosphere. Ozone photochemical formation occurs in two main ways: (1) a rapid formation from reactive hydrocarbons (urban pollution, biomass burning) close to their source, followed by its mixing within the troposphere, and (2) a slow and delayed formation (2 or 3 weeks) from less reactive precursors, such as CO and CH_4, during or after their redistribution in the troposphere by horizontal and vertical atmospheric motions (Logan et al., 1981, 1985). This has been summarized as "cook and mix" or "mix and cook" (Chatfield and Delanay, 1990). For example recently polluted biomass burning plumes sampled during the TROPOZ campaign showed that after only two days ozone concentrations are between 90 and 105 ppbv (Jonquieres et al., 1998).Thus the photochemical impact of pollutants must be considered both on the regional and hemispheric scales.

The equatorial African region is of great interest due to a combination of dynamical processes, high chemical activity from intense biomass burning, and the diversity of the vegetation (associated with different emissions factor when burnt).

Its main dynamical feature is the West African monsoon (a south-westerly flow, due to the strong thermal gradient between continent and ocean) in boreal summer. The zone of confluence between northeasterly (Harmattans) and southwesterly winds is represented by the intercontinental convergence zone (hereafter, ITCZ), which shifts each summer into the opposing hemisphere. The African Easterly Jet (hereafter, AEJ) is associated with these low level transport paths and the strong convection at the

vicinity of the ITCZ, and is located around 700 hPa. The equatorward side of the jet coincides with maximum cyclonic shear and may also be the birthplace of easterly waves. Two anticyclones dominate at 500 hPa. The Saharan Anticyclone is centred at 15° N–10° E and the South Atlantic Subtropical Anticyclone (thereafter SASTA, or St. Helena) which is centred at 30° S–15° W. They both contribute to the well defined and stable trade-wind inversion and the highly stratified nature of the atmosphere (Andreae et al., 1994). Compared with West Africa, East Africa presents slightly different regimes due to the orography and the proximity of the Indian Ocean. Several major structural elements can be identified: the ITCZ and the Arabian and Mascarene anticyclones. The East African low-level jet or Somali jet, originates at the northeastern tip of Madagascar (Hastenrath, 1990); and the African Easterly Jet, which is situated in the upper troposphere.

In Africa as a whole, the forest and the savanna occupy areas of 2×10^6 and 1×10^7 km^2, respectively. The annually burned dry matter biomass is estimated at 1.3×10^8 tons yr^{-1}(Delmas et al., 1991). Human activities are the primary cause of biomass burning, which serves a variety of purposes: cleaning of forest and brush land for agricultural use, conversion of forests to agricultural and pastoral lands, energy production for cooking, heating and fuel, control of pests, insects and weeds, nutrient mobilization, and removal of brush and litter (Crutzen and Andreae, 1990). Biomass burning has long been recognized as a significant source of reactive species such as CO, hydrocarbons and NO_x, which play an important role in the chemistry and the radiative budget of the troposphere (Crutzen et al., 1985; Crutzen and Andreae, 1990). Emissions of ozone precursors from biomass burning and their consequent photochemistry have been observed at different scales (Andreae et al., 1988, 1992; Marenco et al., 1990; Fishman et al., 1990; Jonquieres et al., 1998).

Africa remains a poorly understood and documented continent. Several campaigns have been conducted within the last decades to learn more about the emissions and transport of biogenic and anthropogenic trace constituents over the South Atlantic and Africa (Table 1). These experiments mainly focused on emissions from tropical ecosys-

ACPD
4, 3285–3332, 2004

tems and on the photochemistry of the lower and middle troposphere. The first results came from DECAFE (1988 – Congo); FOS/DECAFE (1991 Ivory Coast); and SAFARI/TRACE-A (1992–2000 Southern Africa) and provided evidence of biomass burning emissions. High concentrations of gases (CO, CO_2, NMHC, NO and O_3) and particles were observed in the biomass burning plume and haze layers occurring in the mid-troposphere (altitudes of 1 to 4 km) (Andreae et al., 1988, 1992). However, no measurements were made higher in the atmosphere. The few ones performed during the TROPOZ I and II campaign did confirm that fires in northern Equatorial Africa are effective in producing large amounts of ozone (photochemical ozone formation rates of 15–35 ppbv O_3.d $^{-1}$) in the middle troposphere (Jonquieres et al., 1998). During the SAFARI 2000 experiment (Swap et al., 2003), the flux of organic and elemental carbon from African savannas burning were estimated at 14±1 Tg.yr^{-1}, and the flux of nitrogen species at 2±2 Tg.yr^{-1} with much of this pollution exported from the east coast towards the Indian Ocean.

From a climatological point of view, observations of African ozone are only available from satellite observations (TOMS) that can be processed to give a total tropospheric column ozone (Fishman and Brackett, 1997; Kim et al., 2001). There is however significant uncertainty in these tropospheric ozone column products because of their insensitivity to the lower troposphere due to Rayleigh scattering. Even the latest TOMS product using the Scan-Angle Method (Kim et al., 2001) overestimates clear-sky tropospheric ozone columns in areas with particular low ozone and underestimates amounts in areas with high ozone. Because of this, the TOMS Tropical Tropospheric Ozone Contents (TTOC) does not capture the seasonal maximum due to biomass burning found in observations and models over sub-Saharan northern Africa (Martin et al., 2002). This lower tropospheric insensitivity increases the "tropical Atlantic paradox" (Thompson et al., 2000), i.e. the north-south gradient reversal in ozone over the tropical Atlantic during the northern African biomass burning season in December-February (DJF).

The lack of in-situ tropospheric measurements over Equatorial Africa restricts the assessment of its regional budget and validation of global models. The objective of

ACPD
4, 3285–3332, 2004

this paper is to establish the regional and seasonal distribution of tropospheric ozone over Equatorial Africa from the vertical profiles provided by the MOZAIC program. The MOZAIC program will be described in Sect. 2. We aim to describe the key features of the ozone distribution in Sect. 3, from the perspective of both those processes defining the vertical profiles and the seasonal cycles. To assess the role of transport processes, we analyse back-trajectories to investigate connections between the main features of the vertical ozone distributions and the regions of intense biomass burning according to three different regions. Seasonal TTOCs computed with MOZAIC data will be investigated in Sect. 4 with regards to the "tropical Atlantic paradox" and an assessment of the regional ozone production will be presented in Sect. 5 followed by the conclusions.

2. The MOZAIC database

The MOZAIC program (Measurement of Ozone and Water Vapor by Airbus In-Service Aircraft) was designed to collect ozone and water vapor data, using automatic equipment installed aboard 5 long range Airbus A340 aircraft flying regularly all over the world (Marenco et al., 1998). The program started in 1994, and more than 20 000 flights have been performed. The MOZAIC program provides measurements every 4s for ozone, water vapor. The vertical take-off speed of the aircraft is around 5–7 $m\,s^{-1}$, leading to a vertical resolution of the data of about 20-28 m. For ozone a dual-beam UV absorption instrument (Thermo-Electron, model 49-103) is used, and the measurement accuracy was estimated at ±2 ppbv +2%; details are given by Thouret et al. (1998a). An inter-comparison with the available ozone soundings has shown the ability of the MOZAIC ozone data to produce accurate and reliable ozone distributions (Thouret et al., 1998b). Note that since the end of 2001, MOZAIC also provides additional measurements of NO_y (onboard only 1 aircraft) and CO (Nédélec et al., 2003). The data set used for this analysis corresponds to measurements of ozone and water vapor recorded between April 1997 and March 2003.

In this paper, we use vertical profiles corresponding to takeoff and landing at 8 air-

ACPD
4, 3285–3332, 2004

ports regularly visited by MOZAIC aircraft. We divide the airports into 3 groups based on their geographical location: Gulf of Guinea (Abidjan, Douala and Lagos), Central Africa (Brazzaville and Luanda), and East Africa (Entebbe, Kigali, Nairobi). Figure 1 shows their locations on the map.

To study monthly mean concentrations, the ozone concentrations are vertically averaged into 20 layers, 12 between the surface and 400 hPa with a 50 hPa thickness and 8 between 400 hPa and 200 hPa with a 25 hPa thickness. This resolution is fine enough to assess the general characteristics of the monthly mean distributions. We consider the aircraft measurements during ascent and descent to be vertical profiles even though there is significant (350 km) horizontal travel between the ground and cruising altitude. Comparisons with ozonesondes (which ascend more steeply than aircraft) (Thouret et al., 1998a) and lidar (which provides a two-dimensional coverage of layers) (Stoller et al., 1999) have shown that the assumption is satisfactory.

To analyze the seasonal variations we will consider a set of three different layers 100 hPa thick, centered at 750, 500 and 250 hPa, chosen for a complete sample of the tropospheric column.

Table 2 gives the number of available daily profiles over Equatorial Africa since April 1997 as a total and per month. Note that each region is well documented within the year with at least one visited airport per region (with the exception of East Africa from February to April).

In order to present reliable ozone climatology over equatorial Africa, we need to assess the statistical significance. The methodology is based on statistical laws, like in the study of Logan (1999), which used ozonesondes data. Figure 2 gives the standard error on the ozone monthly mean (SE=$\sigma/\sqrt{N}$, with σ the square root of the sample variance and N the number of flights) as a fraction of the monthly mean versus the number of flights per month. The 95% confidence intervals for the monthly means (2SE) clearly depend on both the inherent variability of ozone and the number of measurements. Logan (1999) found that in the tropics, 20 ozone soundings were required for 1 SE to be <7.5% of the mean below 500 hPa. The tropical region is of high tropospheric ozone

variability as shown by Thompson (2003b).

