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Atmospheric Chemistry and Physics Discussions

Interactive comment on "The influence of African air pollution on regional and global tropospheric chemistry" *by* A. M. Aghedo et al.

A. M. Aghedo et al.

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General comments

We would like to thank our referees for their detailed and thoughtful comments on our paper. Here is our final response to the comments raised by the anonymous referees. The referee's original comments are in italics.

Responses to Anonymous referee

I-) Major general concerns

1) Introduction: The goal is not very well presented here. I think it deserves more

than the two lines sentence lines 7 to 9. That should be stated at the end after having exposed the rationale of the study, the state of the art and brief details concerning the different sources investigated here.

We will restructure the introduction, and add more statements to strengthen the aim of the study.

2) Section 2, as well as the entire study is based on Rast et al., 2006 which is in preparation. It is a shame ! I think it is difficult for readers to fully trust such an important study drawing quantitative results without having access to the reference study for the ECHAM model. I found very frustrating the type of sentence "...described in Rast et al., 2006" for key informations like the sensitivity tests, the reference experiment and the influence of the injection heights for some emissions for example. I would suggest if possible to include some results of the future publication in this one. End of section 2, although with a lesser importance, I would like to read more details instead of "...described in Roeckner et al., 2006 and Aghedo et al., 2006" (non available papers by now.) Just add a few sentences to justify your choice and make the following more robust.

We will include a short summary of Rast et al. (in prep.) and the point in Aghedo et al., 2006 that justifies our decision. Roeckner et al., 2006 has now been published.

3) Section 4.1: Model validation This is my major concern overall. The paragraph and the analysis coming along are definitely too short and not convincing. Such a global analysis on the influence of the African air pollution to the rest of the world deserves a thorough evaluation over the African continent. Before drawing the conclusions, it would be nice to check the "accuracy" of the model in reproducing the main characteristics of the ozone (at least) distribution over Africa. The authors use the MOZAIC data recorded over Africa and some other profiles over the other

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continents (US, South-America, Europe and Japan). Some profiles are presented in January and in July and the overall good agreement is claimed without a proper discussion. They have chosen not to show the seasonal variations. I think this information is always important to give because it really indicates the influence of the most important sources, like biomass burning for example. The authors should have used the Sauvage et al., 2005 (ACP), 2006 (JGR) papers to perform their model evaluation over Africa. These two recent studies make a use of the MOZAIC data available over Africa to draw the main characteristics of the ozone distribution over the continent. Besides, by combining the MOZAIC and the SHADOZ data, they have also presented a regional aspect of the ozone distribution over Africa and the adjacent oceans. I suggest to substantially modify this sub-section to make it a more solid evaluation. I think the Sauvage et al., 2005,2006 papers give the lines to compare with the model. For example, the Brazzaville profiles (available in the MOZAIC data base and not shown here) should be very interesting to show as this region experiences an influence of the northern biomass burning although it is in the southern hemisphere. Besides, is the model able to reproduce the South Atlantic Maximum throughout the year ? This would argue in favour of a good representation of the transport outside Africa, before quantifying it in the following sections.

As suggested in this comment, we will extend MOZECH evaluation to include:
1. Detailed comparison with MOZAIC vertical profiles, with focus on six African stations (Cairo [Egypt], Abidjan [Cote d'Ivoire], Lagos [Nigeria], Brazzaville [Congo], Windhoek [Namibia] and Johannesburg [South Africa]). Brazzaville is included as suggested here, although no measurements were conducted over Brazzaville in November and December of years 1997 to 2002 (Table 2 in Sauvage et al., 2005). We will discuss the variability throughout the year, but we will show the profile plots for only December, January, February, June, July and August. We include 6 additional plots to show the influence of each of the African emissions on ozone at these six stations. Our model calculation also shows an enhancement over Brazzaville in DJF months, which our

