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# Two years of Ozone radio soundings over Cotonou as part of AMMA: overview

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### **ACPD**

9, 11221–11268, 2009

### Overview of Cotonou ozone soundings

V. Thouret et al.





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#### Abstract

As part of the African Monsoon Multidisciplinary Analysis (AMMA) program, a total of 98 ozone vertical profiles over Cotonou, Benin, have been measured during a 26 month period (December 2004-January 2007). These regular measurements broadly document the seasonal and inter annual variability of ozone in both the troposphere and the lower stratosphere over West Africa for the first time. This data set is complementary to the MOZAIC observations made from Lagos between 0 and 12 km during the period 1998–2004. Both data sets highlight the unique way in which West Africa is impacted by two biomass burning seasons: in December-February (dry season) due to burning in the Sahelian band and in June-August (wet season) due to burning in southern Africa. High inter annual variabilities between Cotonou and Lagos data sets and within each data set are observed and are found to be a major characteristic of this region. In particular, the dry and wet seasons are discussed in order to set the data of the Special Observing Periods (SOPs) into a climatological context. Compared to other dry and wet seasons, the dry and wet season campaigns took place in rather high ozoneenvironments. During the sampled wet seasons, southern intrusions of biomass burning were particularly frequent with concentrations up to 120 ppbv of ozone in the lower troposphere. An insight into the ozone distribution in the upper troposphere and the lower stratosphere (up to 26 km) is given. The first tropospheric columns of ozone based on in-situ data in this region are assessed. They compare well with satellite products on seasonal and inter annual time-scales, provided that the layer below 850 hPa where the remote instrument is less sensitive to ozone, is removed.

#### Introduction

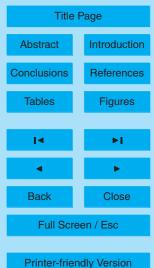
Tropospheric ozone is an important trace gas in particular via its role in the oxidative capacity of the global atmosphere and its climate effect. Two main reasons make the tropical regions of interest regarding tropospheric ozone. Firstly photochemistry

### **ACPD**

9, 11221–11268, 2009

### Overview of Cotonou ozone soundings

V. Thouret et al.





and OH formation are more active in the tropics due to high UV radiation and humidity. Secondly, the tropics are important source regions of ozone precursors especially through biomass burning (Andreae and Merlet, 2001; van der Werf et al., 2006), biogenic (Guenther et al., 1995; Serça et al., 1998; Jaeglé et al., 2004, 2005; Guenther et al., 2006; Aghedo et al., 2007) and lightning emissions (Moxim and Levy, 2000; Sauvage et al., 2007b; Höller et al., 2009).

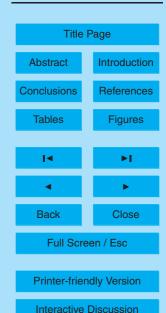
The previous African campaigns (from TROPOZ 1987 to SAFARI 2000) have effectively highlighted the importance of the tropical region, and particularly Africa, as a source region of ozone precursors (e.g., Cros et al., 1992, 2000; Delmas et al., 1999; Jonquière et al., 1998). Most of the data obtained during those campaigns come from low spatial or temporal sampling, meaning that regional and/or seasonal, annual or interannual variabilities are not well documented. More recently, the only data set providing regular ozone profiles over West Africa has come from the MOZAIC program (Marenco et al. (1998) and http://mozaic.aero.obs-mip.fr/web/). This program recorded ozone concentrations over Abidian, Ivory Coast and Lagos, Nigeria from 1998 to the beginning of 2004, with most of the data in 2000 and 2003. Thus the first climatology of tropospheric ozone over West Africa up to 200 hPa (12 km) was established by Sauvage et al. (2005). In particular, they showed that the ozone distribution over West Africa is influenced by two biomass burning seasons, in December to February (DJF) due to fires in the Sahelian area and in June to August (JJA) due to fires in the northern part of the Southern Hemisphere. Further details can also be found in the following studies: Sauvage et al. (2006, 2007b,c).

As part of the African Monsoon Multidisciplinary Analysis (AMMA) program, which aims in particular to document the chemical composition of the West African atmosphere and its variations linked to dynamics and climate, an ozone sounding station was set up in Cotonou, Benin (6.21° N, 2.23° E) (Fig. 1) thanks to the IRD (Institut de Recherche et Développement) and the DMN/ASECNA (Direction de la Météorologie Nationale/Agence de securité et de navigation aérienne) people located there. These regular soundings were performed during the Enhanced Observation Period (EOP)

#### **ACPD**

9, 11221–11268, 2009

### Overview of Cotonou ozone soundings



in 2005–2006 and aimed to better assess the seasonal and interannual variability of ozone vertical profiles in Equatorial Africa.

This new data set supplies complementary and additional knowledge regarding the ozone vertical distribution in the tropics and in particular in West Africa where ozonesondes were launched for the first time. Indeed, these profiles have also been archived in the SHADOZ (Southern Hemisphere Additional Ozonesondes; http://croc.gsfc.nasa.gov/shadoz/ and Thompson et al., 2003a,b) data base. The uses of such a data set range from ozone distribution studies (its main features and variations related to dynamics and climate), remote sensing satellite validation (in particular the tropospheric ozone column) to validation of chemistry-transport models used to interpret observations from various platforms (satellite, airborne, in-situ). The objectives of this paper are the following:

1. Present the entire data set from December 2004 to January 2007, including the evaluation against aircraft measurements during the campaign in summer 2006 and against the MOZAIC climatology (Sect. 2)

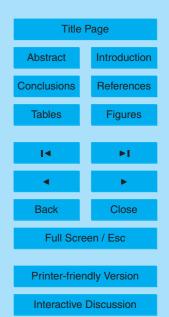
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- 2. Detail the measurements performed during the SOPs during the dry and wet seasons of 2006 (December–February and June–August, respectively) and set these data into a climatological context (Sect. 3).
- 3. Highlight new features identified in this data set. In particular, we will characterize the UT/LS (Upper Troposphere/Lower Stratosphere above 200 hPa) that has not been sampled previously (Sect. 4). In Sect. 5, we will give a first assessment of the tropospheric columns over this region based on the in-situ data.

### **ACPD**

9, 11221–11268, 2009

# Overview of Cotonou ozone soundings





### 2 Data presentation and evaluation in the lower and mid troposphere

### 2.1 Data acquisition

Regular weekly measurements were performed from December 2004 to January 2007. This frequency was doubled during the SOPs (Special Observing Period) in January 2006 and July–August 2006 to provide better statistics during these particular periods of the year (dry and wet seasons). A total of 98 profiles are now archived and available in the AMMA data base (http://database.amma-international.org/). Table 1 gives the number of profiles for each month. Due to serious technical problems in July 2005, end of January 2006 and December 2006, we were not able to perform as many soundings as scheduled (4, 8 and 4, respectively).

The ozone measurements were made with balloon-borne ECC (Electrochemical Concentration Cell) ozonesondes coupled with a standard radiosonde including a sensor for relative humidity and temperature. We used the Vaisala manufactured RS80 radiosondes during the entire AMMA period. The air for the ozone measurements was sampled with a Science Pump 6A type. Briefly, the principle of an ECC sensor is the following. A potential difference is set up between two cells (anode and cathode) with different strengths of KI (potassium iodide) solution (Komhyr, 1967). Then, the amount of ozone present in the sampled air is given by a formula including the current developed due to electrochemical reactions, the current relative to a zero-ozone background value, the temperature of the pump, the flow rate, and a correction factor accounting for the response time of the solution and the slowdown in the efficiency of the ozonesonde pump at high altitude and low temperature. All these terms are sources of uncertainties. It has long been recognized that the pump efficiency correction is the greatest source of uncertainty in the profile as a whole (Komhyr, 1986; DeBacker et al., 1998). Similar to the SHADOZ data (Thompson et al., 2003a), no correction factors from a co-located total ozone instrument have been applied to these AMMA soundings data. The overall uncertainties in the ozone soundings measurements have been evaluated to be in the range 4-12% for the ECC sondes (Barnes et al., 1985; Beekmann et al., 1994; Komhyr

#### **ACPD**

9, 11221–11268, 2009

# Overview of Cotonou ozone soundings

V. Thouret et al.





et al., 1995). More recently, Newchurch et al. (2003) have shown that electrochemical concentration cell sensors have an accuracy of about 10% in the troposphere, except when ozone is less than 10 ppbv when accuracies can be degraded to 15%. Note that such low ozone concentrations were very rarely recorded over Cotonou during the AMMA period. As part of the SHADOZ programme (tropical area), Thompson et al. (2003a) have shown an ozonesonde precision of 5% when compared to independent Dobson instruments. When compared to UV analysers onboard commercial aircrafts, Thouret et al. (1998) have shown that mean concentrations derived from ozonesondes are about 3 to 13% higher than those obtained by the MOZAIC program in the free troposphere in a similar geographic location.