Applying this criterion (the 95% confidence interval) to our data set shows that only 16 measurements are required in the LT over Gulf of Guinea for 1 SE to be <5% of the mean. In the mid troposphere (MT), 18 measurements are necessary for 1 SE to be <5% and 8 for 1 SE <7.5%. This value falls to 6 measurements in the upper middle troposphere, for 1 SE to be <12.5% of the mean, as the variability of ozone at this altitude is weaker in the tropics than mid-latitudes due the higher tropopause height that reduces the stratospheric influence. For the other regions (not shown) the variability is similar for each tropospheric level and between the sites. However fewer measurements are necessary for the LT and MT over Central Africa, with 12 and 7 measurements for 1 SE <5% respectively. For the Central Africa, UT, 6 measurements are required, for 1 SE<5% this clearly shows the different degrees of variability seen over Africa.

These results show that the ozone monthly means over each airport should be representative with 15 measurements over Gulf of Guinea, 13 over East Africa and 8 over Central Africa. Over the main visited cities (Brazzaville, Lagos, Abidjan), these criteria are satisfied (Table 2).

One of the principal aims of this study is the monthly and seasonal variability of the ozone distribution. Thus, it is reasonable to investigate whether or not these ozone signals are correlated with the main transport processes (discussed in the introduction) that also have a monthly and seasonal dependence. Figure 5 shows the backward trajectories used in this study. They were computed with the Lagrangian-model (LAGRANTO, Wernli and Davies, 1997; Wernli, 1997) using the winds from the ECMWF mean monthly analysis ($0.5^{\circ} \times 0.5^{\circ}$ latitude longitude grid, 61 vertical model level) calculated with a time step of 30 min. As is typical of these Lagrangian approaches convection and turbulence are not fully represented in these calculations. Monthly analyses capture the main features of the synoptic circulation needed to understand the ozone climatologies. Comparisons between back trajectories computed with monthly and 6-h ECMWF analyses did not lead to significant differences in the interpretation of

ACPD
4, 3285–3332, 2004

the regional-scale origin of air parcels. This agrees with the temporal stability of the features seen in the ozone profiles as manifested by the low number of observations needed for the 95% confidence interval.

3. Distribution and variability of tropospheric ozone: seasonal and regional dependence. Mean vertical profiles and the seasonal cycles over Equatorial Africa

3.1. Gulf of Guinea

Figure 3a shows the monthly mean vertical profiles of ozone from December to November, over Gulf of Guinea, averaged between April 1997 and March 2003. Figure 4 represents the seasonal cycle for layers centered on 750 hPa (Fig. 4a), 500 hPa (Fig. 4b) and 250 hPa (Fig. 4c) as described previously.

3.1.1. Lower troposphere

During the Northern Hemisphere dry season, from mid November to mid March (hereafter named DJF), there is a maximum in the concentration of ozone in the lower troposphere. This maximum starts in November over Lagos and Douala, with 50 to 55 ppbv, between 750 and 600 hPa. The highest concentrations are observed between December and February, reaching 60 to 80 ppbv with the overall maximum occurring in January over Lagos. The polluted layers are 150 hPa to 300 hPa thick, with a central pressure at 750–700 hPa. This enhancement is estimated to be about +20–30 ppbv over the background concentration level. From the end of February the concentrations decrease.

In order to explain the origin and formation of the observed layer, Fig. 5a represents trajectories initialized in the ozone maximum at 750 hPa over Lagos and traced backward for 5 days using the monthly field of January 2000. Trajectories show a

ACPD
4, 3285–3332, 2004

northeasterly source of the air mass in the lower troposphere (820–860 hPa) over the eastern African region (Central Africa, Sudan, Zaire, Uganda and Ethiopia). Over these countries and more generally throughout the northern tropics (see Fig. 6a), biomass burning during the dry season is an important source of ozone precursors (Hao et al., 1994).

Figure 7 shows the monthly mean vertical profiles of the MOZAIC dynamical and thermal (u, v, θ), and chemical (O_3, RH, CO) parameters, during January over Lagos. The boundary layer is fed by maritime air masses embedded in the monsoon flow up to 900 hPa and characterized by southwesterly winds and high relative humidity and low ozone (40–60 ppbv). High CO concentrations (500–600 ppbv) are due to local pollution (Lagos is a large city). The strong ozone vertical gradient is characteristic of dry deposition. The monsoon flow is capped by a Harmattan layer (900–750 hPa) with northeasterly winds bringing ozone-rich (>65 ppbv), dry and relatively stable air masses of continental origin. The contribution of thermal winds associated with the meridional temperature gradient between the Gulf of Guinea and the continent, peaks into the Africa easterly Jet (7 ms^{-1} for monthly-mean and up to 10–15 ms^{-1} on individual profiles between 750 and 550 hPa). In the AEJ, the ozone concentration provided by easterly air masses decreases with height (65 to 50 ppbv). Very similar vertical structures are measured at Douala and at Abidjan, so the ozone concentration gradient along the African Easterly Jet is flat. Moreover, the AEJ lies just south of the band of fires (Figs. 6a and 5b) over a large longitudinal band and should thus be constantly sustained with ozone precursors by northeasterly winds. This suggests that after ozone is initially formed on a regional scale in the Harmattan layer by rapid photochemical processes over the band of fires (Jonquieres et al., 1998; Singh et al., 1996), the slower photochemical enhancements of ozone during the transport along the AEJ is either weak or balanced by mixing processes; otherwise the zonal gradient of ozone should be stronger. A Lagrangian aircraft campaign and model studies would be needed to investigate whether the dominant process creating the dry-season ozone maximum can be attributed to rapid and regional photochemistry or to slower and continental-scale

photochemistry.

From June to August (JJA) (Fig. 3a), the lower troposphere exhibits much lower ozone concentrations than during the dry season. Lagos and Abidjan show depleted ozone with 20 to 30 ppbv from the ground up to 780 hPa. The monsoon event with southwesterly oceanic flow is particularly active in this region, from the ground up to 800 hPa with RH of around 90%. The clean oceanic air combined with dry deposition and wet destruction in the ITCZ clouds leads to a decrease in the ozone concentration in the boundary layer, and strong ozone gradient.

Above this ozone-depleted layer, an ozone enhancement is measured over Lagos in July, from 600 to 500 hPa, with 45 ppbv ozone (Fig. 3a). We observe similar profiles over Lagos and Abidjan in August, with an enhanced ozone (55 ppbv) layer, between 750 and 600 hPa. Backward trajectories (Figs. 5c and 5d) finishing near 700 hPa over Abidjan and Lagos in August identify two different origins for air masses in the southern hemisphere. The first origin (Fig. 5d) lies in the lower troposphere (850–800 hPa) west of Lake Tanganyika (Rwanda, Congo Democratic Republic) and is linked to the region of Lagos and Abidjan by the southeasterly trades flow (raising as it goes equatorward) emanating from the eastern flank of the St Helena Anticyclone. The second origin (Fig. 5c) is brought by the southwesterly monsoon flow from the Gulf of Guinea in the Atlantic, and supplied by the trade winds passing over others regions of fires (Gabon, Congo, 750 hPa). The biomass burning, which is highly active during this period south of the Equator (see, Sect. 3.2), could provide the ozone precursors needed to produce the ozone measured over Lagos and Abidjan, which is then advected by cross-equatorial south to north transport. Indeed, as we will see in Sect. 3.2, the shapes of the vertical profiles are similar between north and south, with higher ozone concentrations (85 ppbv) measured in the southern hemisphere at the same time. The decrease in the ozone concentrations during transport may be attributed to convective and turbulent mixing processes, as well as wet destruction in clouds. As cross-equatorial advection is very slow with a wind speed around 2 $m.s^{-1}$, more than 10 days of transport are necessary to go from the regions of fires up to Gulf of Guinea.

ACPD
4, 3285–3332, 2004

During the transition seasons, Spring (MAM) and Fall (SON), the ozone concentrations are lower. This clearly confirms the importance of the biomass burning in the African northern tropics for ozone production.

3.1.2. Middle troposphere

In contrast to the lower troposphere, the middle tropospheric (500 hPa and 250 hPa) seasonal cycles (Figs. 4b and 4c) do not show a maximum during the biomass burning season (December to February). Although the highest concentrations appear during December, with barely 60 ppbv, the monthly means for the rest of the year all range between 45 and 55 ppbv. The seasonal enhancement observed in the lower troposphere is not observed in the mid-troposphere. There is no increase of the background concentrations that remain near 50 ppbv. This can be explained by the subsidence caused by the Saharan Anticyclone found near 500 hPa between 5°–15° N, a feature which is present between November and February. Moreover, the stability of the Harmattan layers restricts ascending motions, and thus allows accumulation of the pollutant in the lower tropospheric layers and restricts ventilation into the middle and upper troposphere. From June to August, the middle and upper middle troposphere show ozone concentrations from 45 to 65 ppbv (Figs. 4b and 4c). The middle troposphere shows higher concentrations than the lower troposphere, as a consequence of the ozone-poor maritime boundary layer origin air of the monsoon flow below 600–700 hPa. During August, the concentrations are relatively constant at about 50 ppbv. Between 300 hPa and 200 hPa, Lagos has higher concentrations than during the dry season, with a +15 ppbv difference (Fig. 4c). A possible entrainment of air across the ITCZ could explain this difference (Thompson et al., 2002) as biomass burning is occurring within the Southern Hemisphere at this time of year. However, we also showed the role of south to north advection to increase the ozone background in the lower troposphere (near 700 hPa) during JJA (Sect. 3.1-A). ECMWF wind analyses show that the South Atlantic Subtropical Anticyclone (SASTA) centered at 15° S–20° E and 500 hPa has southeasterly flow that crosses the burning continental area of Central Africa and reaches the coast of the

Gulf of Guinea.