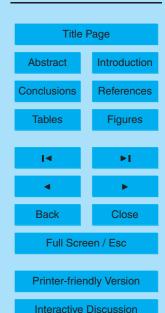
### 2.2 Comparison with aircraft data in August 2006

It is well recognized that ozone soundings are quite difficult to operate. Many sources of errors can be attributed to the sonde preparation 4 to 7 days in advance, to the storage of the sondes and electrochemical solutions and to the balloon preparation just before launching. These errors are particularly sensitive to tropical conditions due to high temperature and humidity. There is no easy way to check the data quality and consistency. Each ozonesonde launched is a new instrument. During the AMMA SOPs, ozone measurements were also made onboard research aircraft using UV photometers. Characteristics of the instruments (accuracies, detection limits) are summarized in Reeves et al. (2009). Unfortunately we did not have the opportunity to coordinate a sounding with the take off or landing from Cotonou airport of one of the aircraft measuring ozone. The comparison of the average vertical profiles of ozone from aircraft data around 6° N during the SOP in July-August 2006 and from ozonesondes from Cotonou over the same period is in general rather good (Reeves et al., 2009). However, the soundings data show higher ozone concentrations in the upper troposphere (between 550 and 150 hPa) due to the difference in measurement technique but also, in part, to the strategy of operations. Balloons were launched every 3 to 4 days regardless of meteorological conditions while some aircraft flights in the upper troposphere

### **ACPD**

9, 11221–11268, 2009

## Overview of Cotonou ozone soundings





were dedicated to the analyses of the mesoscale convective system outflows (explaining the relatively low ozone concentrations in the UT). More details can be found in Reeves et al. (2009). The July–August 2006 mean profiles measured from both the ozonesondes and aircraft show an ozone enhancement in the lower troposphere characteristic of the biomass burning plumes from the Southern Hemisphere as detailed in Sauvage et al. (2005, 2007a) and Mari et al. (2008). Reeves et al. (2009) show that the average for this period from the ozone radiosoundings exhibits lower concentrations in the lower troposphere and suggest that a greater amount of air sampled by the aircraft had been influenced by biomass burning.

A thorough comparison between soundings and aircraft data is made here for a shorter period (8-14 August 2006). This period is characterized by the so-called "extreme event" when the highest ozone concentrations (130 ppbv) relating to biomass burning intrusions were sampled in the lower troposphere. Figure 2 presents the mean vertical profiles between the surface and up to 6 km recorded by the soundings on the 10th and 14th, the BAe-146 aircraft on the 8th and 13th between 5.5-7°N and the D-F20 aircraft on the 13th around 4-5.5° N. They all show maxima from 80 to more than 130 ppbv between 3 and 5 km, well above the seasonal average (black line). The mean ozone profile from the BAe data (dark blue line) at these latitudes shows fairly good agreement with the ozonesonde data regarding the altitude and magnitude of the enhancement. There is also pretty good agreement between the two data sets in the lower troposphere. On 13/08 the D-F20 (green line) sampled a biomass burning pollution plume located below 5 km with highly enhanced concentrations of ozone and ozone precursors (Andrès-Hernández et al., 2009). The RS ozone profile measured on 14/08 (grey line) also shows this layer of enhanced ozone with concentrations in good agreement with the D-F20 data. The ozone profile from the D-F20 data shows a higher maximum and at a lower altitude probably because it has been sampled further south and further west. However, the top of the enhanced ozone layer is very similar to that observed from soundings.

### **ACPD**

9, 11221-11268, 2009

## Overview of Cotonou ozone soundings

V. Thouret et al.





### 2.3 Comparison with the MOZAIC climatology

An objective of the EOP was for this new data set to be a reference for the ozone distribution throughout the troposphere and the lower stratosphere. Therefore, it is necessary to assess how well this AMMA data set agrees with the MOZAIC climatology over Lagos at least for the lower and mid-troposphere as presented by Sauvage et al. (2005). Lagos is situated 110 km east of Cotonou, also by the Gulf of Guinea coast (Fig. 1) and we expect similar seasonal characteristics at least above the boundary layer. Equipped MOZAIC aircraft started collecting data en-route to Lagos in 1998, but unfortunately stopped their regular operation there in March 2004 before the start of the AMMA soundings (December 2004). No direct comparison is therefore possible to evaluate any bias between the two measurement techniques.

Figure 3 presents the monthly mean vertical profiles of ozone over Lagos for the entire period of operation from MOZAIC and over Cotonou for the 2 years of the EOP 2005 and 2006 (both averaged every 100 m). AMMA data for December 2004 and January 2007 are plotted as well. Both data sets show similar seasonal variations. In particular, both are marked with ozone enhancements around 2-3 km (Harmattan layer) during the dry season (December-January-February) and around 3-4 km during the wet season (June-July-August) due to fires in the Sahelian region and in the northern part of the Southern Hemisphere, respectively. However, it seems that the soundings data show higher concentrations than the MOZAIC data, especially in the middle and upper troposphere. This is particularly noticeable in JJA as well as in October, and in DJF. Indeed, Thouret et al. (1998) have shown that there can be up to 13% difference between the ozonesondes and MOZAIC data. However, the overestimation does not seem to be global or systematic. In April and May, soundings and aircraft data show only small differences. September and November 2006 exhibit profiles close to the MOZAIC average whereas large differences relative to the MOZAIC climatology appear in the mid and upper troposphere in 2005. Conversely, the January profiles from soundings are similar to each other and high compared to MOZAIC. In December, all

### **ACPD**

9, 11221–11268, 2009

### Overview of Cotonou ozone soundings





four profiles show very different ozone levels above 6 km.

To further quantify this variability, Fig. 4 gives the difference (%) between MOZAIC (taken as the reference) and the sonde data for the layer 2-8 km and for the 3 years of sampling. This is the layer where the comparison is probably the most relevant. The local surface effects (different urban pollution and time of sampling) do not have a significant influence and the locations of the different measurements are in the range of one to two hundred km. In general, the highest differences relatively to MOZAIC are observed in JJA and DJF. In JJA, it is worth noting how similar the sounding profiles are while the MOZAIC distributions were more variable (Figs. 3 and 9 which is detailed in Sect. 3.2 and presents MOZAIC and AMMA monthly means for each sampled wet season). However, we notice that in December, the differences range from 7 to 60% as also seen in Figure 6 (which is detailed in Sect. 3.1 and presents MOZAIC and AMMA monthly means for each sampled dry season). The difference is noticeable throughout the troposphere. For the annual average, the difference is 20% (sondes higher than MOZAIC) which is higher than the range of uncertainties of the measurements. As the differences show a seasonal cycle similar to that of ozone and as there is no overlap in the sampling periods, the strong interannual variabilities of the processes leading to the ozone enhanced layers in DJF and JJA (biomass burning emissions and strength of the transport pathways) may explain most of this 20%.

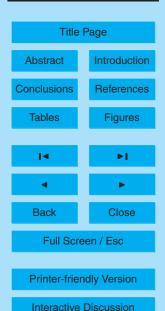
#### 3 Measurements during the special observation periods

One of the objectives of the EOP measurements was to give a more general and regional context for the data from the SOPs. The following sub-sections focus on the dry and the wet seasons specifically.

#### **ACPD**

9, 11221–11268, 2009

## Overview of Cotonou ozone soundings





### 3.1 Climatological context for the dry season DJF 2005–2006

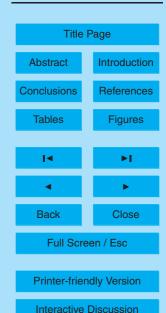
The first SOP, SOP-0, took place in January 2006 (Haywood et al., 2008). As it concerned the dry season it mainly focused on the measurement of aerosols originating from Saharan dust and biomass burning. Details concerning the measurements over Djougou, Benin are given in Mallet et al. (2008) and in Raut and Chazette (2008) for those recorded over Niamey, Niger. Individual soundings measured during the dry season 2005-2006 are presented in Fig. 5 as a time series. The highest values recorded during the dry season 2005-2006 were on 20 and 22 December in the lower troposphere as well for the entire tropospheric column. In addition to the very high ozone concentrations measured above 2 km, extreme ozone values were recorded around 1 km on December the 20th (Fig. 5) (details are given in Minga et al., 2009). Biomass burning signatures are clearly observed in each profile in January in the lower troposphere (3-4 km). High ozone concentrations were also measured in the upper part (8-10 km) of the troposphere in January 2006. These ozone enhanced layers probably originate from biomass burning products lifted by convection in a similar way to that described by Mari et al. (2008) during the wet season. Although these processes are similar, they are symmetrical in that they occur in opposing hemispheres and seasons. Pollutants emitted by fires in the Southern (Northern) Hemisphere during the wet (dry) season are trapped over the continent and advected to the north (south) by the trade winds (Harmattan flow) where they reach convective regions located further north (south) of the fire regions and are injected in the upper troposphere. A further study should be dedicated to better analyse these signatures and to draw symmetrical schemes of the processes involved during the dry and the wet seasons, respectively. In February, the profiles are more homogeneous vertically with lower values in the lower troposphere.

Figure 6b gives the monthly mean ozone vertical profiles over Cotonou recorded in January 2006 (black line) compared to the ones recorded in 2005 and 2007 along with the MOZAIC measurements taken over Lagos, in 2002, 2003 and 2004. Figure 6a and

### **ACPD**

9, 11221–11268, 2009

## Overview of Cotonou ozone soundings





6c present the monthly mean profiles in December and February as well. Three dry seasons 2004-2005 (red), 2005-2006 (black), 2006-2007 (pink) have been sampled during the AMMA campaign. In January, average concentrations in the Harmattan layer (1-3 km) almost reach 80 ppbv. It is quite similar to that observed over Lagos 5 in 2002, 2003 and 2004. However, it is worth noting that MOZAIC aircraft measured more ozone in the lower part of the Harmattan layer in 2002 than the soundings in 2006. Concentrations over Lagos reach 84 ppb at 2.2 km and are greater than 70 ppb between 1 and 3.6 km altitude this month. On the other hand soundings in 2005 and 2007 show average concentrations as high as 100 ppb at 2 km altitude. Within the AMMA data set, overall 2006 shows the highest values above 4 km. This is consistent with December 2005 showing very high ozone concentrations from the ground to the lower stratosphere compared to 2004 and 2006 (Fig. 6a). Indeed, individual soundings from the beginning of January (6, 9, 12) show higher concentrations than those from the end of the month (16, 19) especially in the upper troposphere (Fig. 5). In general, this dry season DJF 2005-2006 presents higher ozone concentrations which are in agreement with higher concentrations of black carbon from biomass burning in 2005 compared to 2006 (Liousse, personal communication).

### 3.2 Climatological context for the wet season JJA 2006

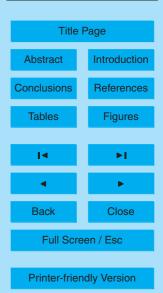
The third SOP, SOP-2, took place in July and August 2006 with various chemical objectives (Janicot et al., 2008) and a large aircraft operation between Niamey, Niger and the Gulf of Guinea. The chemical characterization of the troposphere over West Africa during this period is widely described by Reeves et al. (2009). Other aircraft data regarding the influence of convection on the chemical composition are presented by Ancellet et al. (2009) and Andrès-Hernández et al. (2009). Figure 7a presents the 24 vertical profiles recorded in JJA 2006. In contrast to the dry season DJF, the vertical gradient is more pronounced. In addition, this period is characterized by ozone enhanced layers between 3 and 5 km attributed to southern biomass burning products. This behaviour has been firstly reported by Sauvage et al. (2005, 2007a). Profiles with

### **ACPD**

9, 11221–11268, 2009

## Overview of Cotonou ozone soundings

V. Thouret et al.





such ozone layers correspond to the dates circled. We notice that these southern intrusions were rare in June (only the 30/06) and July (2 on 8) whilst they were nearly a daily occurrence during August 2006. Ozone concentrations in the upper troposphere are higher in June and in the beginning of July compared to August. It is also worth noting that the SOP-2 (20 July-21 August) is characterized by an "extreme event" recorded by the ozone radiosoundings on the 10th and 14th August and by aircraft (BAe and Falcon DLR) on the 8th and 13th of this month (Fig. 2). Concentrations up to 120 ppb were measured on the 14th around 4 km altitude. Figure 8a and b have been designed to highlight the intra seasonal variability and the relationship between the ozone concentrations and the meteorological conditions in the layer 600–800 hPa. For each sounding in JJA 2006, the ozone concentrations and the zonal wind speed have been averaged between 600 and 800 hPa (Fig. 8.a). The ozone background value at this pressure is around 40-50 ppbv. For days with biomass burning signatures, the magnitude of the ozone enhancement is represented with vertical bars. Days without intrusions generally show strong easterly zonal wind  $(<-5 \,\mathrm{m\,s}^{-1})$  attributed to the African Easterly Jet (AEJ-North) while days with intrusions are characterized by weak zonal wind speed, the AEJ-N being further north. A good agreement is found between in-situ observations and NCEP reanalysis zonal wind speed, especially regarding temporal variations during the JJA period. When Cotonou is under the influence of the AEJ-N, the jet prevents biomass burning intrusions going further north ("blocking effect"). Mari et al. (2008) have already suggested that the intra seasonal variability of the biomass burning intrusions is related to the activity of the AEJ-S (located at ~5° S, latitude of the fires regions). Indeed, most of days with intrusions have been preceded by days characterized by strong easterly winds at 2.5-5° S (AEJ-S). This jet allows an efficient transport of pollutants over the ocean and the Gulf of Guinea. The same analysis has been done for JJA 2005 with 5 out of 8 profiles showing biomass burning signatures (Fig. 8b). The zonal wind is much more variable during the 2005 wet season and the AEJ-S is not well pronounced for long periods (as in August 2006 for instance). As for 2006, days with intrusions show weak zonal wind. However, it is difficult to see if all these days

#### **ACPD**

9, 11221-11268, 2009

### Overview of Cotonou ozone soundings

V. Thouret et al.





were preceded by a strong transport by the AEJ-S over the ocean. These two figures highlight the strong intra-seasonal and interannual variability regarding wind patterns and ozone concentrations in this region. They suggest that intrusions are favoured by an AEJ-N located north of Cotonou and possibly by an active AEJ-S over the fires region in the Southern Hemisphere. However, other processes are still to be investigated to completely explain these southern intrusions. At the monthly mean scale, different dynamical situations in August 2005 and 2006 lead to similar ozone vertical profiles as shown below.

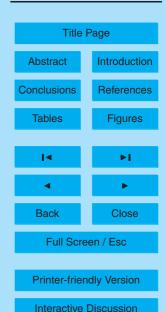
Figure 9 presents on overview of the interannual variability of the JJA profiles as seen in the AMMA program and in the MOZAIC measurements over Lagos. In August, it is worth noting how similar the two mean profiles from soundings are while the MOZAIC profiles in 2000 and 2003 show the two distinct behaviours (with and without the southern intrusions, respectively). Cotonou data exhibit thicker ozone layers with higher concentrations compared to the aircraft data over Lagos. The years 2005 and 2006 seem to have had more favourable conditions for these southern intrusions. Indeed, within the MOZAIC data set, 28% of the profiles in JJA 2003 presented such layers while the soundings time series (2005 and 2006) exhibited this phenomenon for 41% of the samples. The monthly mean for 2006 in July is closer to the one observed over Lagos in 2000, at least up to 5 km altitude. The mean vertical profiles in June do not present a well marked layer in general. Little signature of the southern intrusion is seen in 2005 and to a less extent in 2006 (only one profile presents such layers out of the 9 recorded).