3.2. Central Africa region: Angola and Congo

Figure 3b shows the monthly mean ozone vertical profiles over Brazzaville and Luanda for the whole period of measurements. Figure 4 presents the seasonal cycles for layers centered on 750 hPa (Fig. 4a), 500 hPa (Fig. 4b) and 250 hPa (Fig. 4c).

3.2.1. Lower troposphere

During the southern hemisphere dry season, from June to August/September, there is an ozone enhancement in the lower troposphere over both Brazzaville and Luanda, between 900 to 600 hPa (Figs. 3b and 4a). The background ozone concentration is estimated at 50 ppbv, and the concentrations maximize at 700 hPa with 90 ppbv.

The back trajectories shown in Fig. 5e clearly demonstrate that the lower tropospheric air masses travel southeasterly from east of Lake Malawi. This is a region of intense fire activity (Fig. 6b), with the ozone precursors being injected into the continental southeasterlies as they pass over the fires of Democratic Republic of Congo, Zambia and Zimbabwe before moving up to Brazzaville and Luanda. This is also visible in the MOZAIC dynamical parameters (Fig. 8), with strong negatives zonal wind, u=−10 m.s^{-1}; v=+5 m.s^{-1}. Ozone deposition (suggested by a strong vertical ozone gradient of about 30 ppbv over 100 hPa is seen in Fig. 8) is at work in the layer below 900 hPa. In the lower troposphere (up to 900–850 hPa) Congo and Angola are subject during the entire year to the southern monsoon flux, with positive zonal and meridional winds (Fig. 8) of up to 3–4 m.s^{-1} on the monthly mean basis.

Figure 9a shows the vertical ozone profile over Lagos and Brazzaville in JJA. Up to 500 hPa the vertical profile over Lagos is similar in shape to that over Brazzaville, except that concentrations are lower and that characteristic points (ozone enhanced layers) are shifted vertically. Regional transport explains the similarities in the vertical profiles. Southeasterlies emanating from the SASTA anticyclone situated over Angola

ACPD
4, 3285–3332, 2004

reach the Gulf of Guinea coast, as the ITCZ peaks in its northern position (10–15° N) at this time of the year. The vertical displacement of the ozone enhanced layers between the two profiles is associated with an increase in altitude of the trade wind inversion level downstream of the anticyclone. During the regional transport in southeasterlies, deposition, convective and turbulent mixing processes participate to reduce ozone concentrations.

In the opposite season (DJF), Brazzaville shows an ozone enhancement, near 650 hPa, of 200 hPa thickness and with concentrations of 50 to 65 ppbv (Figs. 3b and 4a right). Such enriched ozone layers over Brazzaville during the boreal dry season were observed during the TRACE-A campaign (Nganga et al., 1996). Figure 5f shows that the trajectories ending in the lower tropospheric ozone maximum over Brazzaville have a northeasterly origin (over the Sudan) in the northern boreal hemisphere where biomass fires are active. Such transport by northeasterlies and easterlies emanating from the semi-permanent Saharan High affects both the Gulf of Guinea and Central Equatorial Africa (compare Figs. 5a and 5f) in DJF when the ITCZ peaks is in it southernmost position. This explains the similarities of the shapes of the vertical profiles over Lagos and Brazzaville during DJF (Fig. 9c). Note that the ozone maximum over Brazzaville is shifted vertically compared to the one over Lagos. This reflects the higher altitude of the trade wind inversion in the northeasterlies compared to easterlies. Further south, this feature is absent from the January and February vertical profiles over Luanda where ozone concentrations are quite low. The absence of such enhanced layers is explained by the lack of northeasterlies influence as seen in the ECWMF wind fields.

It is interesting to compare the dry seasons of Angola-Congo (in JJA) and Gulf of Guinea (in DJF). Figure 9b shows that they have the same lower tropospheric shape, but with ozone concentrations over Brazzaville being +15% higher with a thicker polluted layer (250 hPa against 150 hPa over Abidjan). Given that the magnitude of burning in northern and southern African tropical belts is very similar (48% of the African fires are in the north and 52% in the south, Hao and Liu, 1994), the different ozone con-

centrations may reflect the different types of vegetation found in the African northern and southern hemispheres. Figure 1 shows that in the southern hemisphere deciduous forest-woodland savanna predominates whereas brush-grass savannas become important in the north. According to Duncan et al. (2003) emissions factors of pollutant fires from tropical forest are higher for CO (104 g CO/kg DM), than fires from savannas (65 g CO/kg DM).

3.2.2. Middle troposphere

During JJA, middle tropospheric ozone is fairly homogenous (Figs. 4b and 4c), with concentrations of between 60–65 ppbv. The upper level, centered at 250 hPa shows lower concentrations of 45 to 60 ppbv (Fig. 4c right). These JJA concentrations are lower than those found in the lower troposphere. However the middle and upper troposphere, from 650 hPa up to 200 hPa, shows relatively strong seasonal concentrations, with a seasonal cycle enhancement in JJA, as observed in the lower levels (Fig. 4a), which is the opposite of that observed over the Gulf of Guinea (Figs. 4b and 4c left).

There is a large influence of the SASTA anticyclone, centered at 20° E–15° S, with strong easterlies and southeasterlies emanating from the eastern flank of the high (Hastenrath, 1990). As we can see in the back trajectories (Figs. 5g and 5h), easterlies and southeasterlies bring lower tropospheric air laden with ozone from the regions of fires (Zambia and Tanzania) up to Brazzaville and Luanda. The same winds influence the middle troposphere of Gulf of Guinea has seen previously (Part 3.1-B, Fig. 3a), with ozone loss during the transport over the Atlantic before it reaches the Gulf of Guinea coast.

During the intermediate season SON (Figs. 4b and 4c), there is an extension of the seasonal maximum that started in June, at both 500 hPa and 250 hPa. The vertical profiles (Fig. 3b) show two enhanced layers in September over Luanda, one centered at 450 hPa (+20 ppbv with respect to the background profile) and another at 250 hPa (+10 ppbv with respect to the background concentration of around 50 ppbv). During October and November, there is just one thick layer with ozone mixing ratio

up to 80 ppbv, centered near 350 hPa, and a large perturbation from the background (+20/+25 ppbv). Brazzaville shows the same behavior in October, with weaker ozone maximum of 70 ppbv.

In Fig. 5i, we see that strong easterlies dominate in September between the equator and 15° S. These winds give the rise to the advection of biomass burning ozone precursors from the eastern part of Africa (Malawi, Zimbabwe and Mozambique), after they have been transported vertically by convection into the middle troposphere over Brazzaville and Luanda. Indeed, at the beginning of September, biomass burning activity moves eastward, from Central Africa up to the east coast of Africa, and over Madagascar. A similar fire influence has been observed by Thompson et al. (1996) and Jenkins et al. (1997), during the TRACE-A and SAFARI 92 campaigns.

The other origin of the high ozone values during this season is the emissions of precursors from biomass burning in Brazil. This is not surprising as during this period (Hao et al., 1994), especially in the upper troposphere, northwesterly and westerly transport predominates. Convection lifts ozone and its precursors from the boundary layer over Brazil to the middle and the upper troposphere, where it is transported over the Atlantic by high-level westerlies, and with the easterlies emanating from the St-Helene high (SASTA) centered over Namibia. Figure 5j shows the path followed by pollutants in back trajectory calculations. This middle tropospheric enhancement during SON is in agreement with the analyses of Cros et al. (1992), Kirchhoff et al. (1996), Nganga et al. (1996), Thompson et al. (1996), Diab et al. (1996) and Olson et al. (1996) during TRACE-A and SAFARI 92 campaigns.

Another attributable origin of the high ozone concentrations measured over Angola and Congo near 250 hPa in SON is ozone production from lightning NO_x emissions. Observations from space by the LIS sensor (http://thunder.msfc.nasa.gov/data/query/distributions.html) show high levels of lightning activity over Angola and Congo in SON, as well as during DJF. The strong ozone enhanced layer appearing in DJF over Brazzaville (Fig. 9c) (which has higher concentrations than those observed over Lagos at the same time) could be explained by such an origin, as suggested by Jenkins et

ACPD
4, 3285–3332, 2004

al. (2004). The impact of lightning NO_x emissions on ozone production is badly quantified, as is the magnitude of the emissions. Estimates of the source magnitude are highly variable, from 2 Tg N yr^{-1} to 20 Tg N yr^{-1} (3–5 Tg N yr^{-1}, Levy et al., 1996, 5 (2–20) Tg N yr^{-1}, Lee et al., 1997, 12 (5–20) Tg N yr^{-1}, Price et al., 1997, 5 Tg N yr^{-1}, Jourdain and Hauglustaine, 2001, 6 Tg N yr^{-1}, Staudt et al., 2002a). The uncertainty of the global NOx lightning source and its impact on ozone concentrations remain one of the major issues in understanding the tropical ozone budget.

3.3. East Africa

Figure 3b shows the vertical profiles of ozone over Nairobi, and Fig. 4a the seasonal cycles for the three different airports: Entebbe, Kigali and Nairobi.

3.3.1. Lower troposphere

From May to November, the three sites have the same seasonal cycle, with maxima in July/August. However, the amplitude is highly variable from one place to another, even though the airports are situated East, West and North of Lake Victoria (Fig. 1). The lowest concentrations appear over Nairobi, with 30 ppbv in August, and the highest over Kigali, with 60 ppbv in July/August. In contrast to the northern region (Gulf of Guinea), there is a strong zonal gradient in ozone concentration.

The observed zonal gradient shows that the more continental the station, the higher the ozone concentration (Nairobi 30 ppbv, and Kigali 60 ppbv). This process is exacerbated at Nairobi that is enclosed between Indian Ocean and Victoria Lake. The oceanic origin of the southeasterlies, bringing clear air on contrary of Angola and Congo, explain why the ozone values are lower over East Africa than over Central Africa, where the continental origin is more pronounced.

Large north-south mountain ranges on either side of the Great Rift Valley and large inland lakes (Tanganyika and Victoria) may make the flow in this region far from zonal and may modify monsoon flow, resulting in this region begin dynamically more complex

than those discussed previously.

This increase in the seasonal cycle of lower tropospheric ozone is correlated with the local biomass burning, occurring over Rwanda, Tanzania, Mozambique, Zimbabwe and Kenya in JJA, along the southeastern coast of Africa. Figure 10 shows the stable (strong θ gradient) southeasterly flow (v=2.5 m.s^{-1}, u=−2.5 m.s^{-1}), characterized by high RH (80%). This flow, originating from the southwestern regions of the Indian Ocean in the area of the Mascarene anticyclone passes over the regions of fires, where it is loaded with ozone precursors, and then passes up to Kigali. Another important dynamical process participating in the transport of southeastern ozone precursors is the development of the East African low level jet, at 850 hPa, from May to September. This develops in association with the Indian and the East African monsoon, and has a southeasterly/southwesterly flow which penetrates inland (north westward) over eastern Africa through topographic breaks in the East African plateau (Findlater, 1974), before taking a north eastwards direction ending over the Indian Ocean and Arabian sea.

3.3.2. Middle troposphere

Figures 4b and 4c show the middle tropospheric seasonal cycle over East Africa. There is homogeneity of concentrations over the three sites, both at 500 hPa and 250 hPa.

Concentrations increase from May to September/October, at both levels, and are lower from November to January. The maximum occurs from August to September at 500 hPa, but continues until October at 250 hPa, with concentrations within the range of 50 to 55 ppbv. From June to November, concentrations increase with altitude over the three sites, with greater mixing ratios in the upper troposphere than in the lower.

The most noticeable difference in the dynamical situation between the lower and upper troposphere is a south-easterly flow in the lower troposphere, mixing continental polluted and oceanic clean air over the Lake Victoria region, as compared with a permanent easterly flow (u=−5/−10 m.s^{-1}, weaker θ gradient) (Fig. 10) in the middle and upper troposphere, with less humidity. As noticed by Thompson (2003b), this

origin confined between 40°–70° E, over the Indian Ocean, corresponds with a region that may be influenced by pollution from nearby East African regions (fires), or India (Lelieveld et al., 2001).

4. Comparison of MOZAIC and TOMS TTOCs

In order to compare our results to the previous climatology available over Equatorial Africa (i.e. from TOMS) we calculate the seasonal TTOC (Tropical Tropospheric Ozone Column) over Gulf of Guinea (average between the three visited airports) and Brazzaville. As the MOZAIC aircraft do not provide measurements above 200 hPa, we used seasonal mean values from the previous TRACE-A soundings between 200 and 100 hPa (considered as the tropopause level) for Brazzaville. For Abidjan, Lagos and Douala we used a constant concentration fixed at 70 ppbv between 200 and 100 hPa because of the absence of any strong seasonal cycle in the middle and upper troposphere.

The results in Fig. 11 show that the tropospheric column in DJF maximizes over the Northern Hemisphere (40 DU) and in JJA over the southern one (49 DU) due to the local biomass burning season. However, the inter-hemispheric difference during DJF and MAM is only 4 DU while it is 15 DU between JJA and SON. The lower tropospheric enhancement due to the northern hemisphere biomass burning season (DJF) is thus described in detail by the MOZAIC data, with 4 DU more than in the southern regions (Brazzaville). The ozone-enhanced layers between the ground and 700 hPa contribute to 40% of the total ozone column (16 DU) over Lagos and only to 30% (11 DU) over Brazzaville. The high ozone in the lowermost troposphere explains the problems encountered by satellites attempting to observe the northern tropospheric ozone column over Equatorial Africa. If we omit the lower troposphere (reflecting uncertainties regarding the effect of Rayleigh scattering) our columns between 700 hPa and 100 hPa would be 24 DU over Lagos and 25 DU over Brazzaville. This is obviously due to the low hemispheric difference in DJF, probably because of the north to south redistribution

ACPD
4, 3285–3332, 2004

of the pollutants, as explained in Sect. 3.2. Indeed, the huge fires occurring on the eastern part of the northern tropics influence both the Gulf of Guinea and the Brazzaville region. They also probably influence the South Atlantic Ocean as they are correlated to the spatial influence of the north-easterlies. Such a problem is not possible in JJA because the difference is big enough (15 DU) between Lagos and Brazzaville. This is probably one part of the explanation to the apparent "ozone paradox". Nonetheless, the MOZAIC data does give evidence to support the fact that the maximum of TTOC over Africa swings from West Africa in DJF to Central Africa in JJA. Finally, an interesting point raised in this study and highlighted with these TTOC over Gulf of Guinea and Brazzaville, is the higher ozone concentrations in the South (40.6 DU) compared to the North (36 DU), on annual mean basis.

5. Regional budget assessment

To further investigate the contribution of the northern tropics fires, we build a rough ozone budget for the region to assess the magnitude of the chemical ozone production in Africa in NH winter due to biomass burning.

The rectangular box chosen for the budget (Fig. 12a) extends vertically from the ground to 500 hPa. It is delimited to the North at 10° N, to the South at the latitude of Brazzaville, 4.3° S, to the West at the longitude of Abidjan, 4.0° W and to the East at the longitude of Nairobi, 36.7° E. Thus the box covers most of the Northern Hemisphere winter African biomass-burning area (see Fig. 6a). The fluxes across the western, southern and eastern faces are calculated based on the monthly averaged (January) profiles of wind and ozone for Abidjan, Brazzaville and Nairobi, respectively, integrated over the whole face (we assume a horizontally-uniform profile over the face). This hypothesis is partly supported by the monthly-averaged wind ECMWF analyses in January at 850 and 700 hPa (Figs. 12b and 12c).

Flux calculations across the different box faces were performed by vertical integration of the ozone mixing-ratio ($[O_3]$) and normal wind component (v) profiles in pressure

coordinate following: $\int_{P_{bot}}^{P_{top}} [O_3](P) . \frac{M_{O_3}}{M_{air}} . v(P) . L . dP/g$. This expression is derived as follows. $[O_3](P) . \frac{M_{O_3}}{M_{air}} . \rho_{air}(P) . v(P)$ represents the ozone mass flux crossing a surface unit normal to the wind component v (where M_{O_3} and M_{air} are the molar masses of ozone and air, and ρ_{air} the air density). By virtue of the hydrostatic equation, a surface element of height dz and width L (the width of the considered box face, namely 1590 km for the western and eastern faces and 4520 km for the southern face) has an area $L dz = L dP/\rho_{air}(P)g$ (where $g = 9.81\,m.s^{-2}$). For the numerical integration a finite pressure step of 50 hPa was taken for dP.

The wind-fields from the monthly ECMWF analyses prompt us to distinguish:

- on the western face (Abidjan), between the outflows due to the (north-easterlies) Harmattans below 750 hPa (term $F_{W\,HAR}$) and to the African Easterly Jet above (term $F_{W\,AEJ}$).
- on the southern face (Brazzaville), between the monsoon inflow below 900 hPa (term $F_{S\,MON}$) and the Harmattans outflow above (term $F_{S\,HAR}$).

The inflow through the eastern face (Nairobi) is referred to as F_E.

The box's northern face is assumed to have a zero flux (quasi-zonal wind in ECMWF analyses at all levels) except to the east in a 10°-wide longitude window (1110 km) where there is a significant inflow below 800 hPa due to a north northeasterly low-level jet (northerly component of about 5 m/s) descending from Egypt. The transported ozone mixing-ratio is estimated to 40 ppb (according to the MOZAIC seasonal mean profile over Cairo in DJF). The corresponding term is referred to as F_N.