Similar to the dry season, the AMMA SOP-2 took place in a rather "high ozone" environment with frequent enhanced layers originating from the Southern Hemisphere. The SOP-2 has also been characterized by the "extreme event" on 14 August 2006 when up to 120 ppbv were measured in the lower troposphere, a value similar to the highest measured during the dry season.

### **ACPD**

9, 11221–11268, 2009

## Overview of Cotonou ozone soundings





### 4 Upper troposphere/lower stratosphere

MOZAIC vertical profiles are limited to 12 km, the aircraft cruise altitude, which is well below the tropical tropopause. The soundings have the capability of sampling ozone throughout the troposphere and the lower stratosphere (balloons have reached altitudes of 26 km). Consequently they have provided the first data regarding the ozone distribution in the UTLS over Equatorial Africa. The aim of this section is to present the seasonal cycles of ozone and temperature in the UTLS.

Figure 10 shows the monthly mean time series of ozone concentrations averaged every 100 m from the ground to 17 km altitude between December 2004 and January 2007. The UT over Cotonou presents ozone concentrations higher than expected from the interpolation presented in Thompson et al. (2003b) and from the study by Sauvage et al. (2006) when they assumed the values of 70 ppb recorded at 12 km to be valid up to the tropopause. This AMMA data set shows that ozone concentrations between 12 and 17 km are closer to 90–110 ppbv on average, and up to 150 ppbv in August and September.

For further details of this UTLS region and for clarity of colour scale, Fig. 11a and b give the monthly mean ozone and temperature distributions between 12 and 22 km averaged every 100 m. The white line on the figures gives an indication of the tropopause altitude. This has been defined as the lowest altitude where the temperature vertical gradient reaches the value of  $-2 \, \text{K/km}$ . As expected, the tropopause altitude is around 17 km (16.9 $\pm$ 0.4). However, from June to October it is clear that the tropopause is a little bit lower and warmer than in the period from December to May. The difference is up to 10 K, in agreement with Reed and Vicek (1969) and Reid and Gage (1981). This behaviour is visible in both 2005 and 2006. The ozone seasonal cycle seems to be in phase with the well-known annual cycle of temperature. For better clarity we have averaged over the two years of sampling and we present the mean seasonal cycle of ozone concentrations and temperature at 12–14, 14–16, 16–17 and 17–18 km altitude in Fig. 12. It is worth noting that the lowest layer shows no significant seasonal vari-

### **ACPD**

9, 11221–11268, 2009

## Overview of Cotonou ozone soundings





ation (70–80 ppb throughout the year) while a broad summer maximum of ozone is observed in the three upper layers. Nevertheless, the amplitude below 16 km is very weak (less than 20 ppb or 20%) while it increases to 50 ppb at 16–17 km and up to around 150 ppb or 100% in the layer just above the tropopause, at 17–18 km. Concentrations between 17 and 18 km above Cotonou are in the range of 140–290 ppb, in agreement with that shown in Randel et al. (2007) based on a study of the SHADOZ profiles not including Cotonou. They linked this large annual cycle in ozone above the tropical tropopause to the Brewer-Dobson circulation. The amplitude of the temperature seasonal cycle is also higher above 17 km (up to 10 K) compared to the tropospheric layers (less than 3 K) and in phase with the ozone cycle, as expected. Ozone concentrations at 16–17 km are attributed to the upper troposphere and its seasonal cycle is very similar to the one just above the tropopause (17–18 km), but with a lower amplitude though. A similar pattern is observed for the temperature cycle. This may reflect a non-negligible influence of the stratosphere on the ozone distribution in the UT over Cotonou during the broad summer period (June to October).

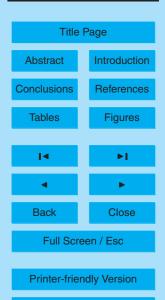
This ozone data set is of particular interest for further investigation of the different origins of air masses recorded in the TTL (Tropical Tropopause Layer, between 150 hPa or 14.5 km and 70 hPa or 17 km as defined by Fueglistaler et al. (2009) and references therein) over this location throughout the year. Indeed, Barret et al. (2008) have shown the influence of the Indian Monsoon and the Tropical Easterly Jet on the composition of the TTL over Northern Africa. Moreover, Law et al. (2009) systematically analyzed the high altitude aircraft data recorded during the SOPs in JJA 2006. They show that the region is mainly under the influence of African convection around 200 hPa (12.5 km), and of the Indian Monsoon around 150 hPa (14.5 km) while air masses sampled around 100 hPa (17 km) are related to stratospheric origins.

### **ACPD**

9, 11221-11268, 2009

### Overview of Cotonou ozone soundings

V. Thouret et al.





#### 5 Tropospheric columns of ozone

This present data set is actually the first one that allows a thorough quantification of tropospheric columns of ozone (TCO) over Equatorial Africa. The objectives of this section are to present the seasonal variations of these TCO and to compare them with satellite retrievals. Comparisons are based on monthly means to evaluate the seasonal and interannual variations of the columns. The goal is not a thorough comparison to the satellite data on a daily basis. Given that the satellite data are the only available records representative of monthly mean columns over this region, we aim to check whether our data set matches them despite its low sampling frequency. Meanwhile, we specifically discuss the sensitivity of remote instruments in the lower troposphere.

#### 5.1 Seasonal variations

Figure 13 and Table 2 present the annual variations of monthly mean TCO derived from the soundings and calculated in Dobson Unit (DU, 1 DU=2.69×10<sup>16</sup> molecules cm<sup>-2</sup>). The TCO range from 32.5 to 67.2 DU, with high annual and interannual variabilities (up to 20 DU). March, September and December show the strongest interannual variability. To better assess which atmospheric layer contributes to these variabilities, the TCO were divided into 5 layers, related to air masses origin. The first layer from 1000 to 900 hPa is under monsoon flux influence, the air masses coming from S-SW. The second layer (900-700 hPa) roughly corresponds to the Harmattan layer during boreal winter (DJF) and to trade winds during boreal summer (JJA). The third one from 700 to 500 hPa generally relates to the African Easterly Jet altitude, with large scale transport influence. The fourth and fifth ones represent for mid-upper and upper tropospheric layers which can be both effected by large scale transport and deep convection. The fifth one is also influenced by the lower stratosphere as seen in Sect. 4. The tropospheric ozone columns show little difference between 2005 and 2006 (2-3 DU) except for the beginning of the dry season (November and December) and the months of March and September (Fig. 13).

#### **ACPD**

9, 11221-11268, 2009

## Overview of Cotonou ozone soundings

V. Thouret et al.





The seasonal variations appear to be largely driven by the first three layers partly due to surface emission changes between dry and wet seasons. A maximum of partial TCO is clearly seen during the dry season (DJF) when emissions from biomass burning maximize with around 15 DU from the surface up to 700 hPa whilst rarely exceeding 10 DU the rest of the year. The mean ozone column amount averaged over the DJF season show 48 DU over Cotonou which is higher than that found over Lagos (41 DU) by Sauvage et al. (2006) using MOZAIC data. The difference in total ozone amount can be partly attributed to the climatological values of 70 ppbv between 186 and 100 hPa used by Sauvage et al. (2006) to augment MOZAIC vertical profiles. Figures 11a and 12 have shown that ozone values over Cotonou can reach up to 130 ppbv near the tropopause, higher than the assumed value used in their study. Nevertheless interannual and geographical variabilities might be part of the difference too. The ozone column below 700 hPa contributes to 14.9 DU of the TOC in DJF which is in fairly good agreement with the Sauvage et al. (2006) results over Lagos (17.3 DU).