The other terms of the budget are surface deposition SD, the flux across the top face F_{top} and the net photochemical production P. The magnitude of these terms is not known. However, as F_{top} is expected to be directed out of the box (dominated by convective transport), $P - F_{top} - SD$ provides a lower limit for ozone production by

biomass burning within the box.The budget can therefore be written as:

$$P - F_{top} - SD = F_{W\,HAR} + F_{W\,AEJ} - F_{S\,MON} + F_{S\,HAR} - F_E - F_N.$$

However, F_{top} can be considered negligible compared to the other two terms because of the absence of any seasonal cycle in the middle and upper troposphere (as seen in Sect. 3.1). The calculated values for these terms are shown in Table 3. The net ozone production and deposition term over the Northern Hemisphere Africa integrated over the whole biomass burning season (120 days from 15 November to 15 March) is thus estimated to be 61.9 Tg. This represents 14 to 22% of the global annual ozone photochemical production in the troposphere (estimated to be between 288 and 451 Tg according to recent global model calculations by Hauglustaine et al., 2004). The Harmattan and the AEJ appear to be the most efficient carriers of the produced ozone. The total westward transport ($F_{W\,HAR} + F_{W\,AEJ}$=46.3 Tg) is probably responsible of the high ozone concentrations recorded over the Atlantic Ocean and further west up to Brazil. This explanation to the "ozone paradox" is also further supported by inspection of the southward transport towards the southern Atlantic ($F_{S\,HAR}$=40.4 Tg) that is of the same order of magnitude as the transport by the Harmattan and AEJ.

6. Summary and conclusions

More than 800 flights over Equatorial Africa, recorded between April 1997 and March 2003, provide a new and unique view of the tropospheric ozone distribution over this previously poorly documented region. Regional and seasonal climatologies of ozone have been established for three different regions, and a detailed picture of the ozone vertical distribution is provided from in-situ data.

Analyses of geographical and seasonal ozone variations have shown that the lower tropospheric ozone enhancement results from biomass burning in both hemispheres. Over the Gulf of Guinea, an average maximum concentration of 80 ppbv of ozone is measured from mid-November to mid-March within a layer of about 150 hPa thickness.

ACPD
4, 3285–3332, 2004

Two mechanisms of ozone transport are identified. First, we suggest that ozone is initially and regionally formed in the Harmattans layer by rapid photochemistry processes over the band of fires. Second, the absence of any zonal gradient suggests a slower photochemical production or stronger mixing processes during the transport along the African Easterly Jet. Further south, over Central Africa (Congo and Angola), a similar enhancement is observed from June to August, with greater concentrations (up to 100 ppbv) within a thicker polluted air layer (about 250 hPa thickness). The greater concentrations over Brazzaville are probably due to greater ozone precursors emissions by the forests fires influencing the Brazzaville region in comparison to the savannas fires influencing Lagos and Abidjan as noted by Duncan et al. (2003). Over East Africa, the burning season also occurs during JJA. However, the monthly mean concentrations never exceed 60 ppbv over Kigali and are only 30 ppbv over Nairobi. This region is the only region where a zonal ozone gradient in the lower troposphere during the burning season was observed. This is probably due to the particular dynamical processes in the eastern part of Africa due to the mountain chain on one side and the Indian Ocean on the other side.

The Gulf of Guinea exhibits no seasonal variation above 600 hPa (monthly mean concentrations between 50 and 60 ppbv). The lower tropospheric enhancement recorded in DJF remains confined between 800 and 650 hPa. This characteristic feature is due to the dynamical and geographical situation over West Africa. Indeed, the polluted layers are enclosed below the trade wind inversion, capped with subsiding warming air from high altitude, provided by the anticyclone. The stability of the Harmattans and the presence of the AEJ on top of them, prevent any efficient vertical transport. In comparison, the middle troposphere over Central Africa presents a broad June–November ozone maximum. The first part of the maximum (JJA) corresponds to the lower tropospheric enhancement while the second part (SON) may be attributed to polluted air masses from Brazil and South Africa biomass burning events reflecting the trans-Atlantic transport, occurring at the same time as a re-circulation around the St. Helena anticyclone. An additional ozone formation at this time of the year may come from lightning.

ACPD
4, 3285–3332, 2004

The most interesting result of this study probably lies in the "coupling" between the regions of Gulf of Guinea and Brazzaville. The ozone vertical distribution over Brazzaville in DJF clearly shows the influence of the northern hemisphere fires on both regions. This corresponds to the spatial influence of the north-easterlies and further argues in favor of a rapid ozone formation over the eastern part of the northern tropics before being transported towards both Gulf of Guinea and Brazzaville. The TTOCs calculations reflects such a redistribution, with only 4 DU difference between Gulf of Guinea and Brazzaville in DJF, but with higher column ozone north of the equator than previously observed with satellites. During DJF Brazzaville the upper tropospheric maximum at 200 hPa is probably attributable to lighting or a mixing of biomass burning emissions from the northern hemisphere and lightning NOx emissions. In JJA, the "coupling" is reversed. Ozone over the Gulf of Guinea is mainly from the southern regions of fires through the monsoon flow.

Finally the northern African tropics play an important role in the regional and global ozone budget. Our first rough analysis shows that biomass burning in this region might contribute up to 20% of the global ozone photochemical production. Not only is the magnitude of this ozone production important but also its spatial distribution. African fires in DJF are a major source of ozone precursors and they also affect the southern hemisphere due to the particular dynamics over this part of the continent. These consequences have been called the "ozone paradox". The MOZAIC data presented here partly provide an explanation of this phenomenon. A study in preparation will further discuss this apparent "ozone paradox", along with the African maximum previously described as the zonal wave-one found by satellite data and confirmed by the SHADOZ network (Thompson et al., 2003b).

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Table 1. Major African campaigns with ozone measurements.

Acronym	Publication	Period / Place
TROPOZ I and II "**TROP**ospheric **O**Z**o**ne experiment"	J. Geophys. Res., 103, D15, 1998; Jonquieres et al., 1998.	11-22 December 1987 West Africa 9 January – 1 February 1991 West Africa
DECAFE « **D**ynamique **E**t **C**himie **A**tmospherique en **F**oret **E**quatoriale » FOS/DECAFE	J. Geophys. Res., 97, D6, 1992, special issue "FOS/DECAFE 91 Experiment" in J. Atmos. Chem, 22, 1, 2, 1-239, 1995	February 1988 Congo Dry season HN 1991 Ivory Coast
SAFARI-92/**TRACE-A** "Transport and Atmospheric Chemistry near the Equator-Atlantic"	J. Geophys. Res., 101, D19, 1996	South Africa 21 September 1992 to 26 October 1992
SAFARI-2000 "Southern African Regional Science Initiative"	South African Journal of Science, 98, 2002, special issue, J. Geophys. Res., 108 D13, 2003	March 1999 and March 2001 Dry season HS (6 August – 26 September 2000) Zambia (Lusaka)
Polarstern cruises	1987,1988; Smit et al., 1989; Weller et al., 1996	
Aerosols99 cruise	Thompson et al., 2000	January February 1999 Dry season HN Gulf Of Guinea (N.Y. → Cape Town)
SHADOZ "**S**outhern **H**emisphere **AD**ditional **Oz**onesondes"	Thompson et al., 2002	1998-2003 once every week Nairobi (over Africa)
EXPRESSO Part of DECAFE program		November 1996 to December 1996 Central Africa

Table 2. Number of MOZAIC observations per months over the Central African area, for all the airports served by the MOZAIC program, from April 1997 to May 2002.

AFRICA Monsoon Season / B.B. Season Town/Month	Jan.	Feb.	Mar.	Apr.	May	Jun.	Jul.	Aug.	Sep.	Oct.	Nov.	Dec.
Abidjan Ivory Coast, 5.4°N/4°W	32	12	12	18	6	12	16	14	8	26	18	6
Brazzaville Congo, 4.3°S/15.3°E	7	16	6	10	9	12	18	12	14	8	-	-
Dakar Senegal, 14.6°N/17.5°W	-	4	8	18	12	2	14	2	4	14	2	6
Douala Cameroon, 4°N/9.8°E	8	24	31	4	-	-	-	10	2	6	9	-
Entebbe Uganda, 0°N/32.4°E	2	-	-	-	8	4	8	12	6	7	2	-
Kigali Rwanda, 2°S/30.1°E	-	-	-	-	10	10	8	12	14	6	2	2
Lagos Nigeria, 6.6°N/3.3°E	14	28	20	55	12	2	14	16	4	24	20	10
Luanda Angola, 8.8°S/13.3°E	6	-	2	8	5	1	10	4	4	4	2	2
Nairobi Kenya, 1.1°S/36.7°E	2	-	-	-	18	14	16	24	20	14	4	2
Total	71	84	79	113	80	57	114	106	76	109	59	28

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4, 3285–3332, 2004

Table 3. Ozone fluxes and time-integrated masses for the ozone budget defined in Sect. 5.

Budget term	O_3 flux (kg/s)	120 days integration (Tg)
$F_{W\,HAR}$	1950 (out)	20.2
$F_{W\,AEJ}$	2500 (out)	25.9
$F_{S\,MON}$	153 (in)	1.59
$F_{S\,HAR}$	3900 (out)	40.4
F_E	1470 (in)	15.2
F_N	932 (in)	9.66
P - SD (- F_{top})	**+ 5975**	**+ 61.9**

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4, 3285–3332, 2004

Fig. 1. Positions of the different airports in the four different regions defined in the text, and the different associated vegetation type.

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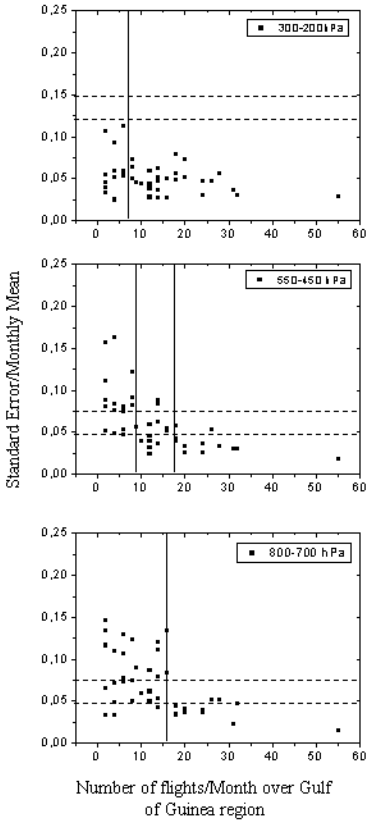

Fig. 2. Fractional standard error versus the number of flights in a month, for the pooled data and over Gulf of Guinea region. The horizontal dashed lines are for relative standard errors of 0.05 and 0.075 for the lower and the middle troposphere (800–700 and 550–450 hPa, respectively), and for 0.125 and 0.15 for the upper troposphere. Each point is the value for a given station and given month, for one of the pressure level indicated.

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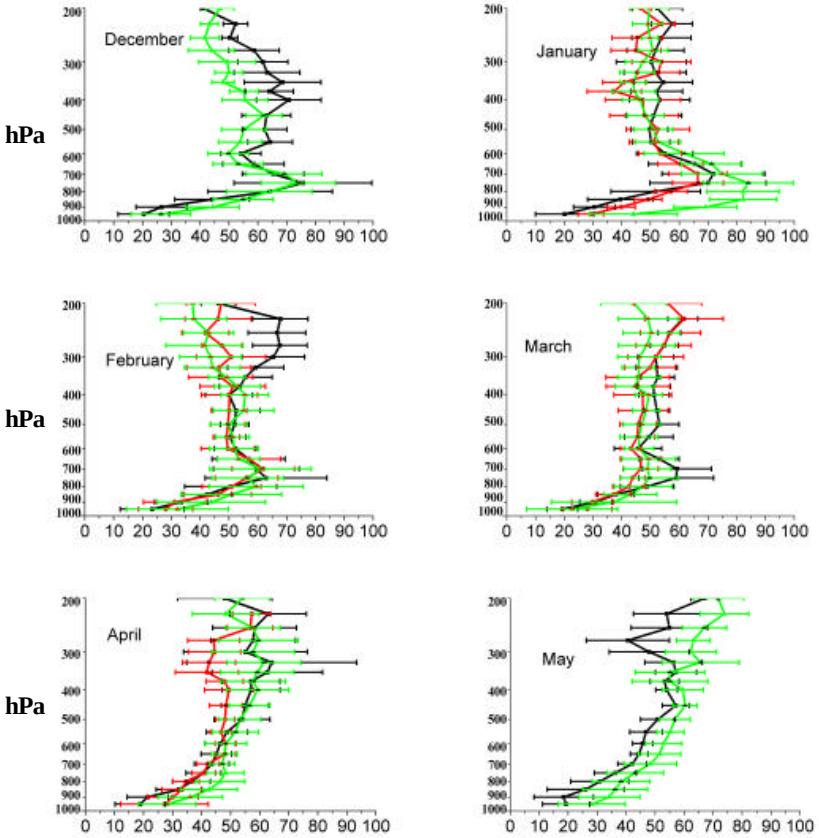

Fig. 3. (a)

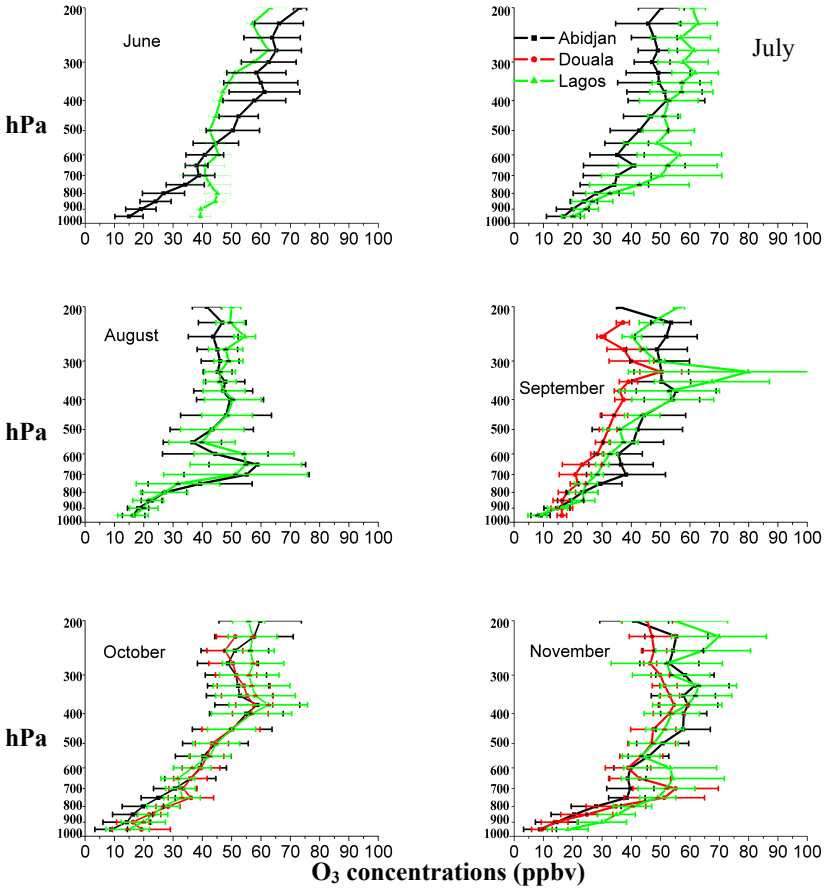

Fig. 3. Ozone mean vertical profiles in parts per billion by volume (ppbv) as a function of altitude, over the different areas: for the Gulf of Guinea in **(a)** with: Abidjan (black), Douala (red), Lagos (green). The horizontal bars represent one standard deviation about mean.

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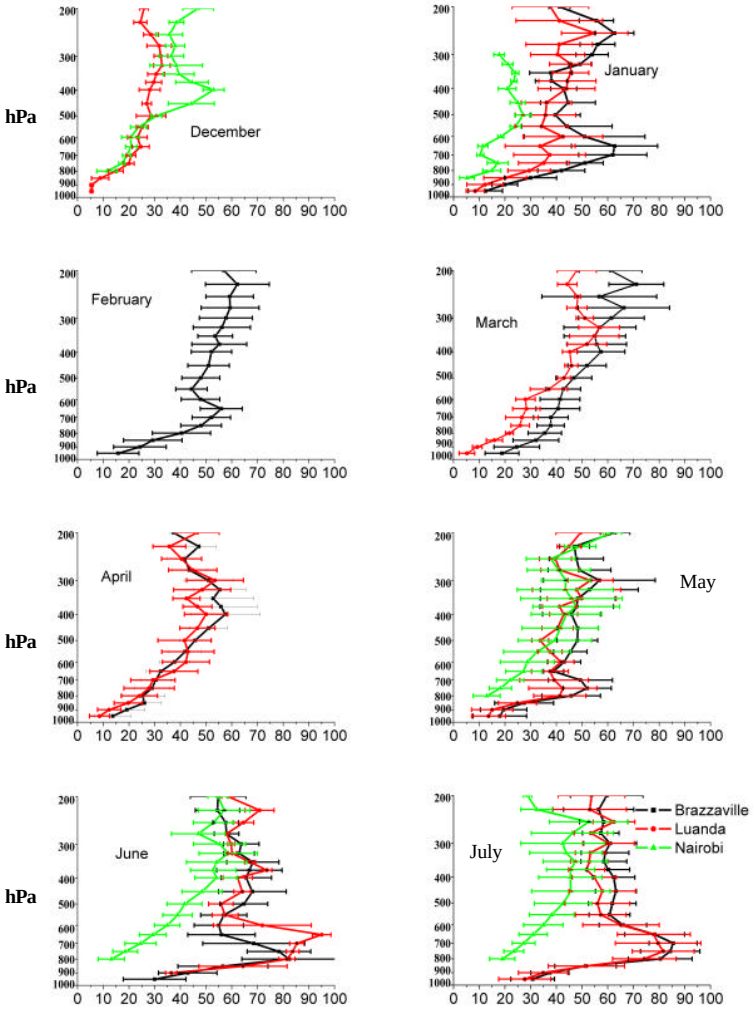

Fig. 3. (b)

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Tropospheric ozone over Equatorial Africa

B. Sauvage et al.

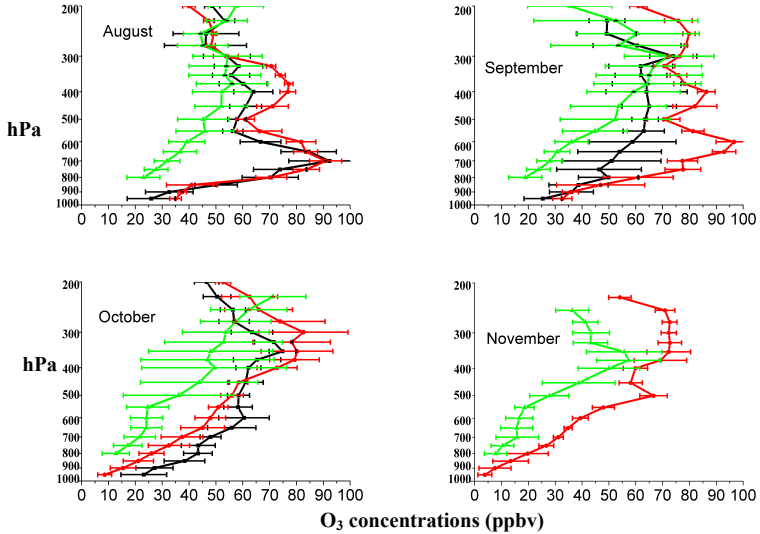

Fig. 3. Ozone mean vertical profiles in parts per billion by volume (ppbv) as a function of altitude (in hPa), over the different areas: for the Southern Hemisphere in **(b)** with: Brazzaville (black), Luanda (red) (Central Africa region), Nairobi (green). The horizontal bars represent one standard deviation about mean