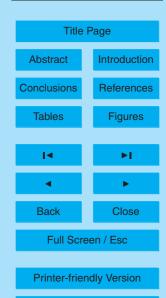
In the following, some observed interannual variabilities are highlighted. However our goal is more to give a first assessment of this tropospheric ozone variability and its probable causes rather than a detailed explanation of them that would require further studies. A 12 DU difference, essentially confined to the lower layers (below 500 hPa), is calculated between March 2005 and March 2006 showing a lower (higher) amount of ozone in 2005 (2006). Such a difference is less pronounced in February and April. However OMI/MLS does not capture this difference (Fig. 14 and next subsection), suggesting that unfortunately sparse sampling (3 soundings each month) might be responsible of this difference. An 8.4 DU difference is calculated between September 2005 and September 2006 showing a lower amount of UT ozone in 2006 in agreement with the mean ozone profiles in Fig. 3. Only three ozone soundings were performed for each September month, however this difference is supported by OMI/MLS TCO retrievals (Fig. 14 and next subsection). NCEP Reanalysis show lower Outgoing Long wave Radiation (OLR) above Western and Equatorial Africa in September 2006 compared to September 2005 (not shown). Thus more convective events in 2006 decreasing UT

#### **ACPD**

9, 11221–11268, 2009

### Overview of Cotonou ozone soundings

V. Thouret et al.





ozone by uplifting poor ozone air masses from the boundary layer are likely to be responsible for part of the variability between the sampled profiles. More specific studies should be made to investigate these hypotheses. Lower ozone columns are found during the dry season 2006 compared to 2005 with -7.6 DU (20%) and -21.9 DU (48%) 5 in November and December, respectively. The monthly mean profiles for December (Figs. 3 and 6) show that the differences are essentially in the lower troposphere under the AEJ influence (above the Harmattan layer and below 5 km or 500 hPa) and in the upper troposphere (above 10 km). Those two parts contribute equally to the interannual difference. 61% and 48% of this difference is confined in the lower troposphere below 500 hPa in November and December, respectively. The variability in the lower part of the troposphere is probably due to enhanced biomass burning emissions in 2005 in Africa (as discussed previously in Sect. 3.1.2). Logan et al. (2008) also found lower ozone in the middle troposphere over Equatorial Africa in December 2006 compared to 2005 using data from the Tropospheric Emission Spectrometer (TES) and suggested that these changes could be related to the late 2005 drought in eastern Africa leading to enhanced biomass burning as well as lower NO<sub>x</sub> production from lightning in 2006 compared to 2005 leading to lower ozone production in the UT in 2006. The 2005–2006 difference in the upper part of the troposphere might be due to enhanced convection in December 2005 leading to more NO, in the UT and more biomass burning pollutants up-lifted, which favour enhanced ozone production.

A secondary maximum is seen in JJA related to biomass burning plume intrusions from the Southern Hemisphere as discussed in Sect. 3.2. Here we attempt to assess the contribution of the Southern Hemisphere influence on the tropospheric ozone total amount by selecting and comparing two groups of ozone profiles (with and without ozone enhancement). For this comparison we use only July and August profiles to avoid the variations of upper tropospheric ozone levels which are clearly higher in June compared to July and August. During July and August 2005 and 2006, 18 ozone soundings were performed and 10 of them present an ozone layer located between 750 hPa and 500 hPa. When processing these two groups of profiles separately,

#### **ACPD**

9, 11221-11268, 2009

### Overview of Cotonou ozone soundings

V. Thouret et al.





we found a 3.46 DU difference in the total tropospheric ozone amount (41.4 DU and 37.9 DU for profiles with and without the ozone layer, respectively, see Table 3). Ozone enhancements from southern biomass burning pollution are located between 750 hPa and 500 hPa, consequently the biomass burning influence is seen mostly in the third level (700-500 hPa) and little in the second level (900-700 hPa). Therefore, a difference of 4.99 DU between 900 and 500 hPa is computed. However this positive difference is balanced by a 1.66 DU negative difference between 500 and 100 hPa. These results are consistent with those found by Mari et al. (2008) and support the hypotheses of a mid-troposphere enrichment of CO and ozone from biomass burning during the active phase of the southern AEJ (AEJ-S) whilst during the AEJ-S break phase, pollutants are first confined to the continent where they can reach a convective region further north and then be up-lifted to the upper troposphere. The up-lifting of biomass burning pollutants from the Southern Hemisphere contribute to the 1.66 DU. However, the electrical activity associated with the convective systems produces nitrogen oxides which may also enhance ozone levels in the UT (e.g. Martin et al., 2000; DeCaria et al., 2005; Sauvage et al., 2007d; Saunois et al., 2008a). Overall the biomass burning pollution from the Southern Hemisphere influences West Africa, at least the region of the Guinean coast close to Cotonou, either in the mid- or in the upper-troposphere depending on meteorological conditions and leads to up to 5 DU (12%) ozone enhancement in July and August.

### 5.2 Comparison between ozone soundings and OMI/MLS tropospheric residuals

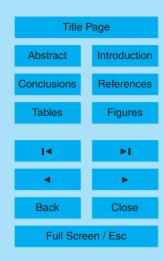
Since the launch of Aura on 15 July 2004, tropospheric ozone columns or residuals can be derived by subtracting the stratospheric column estimated by Aura MLS (Microwave Limb Sounder) from the total ozone column measured by Aura OMI (Ozone Monitoring Instrument). OMI/MLS tropospheric ozone residuals (TOR) have been compared with ozonesondes by Ziemke et al. (2006) (denoted as Z06 in the following) in the tropics and outside the tropics using a pressure-averaged ozone volume mixing ratio (VMR).

#### **ACPD**

9, 11221–11268, 2009

### Overview of Cotonou ozone soundings

V. Thouret et al.





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The tropopause pressure used by Z06 was determined from NCEP reanalysis using the 2 K/km thermal vertical gradient criterion and corresponds to an average of 100 hPa (~17 km) in the tropics with a range from 95 to 110 hPa over the two year period. Z06 did not find a substantial offset relative to ozonesondes using a pressure-averaged ozone mixing ratio (VMR). However their highest differences corresponded to tropical SHADOZ stations, Natal and Ascension with 5.9 and 3.1 ppbv. They found an average offset of 2 DU between OMI/MLS and several WOUDC/SHADOZ stations and pointed out that considering VMR reduces the scatter. Schoeberl et al. (2007) (denoted as S07 in the following) estimate the 200-hPa-to-surface ozone column using a trajectory model to increase the horizontal resolution of the stratospheric columns derived from MLS. Their product, TTOR 200TSC, shows an improvement over Z06, especially in the mid-latitudes. They bilinearly interpolate their TTOR 200TSC product to compare with ozonesonde points. They found a 2.4 DU offset in the tropics, with TTOR 200TSC being lower and a 5 DU standard deviation probably representing the total uncertainty of the measurements.

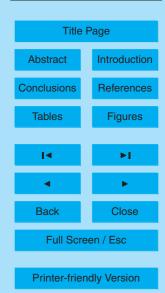
In Fig. 14, we compare the monthly means of OMI/MLS TOR derived by Z06 and publicly available from the NASA Goddard web page (http://hyperion.gsfc.nasa.gov/Data\_services/cloud\_slice) with the monthly means of ozonesonde columns (calculated from the ground to the altitude of the tropopause as defined in Sect. 4). The comparison shows a very good agreement between ozonesonde and OMI/MLS tropospheric columns regarding their seasonal cycle and interannual variabilities. In particular they both agree for smaller values in May 2005 and 2006 and September 2006. During the 26 month period, the highest tropospheric ozone amount over Cotonou was measured in December 2005 by the ozone soundings. This extreme value is also reported by OMI/MLS, but to a lesser extent though. The OMI/MLS tropospheric column reaches up to 41.4 DU in December 2005 whilst its mean value is 35.5 DU through the 26 months (i.e. 17% higher). Generally, OMI/MLS tropospheric residuals from Z06 exhibit a significant negative offset relative to ozonesondes: 11.8 DU and 6.7 DU in DJF and JJA, respectively (Table 4). Those results are greater than those found by Z06 or S07. The

### **ACPD**

9, 11221–11268, 2009

### Overview of Cotonou ozone soundings

V. Thouret et al.





different sampling frequency of ozonesondes (3 to 9 days a month) and of satellite data (almost everyday) used to compute monthly means as well as the missing ozone variation within the pixel size of OMI/MLS data (1°×1.25°) are two basic sources of discrepancy for such a comparison. This could be the main reason for large differences observed in December 2005 when 2 out of our 3 profiles were characteristics of an extreme small scale event. Also MLS stratospheric columns are biased such that they are slightly high by a few DU and assuming no bias for the OMI column, the TTOR would also be low by a few DU (S07). Z06 explicitly added ozone based on a recent evaluation of OMI and MLS offset differences (S07). However the MLS high bias does not explain our 7–12 DU differences.

It is worth noting that compared to Cotonou, most of the SHADOZ stations are subject to low amounts of pollution and pretty clean near the surface. Considering that OMI is not very sensitive near the surface due to strong Rayleigh scattering (S07), we have computed ozonesonde TCO minus the lowest layers below 850 hPa. These reduced columns compare better with the OMI/MLS tropospheric residuals (Fig. 14) and diminish the offsets to 5.5 DU and 2.8 DU for DJF and JJA, respectively (Table 3). A sensitivity test to the tropopause pressure of the column show that the ozone amount is not significantly affected by the tropopause pressure chosen (Fig. 14). These results confirm the low sensitivity of OMI in the boundary layer, which is more pronounced during the dry season when biomass burning pollution is at its highest in the lowest levels. Also using a model comparison Z06 noticed that a band of +10 DU difference centered over Northern Africa is persistent and coincides with mineral dust plumes from desert. This discrepancy was not explained but they suggested that desert dust affects either OMI measurements or the model. This assumption is in agreement with our higher difference in DJF compared to JJA; in DJF the Harmattan flux transporting mineral dust from the Sahara as well as biomass burning pollution reaches the Guinean coast, and so Cotonou.

### **ACPD**

9, 11221-11268, 2009

### Overview of Cotonou ozone soundings

V. Thouret et al.





#### 6 Conclusions

In this paper, we have presented the first ozone data set obtained through balloon-sondes launches in West Africa. During the AMMA campaign, a total of 98 vertical profiles over Cotonou, Benin, were measured over a 26 month period. These regular measurements document the seasonal and interannual variability of ozone in both the troposphere and the lower stratosphere over West Africa for the first time. This data set complements the MOZAIC observations made from Lagos between 0 and 12 km. Moreover it provides a first assessment of the ozone distribution in the UTLS and gives the first tropospheric columns of ozone based on in-situ data in this region.

The comparison with aircraft data has shown that the ozone concentrations from the soundings are in reasonable agreement with the ones recorded by UV analyzers. As expected, Cotonou ozone soundings show the same seasonal characteristics as those derived from the MOZAIC measurements over Lagos in the lower and midtroposphere. Both data sets highlight the unique way in which West Africa is impacted by two biomass burning seasons from two different hemispheres. This leads to ozone enhanced layers in the lower troposphere in DJF (due to burning in the Sahelian band) and in JJA (due to burning in southern Africa) as previously described by Sauvage et al. (2005, 2007a) and Mari et al. (2008). Even though a 20% average difference has been calculated between the MOZAIC data and ozone soundings measurements between 2 and 8 km, no systematic difference has been found between the two data sets. On the contrary, high variabilities appear in each data set (examples of July and August for MOZAIC, March, September and December for the soundings). Thus, the rather high interannual variability of the ozone profiles throughout the troposphere is found to be a major characteristic of this region and has been highlighted in most of the sections. The example of the dry season 2005-2006 shows that the influences of biomass burning emission and dynamics changes on the ozone amount are large. This study has also shown that the enhancement of ozone originating from the biomass burning products in the Southern Hemisphere was more frequent during the wet sea-

### **ACPD**

9, 11221–11268, 2009

## Overview of Cotonou ozone soundings





sons of 2005 and 2006 than previously established based on the MOZAIC data set with 41% of the profiles showing ozone enhancement compared to 28%. This major feature should be considered when analyzing this period. The high interannual variability of ozone in this region seems to be related to the multiplicity of the trace gas sources with their own variabilities (biomass burning, lightning, megacity of Lagos) and also to the particular atmospheric circulation. This latter factor combines the characteristics of a transition zone between the monsoon and Harmattan flows and a region under monsoon and convective influences; the three of them have significant temporal variations. Indeed, such variabilities might be typical of Equatorial Africa. Since December 2005, a MOZAIC aircraft has been operated by Air Namibia and sampled the upper troposphere between Windhoek, Namibia and Europe (either London, UK or Frankfurt, Germany). The mean ozone values in the UT around 6–7° N along the flight tracks also show high interannual variability with differences of up to 20-30 % from one year to the next especially in February, May, June, October and December (Cammas, personal communication). As the flights go east of Cotonou and Lagos (~10–15° E), a quantitative comparison would not have been relevant here. However the Air Namibia data set confirms the high interannual variabilities of ozone concentrations in the African upper troposphere.

The West African UTLS shows the same patterns regarding the ozone and temperature annual cycle, as those found over other tropical stations. However the ozone concentrations registered in the UTLS above Cotonou are higher than expected from previous tropical studies. Ozone concentrations range around 90–110 ppbv on average between 12 and 17 km, and reach up to 150 ppb in August and September. A first quantification of tropospheric ozone columns based on in-situ data has been derived over Cotonou. They maximize in DJF with 45 DU on average. Such a value is higher than the one calculated by Sauvage et al. (2006) from the MOZAIC profiles over Lagos (41 DU). This difference is mainly attributed to their underestimate of the ozone distribution in the upper troposphere. These tropospheric columns derived from the ozonesoundings have been compared to satellite data and a fairly good agreement

#### **ACPD**

9, 11221–11268, 2009

## Overview of Cotonou ozone soundings

V. Thouret et al.





has been found with the OMI/MLS tropospheric column regarding the seasonal and interannual variations. Quantitatively, the difference is around 7–12 DU and can be reduced to 3–6 DU if we omit the lowest layers of the troposphere (below 850 hPa) in the calculation from the soundings, suggesting that much of the negative bias may be attributed to the lack of sensitivity of OMI to the boundary layer.

The AMMA program has allowed the acquisition of the data set presented here that has already been used for various studies such as satellite validations (Schoeberl et al., 2007; Jiang et al., 2007), data analysis (Reeves et al., 2009) and models studies (Mari et al., 2008; Williams et al., 2009b,a). We believe its use will last long after the AMMA programme. Quantifying the tropospheric ozone budget over West Africa will require further analysis. For example it is still a challenge for Chemical Transport Models to be able to reproduce these seasonal characteristics (Saunois et al., 2008b; Williams et al., 2009b,a).

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### **ACPD**

9, 11221-11268, 2009

## Overview of Cotonou ozone soundings





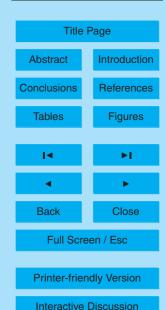
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### **ACPD**

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### Overview of Cotonou ozone soundings



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### **ACPD**

9, 11221–11268, 2009

### Overview of Cotonou ozone soundings

V. Thouret et al.



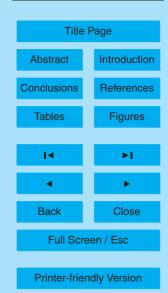


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V. Thouret et al.





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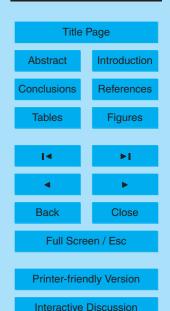
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### **ACPD**

9, 11221–11268, 2009

## Overview of Cotonou ozone soundings

V. Thouret et al.





**Table 1.** Number of ozone soundings recorded over Cotonou during AMMA.

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
2004												2
2005	2	4	2	4	3	4	0	3	3	5	4	3
2006	5	3	4	3	4	9	8	7	3	3	3	1
2007	6											

9, 11221-11268, 2009

# Overview of Cotonou ozone soundings

Title Page					
Abstract	Introduction				
Conclusions	References				
Tables	Figures				
I₫	►I				
4	-				
Back	Close				
Full Screen / Esc					
Printer-friendly Version					
Interactive	Discussion				

**Table 2.** Monthly mean tropospheric ozone columns (from the ground to the tropopause as calculated in Sect. 4) in Dobson Unit (DU) based on ozone soundings over Cotonou.