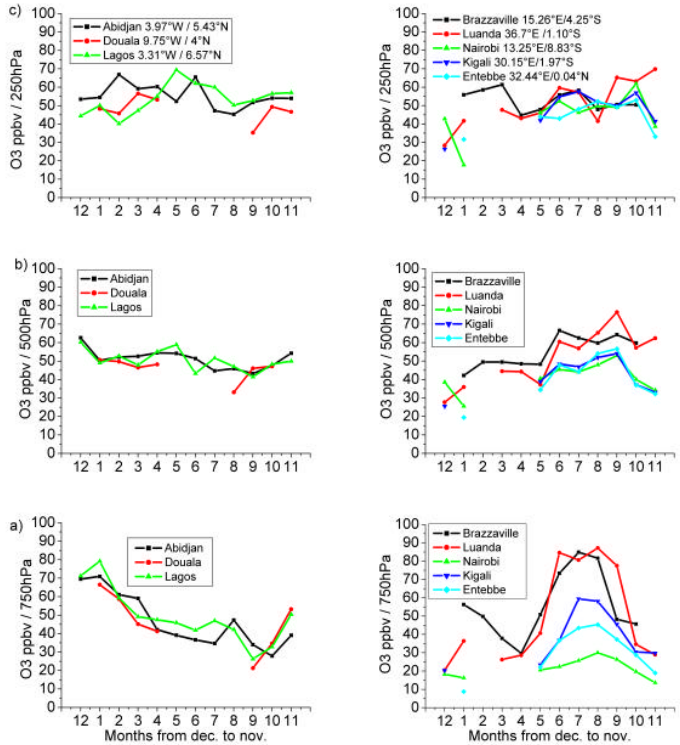

Fig. 4. Ozone seasonal cycle at 250 hPa **(c)**, 500 hPa **(b)** and 750 hPa **(a)** for Gulf of Guinea stations (left) Abidjan in red, Douala in red, and Lagos in green; and those within the Southern Hemisphere (right) Brazzaville in black, Luanda in red, Nairobi in green, Kigali in blue and Entebbe in Cyan. Monthly means are from December to November averaged over the entire period (April 1997–May 2003).

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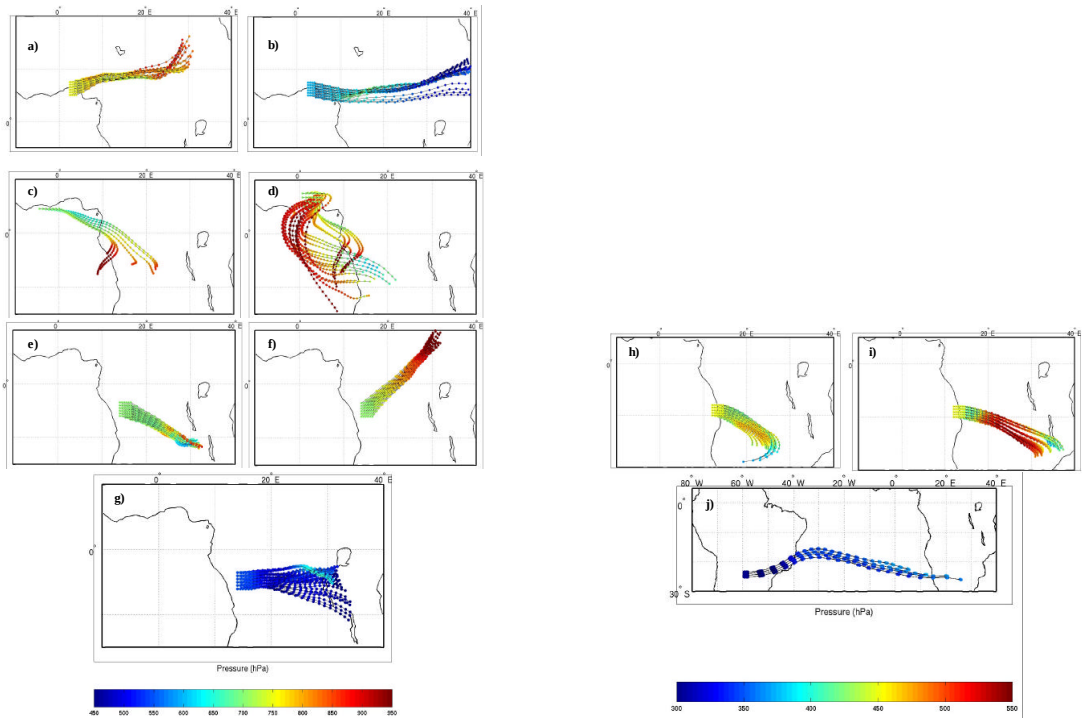

Fig. 5. 5 days back-trajectories computed with monthly ECMWF analyses and initialised over Lagos in January at 750 **(a)**, and at 600 hPa **(b)**. 10 days back-trajectories computed with monthly ECMWF analyses over Abidjan and Lagos region, in August at 700 hPa (**(c)** and **(d)**). Back trajectories initialized at 700 hPa over Brazzaville in August **(e)** and at 700 hPa in January **(f)**. 5 days back trajectories initialized in August over Brazzavile at 550 hPa **(g)**, and over Luanda at 450 hPa **(h)**. 5 days back-trajectories initialized with ECMWF monthly analyses in September over Luanda at 450 hPa **(i)**. Forward trajectories initialized at 300 hPa over Brazzaville **(j)**. Trajectories are color-coded with pressure (hPa), from 450 hPa to 950 hPa for (a), (b), (c), (d), (e), (f), and (g); and from 300 hPa to 550 hPa for **(h)**, **(i)**, and **(j)**.

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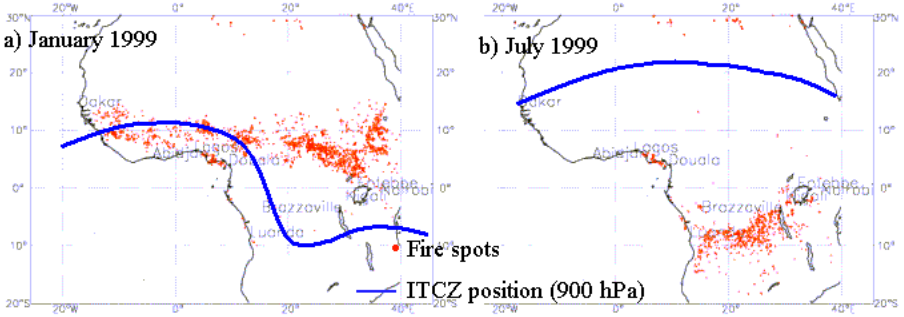

Fig. 6. Fires counts over West Africa in 1999 **(a)** January **(b)** July. The biomass burning season is between December and March in the Northern Hemisphere and from June to August for the southern Hemisphere. (1999 World ATSR Fire Atlas, O. Arino and J. M. Rosaz, ESA). The location of the MOZAIC airports is in grey. The location of the Inter Tropical Convergence Zone (ITCZ) at 900 hPa is in blue.

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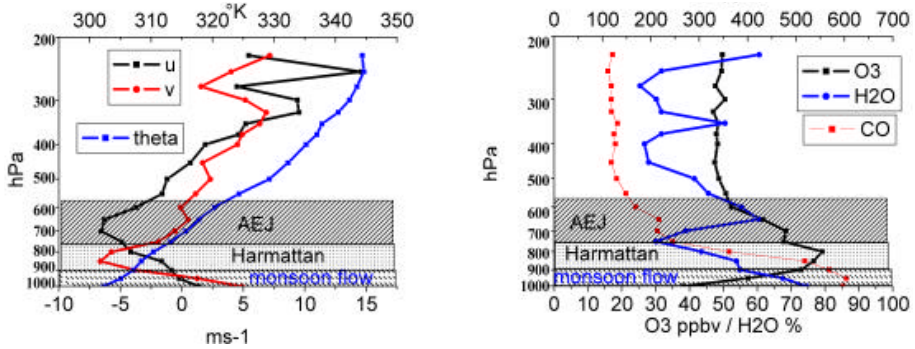

Fig. 7. Lagos monthly mean vertical profiles during Januray from the ground to 200 hPa. Dynamical parameters are on the left. In red meridional components of the wind v (m/s), black zonal components of the wind u (m/s), blue: potential temperature (°K), Chemical parameters on the right: in red: CO (ppbv), black: O_3 (ppbv), blue: RH (%). The three different layers described in the text of highlighted in grey.