	Jan	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep	Oct	Nov	Dec
2004												39.4
2005	49.0	43.4	32.5	41.8	37.7	43.8	_	42.0	39.2	42.3	46.1	67.2
2006	49.5	43.9	44.7	43.3	38.4	45.9	41.8	40.5	30.8	40.3	38.5	45.3
2007	48.26											

9, 11221–11268, 2009

# Overview of Cotonou ozone soundings

V. Thouret et al.

Title Page					
Abstract	Introduction				
Conclusions	References				
Tables	Figures				
I⁴	►I				
4	•				
Back	Close				
Full Scre	en / Esc				

**Table 3.** Mean tropospheric ozone columns in DU over Cotonou during July and August for profiles with and without an ozone enhancement around 700–500 hPa. Standard deviation for each group appears in brackets.

	1000-900 hPa	900-700 hPa	700-500 hPa	500-250 hPa	250 hPa-Top	1000 hPa-Top
With O <sub>3</sub> layer	2.45 (±0.42)	6.95 (±1.3)	11.20 (±2.10)	11.41 (±3.00)	9.50 (±2.20)	41.40 (±3.97)
W/o O <sub>3</sub> layer Diff $(W-W/o)$	,	5.42 (±0.76) 1.53	7.74 (±0.59) 3.46	13.07 (±1.66) -1.66	9.52 (±1.49) 0.02	37.94 (±3.30) 3.46

9, 11221–11268, 2009

# Overview of Cotonou ozone soundings

V. Thouret et al.

**Table 4.** Mean tropospheric ozone columns in DU over Cotonou during DJF and JJA for OMI/MLS and ozonesonde columns and their absolute and relative differences. RS1 correspond to the columns from the surface up to the tropopause level and RS2 correspond to the columns from 850 hPa up to the tropopause.

		Mea	ın DJF	Mea	an JJA	
OMI/MLS (DU)		3	6.4	36.1		
RS1 (surf-Top)	(DU)	4	8.2	42.8		
RS2 (850 hPa-	Top) (DU)	4	1.9	38.9		
RS1-sat (DU)	RS1-sat (%)	11.8	24.5%	6.7	15.7%	
RS2-sat (DU)	RS2-sat (%)	5.5	13.1%	2.8	7.2%	

9, 11221–11268, 2009

# Overview of Cotonou ozone soundings

V. Thouret et al.

Title Page					
Abstract	Introduction				
Conclusions	References				
Tables	Figures				
I⁴	►I				
4					
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Back	Close				
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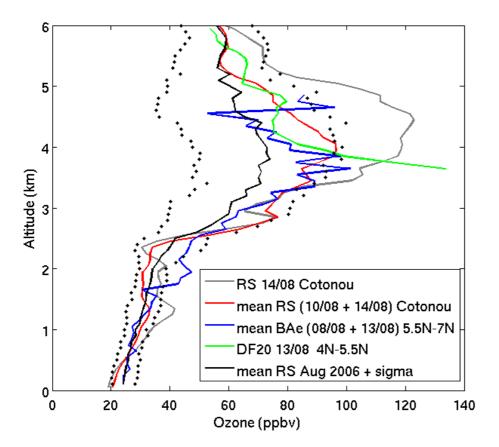


**Fig. 1.** Map of West Africa to highlight the position of Lagos, 110 km east of Cotonou. Cotonou is also highlighted on the map of the SHADOZ sites as visible on the web site http://croc.gsfc. nasa.gov/shadoz/.

9, 11221–11268, 2009

# Overview of Cotonou ozone soundings

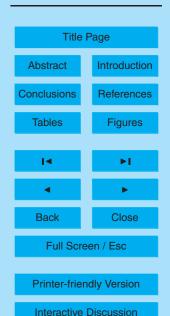




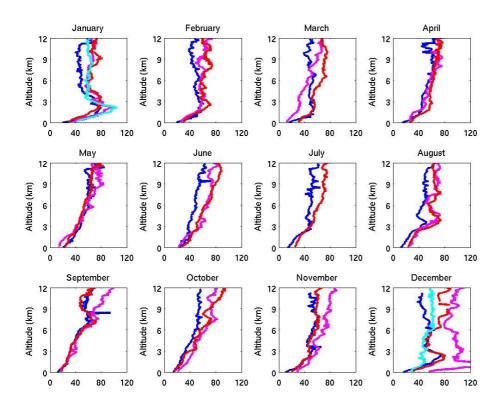
**Fig. 2.** Vertical profiles of ozone up to 6 km from the RS data set: August monthly mean and standard deviation (black), average of the two soundings on 10 and 14/08 (red), sounding on 14/08 (grey); from BAe-146 in the region between 5.5° N and 7° N on the 08 and 13/08 (dark blue); and from D-F20 in the region 4–5.5° N on the 13/08 (green).

9, 11221–11268, 2009

# Overview of Cotonou ozone soundings



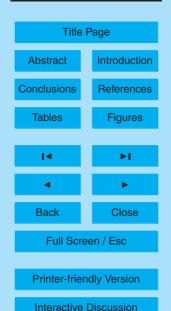




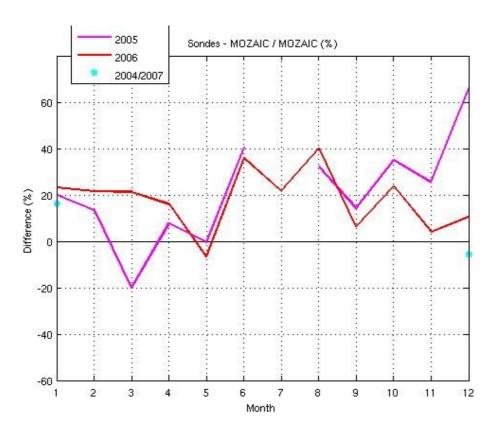
**Fig. 3.** Monthy mean vertical profiles of  $O_3$  from the Cotonou RS (December 2004 and January 2007: light blue, 2005: pink and 2006: red) and from the MOZAIC climatology (dark blue) over Lagos (1° E from Cotonou) from the surface to 12 km.

9, 11221–11268, 2009

# Overview of Cotonou ozone soundings



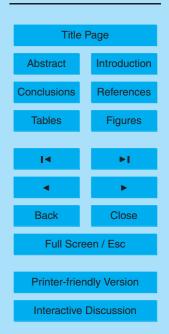




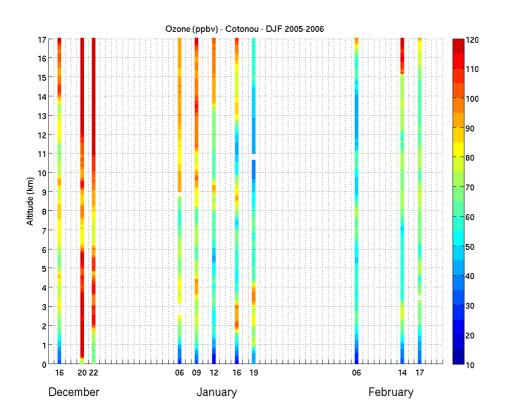
**Fig. 4.** Differences calculated between Cotonou RS and MOZAIC climatology over Lagos between 2 and 8 km for December 2004, years 2005 and 2006 and January 2007.

9, 11221–11268, 2009

# Overview of Cotonou ozone soundings







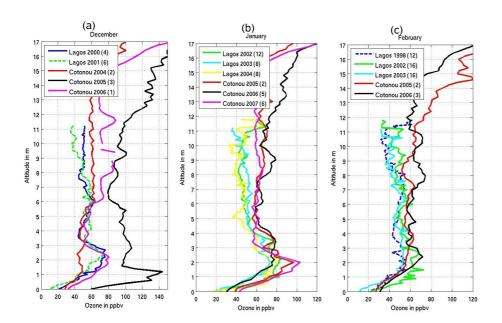
**Fig. 5.** Time series of the ozone profiles from the surface up to 17 km during December-January-February 2005/2006.