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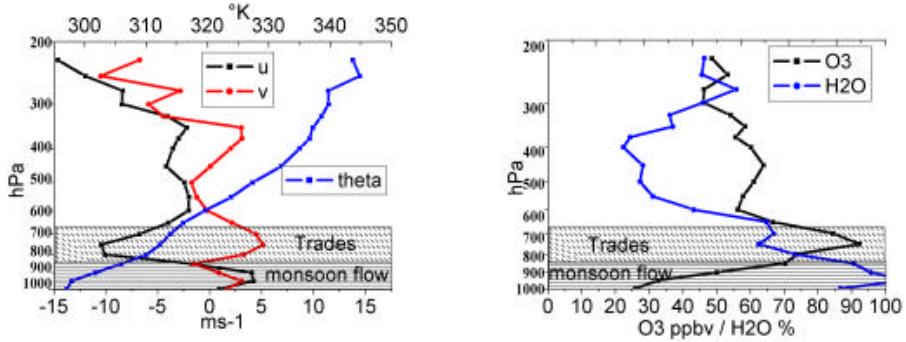

Fig. 8. Monthly mean vertical profiles of ozone over Brazzaville during August Dynamical parameters on the left: in red: *v* (m/s), black: *u* (m/s), blue: potential temperature (°K); Chemical parameters on the right: in red: CO (ppbv), black: O_3 (ppbv), blue: RH (%). Trades layer is in grey.

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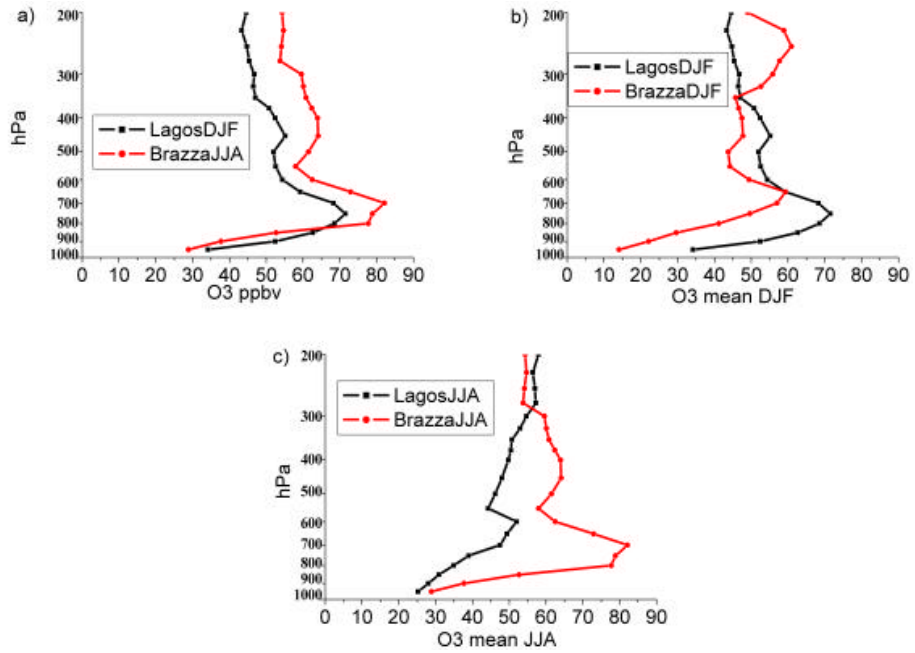

Fig. 9. Seasonal comparison between Lagos (black) and Brazzaville (red) during respective biomass burning season DJF and JJA **(a)**, DJF **(b)**, and JJA **(c)**.

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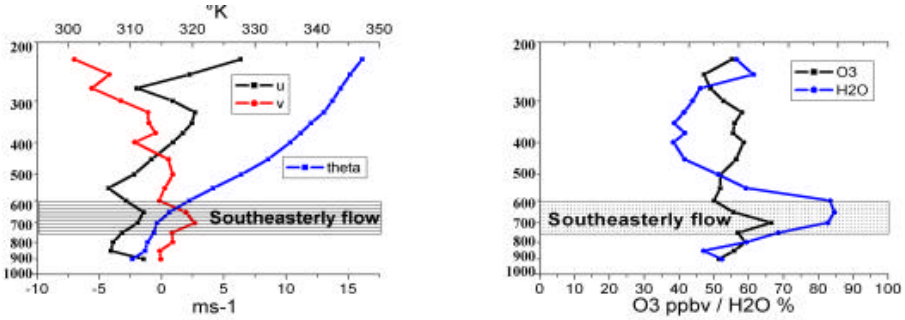

Fig. 10. Kigali vertical profiles monthly means (during August, from the ground to 200 hPa). Dynamical parameters on the left: in red: *v* (m/s), black: *u* (m/s), blue: potential temperature (∘K); Chemical parameters on the right: in black: O_3 (ppbv), blue: RH (%).

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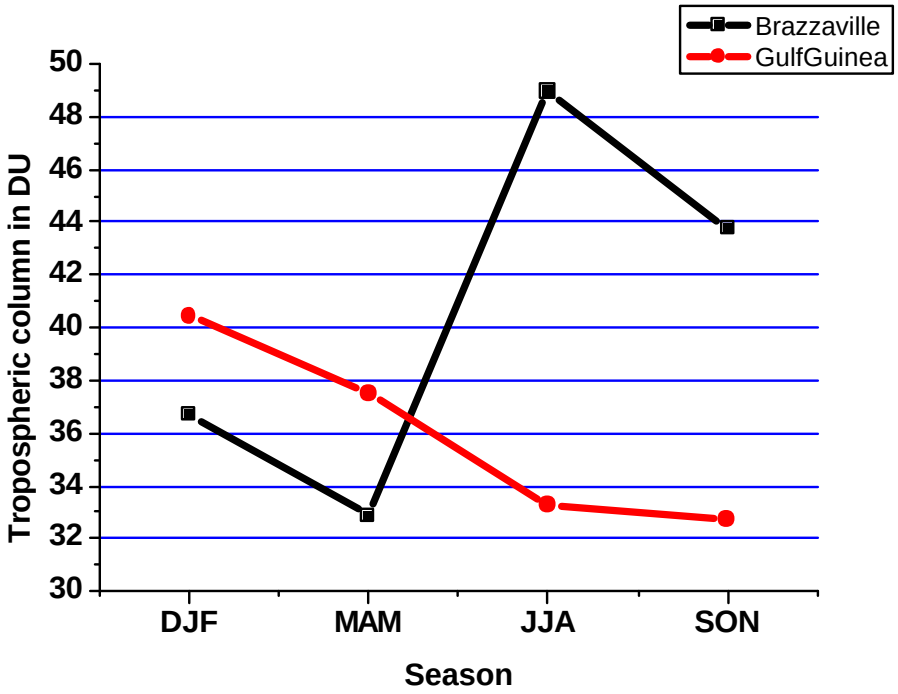

Fig. 11. Seasonal TTOC (DU) variation over Gulf of Guinea (red), and Brazzaville (black). To fill in the observations between 200 hPa TRACE-A soundings are used for Brazzaville whereas a constant O_3 concentration of 70 ppbv is used over the Gulf of Guinea.

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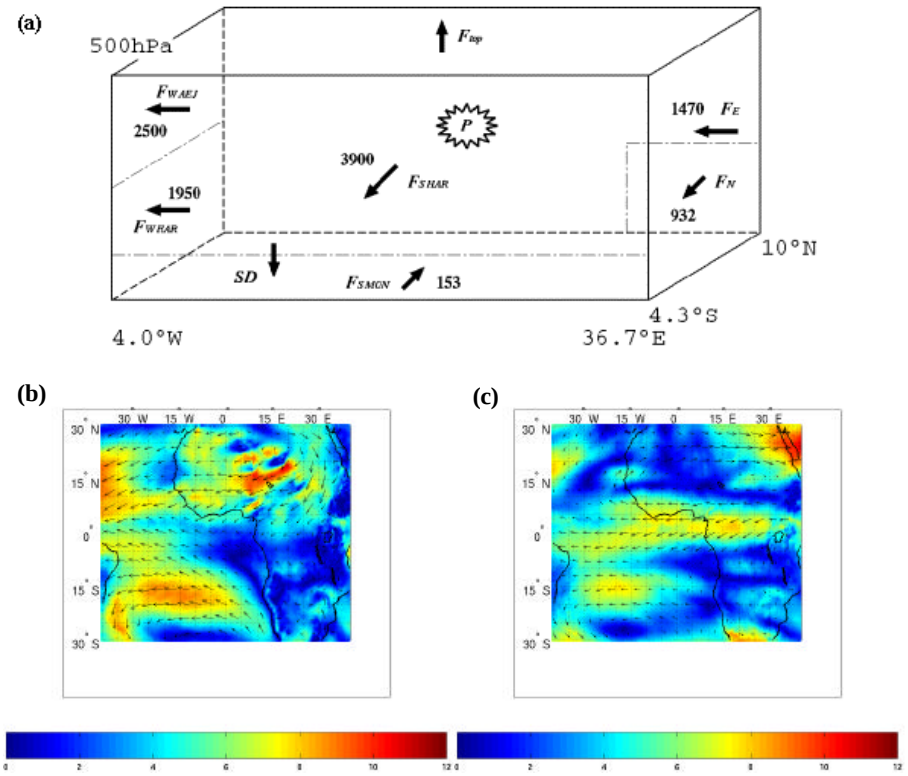

Fig. 12. **(a)** O_3 budget: box definition and considered flux terms with orientation and intensity (in kg/s). **(b)** ECMWF monthly wind analysis at 850 hPa for January 2000. **(c)** As in (a) but at 700 hPa. Arrows represent wind vector and the colour represents wind speed.

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