9, 11221–11268, 2009

# Overview of Cotonou ozone soundings



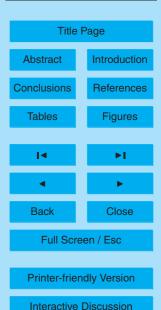




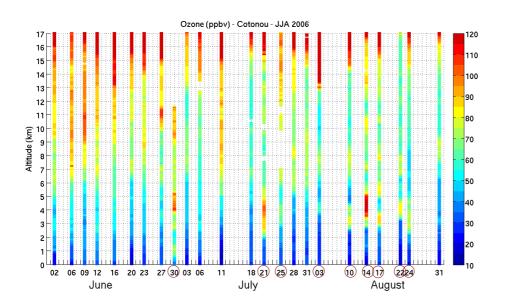
**Fig. 6.** Monthly mean ozone profiles over Lagos and Cotonou in December **(a)**, January **(b)** and February **(c)**. Colour Code for Cotonou RS: dry season 2004–2005 (red), 2005–2006 (black), 2006–2007 (pink).

9, 11221–11268, 2009

## Overview of Cotonou ozone soundings





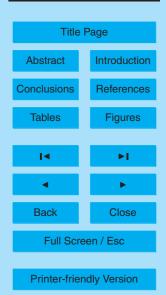


**Fig. 7.** Time series of the ozone profiles from the surface up to 17 km during June-July-August 2006. Dates with  $O_3$  enhancement are circled.

9, 11221–11268, 2009

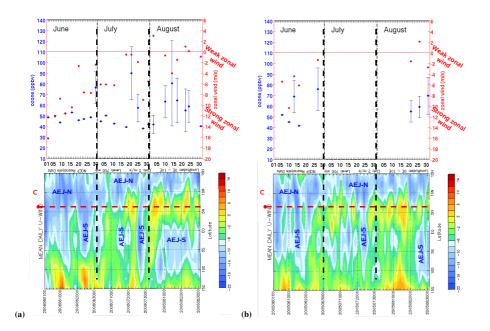
## Overview of Cotonou ozone soundings

V. Thouret et al.





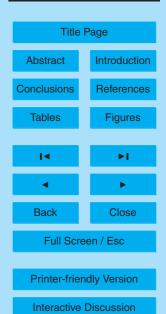
Interactive Discussion

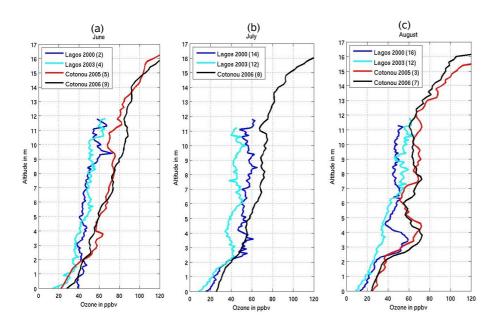


**Fig. 8. (a)** Time series during the period June-July-August 2006. Top panel: ozone concentration (blue) and zonal wind speed (red) measured by radiosounding and averaged between 600 and 800 hPa. Ozone data points for soundings showing an ozone enhancement from biomass burning have vertical error bars. These vertical bars represent the magnitude of the ozone-enhanced layer relative to the background profile. The total length of each bar is calculated from the difference between the maximum of the ozone concentration in the layer and the background value at the same levels. Bottom panel: Time-latitude diagram of the mean zonal wind speed between 0° E and 10° E from NCEP reanalysis at 700 hPa. The red line corresponds to the latitude of Cotonou. **(b)** Same as in **(a)** but for the period June-July-August 2005.

9, 11221-11268, 2009

## Overview of Cotonou ozone soundings



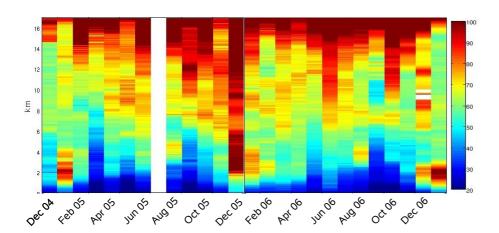


**Fig. 9.** Monthly mean ozone profiles over Lagos (2000: dark blue and 2003: light blue) and Cotonou (2005: black and 2006: red) in June **(a)**, July **(b)** and August **(c)**.

9, 11221–11268, 2009

## Overview of Cotonou ozone soundings

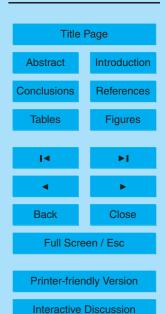




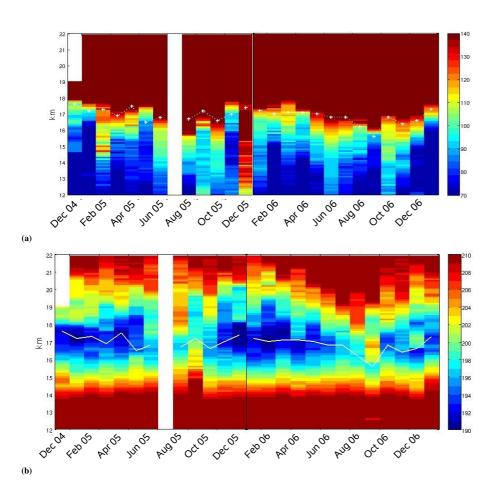
**Fig. 10.** 26 month timeseries of ozone (ppb) monthly means over Cotonou between 0 and 17 km, averaged every 100 m.

9, 11221–11268, 2009

## Overview of Cotonou ozone soundings







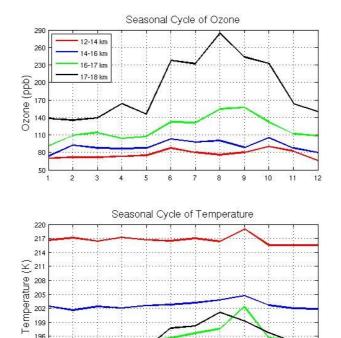
**Fig. 11.** 26 month timeseries of **(a)** ozone (ppb) and **(b)** temperature (K) monthly means over Cotonou between 12 and 22 km, averaged every 100 m.

9, 11221–11268, 2009

# Overview of Cotonou ozone soundings







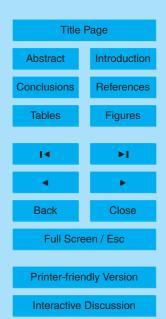
**Fig. 12.** Seasonal cycle of ozone (ppb) and temperature (K) averaged over the two years of the RS data set for four different layers, 12–14 km in red, 14–16 km in blue, 16–17 km in green and 17–18 km in black.

196 193

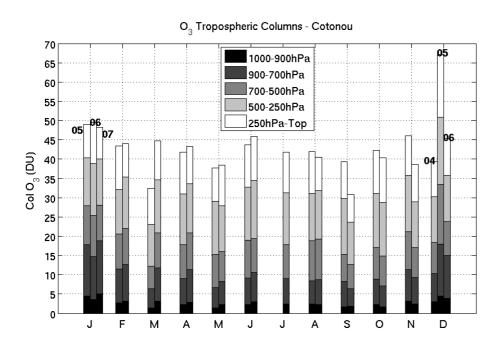
#### **ACPD**

9, 11221–11268, 2009

# Overview of Cotonou ozone soundings







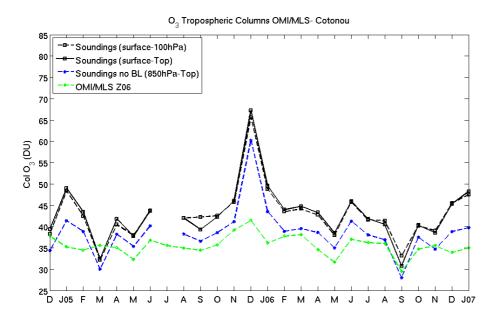
**Fig. 13.** Monthly mean tropospheric ozone columns in Dobson Unit (DU) over Cotonou between Dec. 2004 and Jan. 2007. Columns are divided into 5 layers from 1000 hPa to the tropopause (called Top and calculated as defined in Sect. 4).

9, 11221–11268, 2009

# Overview of Cotonou ozone soundings







**Fig. 14.** Two years of monthly mean tropospheric columns of ozone in DU over Cotonou from OMI/MLS Ziemke et al. (2006) compared with columns derived from ozone soundings from the surface to 100 hPa, the surface to the tropopause (called Top and calculated as defined in Sect. 4), and 850 hPa to the tropopause.

9, 11221–11268, 2009

# Overview of Cotonou ozone soundings