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sensitivity studies show was due to biomass burning emissions. Since local biomass burning is at the barest minimum at this time of the year over Brazzaville (we will show the seasonal cycle of biomass burning CO and NOx emissions), our explanation is that this could be coming from northern part of Africa where biomass-burning activities are occurring, as shown by the backward trajectories in Sauvage et al., 2005. MOZECH does not show ozone enhancement over Western Africa (i.e. Lagos and Abidjan) in July and August as shown by MOZAIC. Another important feature of MOZECH vertical profile is that the dry season ozone enhancement occurs at the surface 750hPa, while that of the measurement occurs higher up in the troposphere at about 850 – 600hPa, leading to a high bias at the lower troposphere. At the surface this bias ranges from 35 – 50ppbv over Lagos and 50 – 100ppbv over Abidjan in DJF and about 35 – 60 ppbv over Brazzaville in JJA. The reasons for this may be three fold. Firstly, MOZAIC African measurements may have missed the high surface to lower tropospheric ozone enhancement occurring outside the airports, considering that most airports are located far away from burning sites, (e.g. Abidjan airport is situated at about 30km east of the city). Secondly, we guess that MOZECH may have problem in simulating the dry deposition well during the dry season. Finally, lack of aerosols (especially dust) in MOZECH may have also contributed to this problem. However, to fully unravel the extent of these three reasons and their contribution to the overestimation would be beyond the scope of this paper.

2. We will also compare MOZECH with SHADOZ tropospheric ozone time series measured at 800hPa, 500hPa and 300hPa over Irene, Ascension Island, Reunion and Nairobi. Due to data gap in SHADOZ data, the 5-year time series include any 5 most-complete consecutive years in 1998 – 2004 at all the stations. These are 1998 – 2002 in Reunion and Nairobi, and 1999 – 2003 in Irene and Ascension Island. Four additional plots are also included to show MOZECH seasonal ozone bias (i.e. MOZECH SHADOZ in JFM, AMJ, JAS, and OND) at these four stations.

3. Surface CO concentration of MOZECH will be compared with CMDL CO surface concentration at Ragged Point (Barbados), Terceira Island (Azores), Ascension

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Island, Tenerife (Canary Island), Assekrem (Algeria), Sede Boker (Negev desert, Israel), Mahe Island (Seychelles), Crozet Island and Syowa, (Antarctica). These 9 stations include 1 continental African station (i.e. Assekrem, Algeria) and 8 stations downwind of Africa. The surface CO concentration calculated by MOZECH shows good agreement with CMDL stations data, with a slight underestimation at Barbados, Azores and Tenerife and an overestimation at Negev desert, Israel.

4. We also include the summary of MOZECH performance in the recent IPCC/ACCENT intercomparison experiment (Stevenson et al, 2006) and a short summary of Rast et al. (in prep.).

The total tropospheric ozone column (TTOC) simulated by MOZECH confirms that there is no tropical ozone paradox over continental Africa as shown in Sauvage et al, 2006. It also confirms the southern Atlantic ozone maximum in JJA and SON, when the TTOC over the southern Atlantic ocean is greater that that over the northern Atlantic, but due to the overestimation recorded over western Africa during the dry season, which definitely affects the downwind transport to northern Atlantic, the DJF and MAM TTOC is greater over northern Atlantic than southern Atlantic.

4) Sections 4.3 and 4.4: As a general comment for the entire paper, I often miss important details. In these sections for example, I would have appreciated some details concerning the calculation of TOB. I am not very familiar with such calculations as many readers probably. Why is the tropopause set at 200 hPa? It is a little bit low in altitude for the tropical regions... especially to quantify the impact of lightning and biogenic components rapidly transported in the upper troposphere as it is shown. Table 3 needs also further explanations. How are calculated the columns 2 to 6 ? Please give also a sentence to explain the big difference between the sum of columns 2 to 6 and column 7 in Table 3. Still concerning Table 3 and comments going along in the text: Is there a real need for making a difference between biogenic VOC and biogenic CO, H2 and soil NOx.

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Tropospheric ozone burden is also known as tropospheric ozone budget. This is the mass of ozone in the troposphere (expressed as $g(O_3)$). For species, s with volume mixing ratio (VMR i.e mole fraction), Y_s and mass mixing ratio (MMR i.e. mass fraction), X_s. Let us define the following: (note that what is usually measured e.g. during ozonesondes or aircraft campaign is VMR)

n_s = mole of species, s

n_a = mole of air

m_s = mass of s at any point in the troposphere

m_a = mass of air at any point in the troposphere

M_s = molecular mass of species,s

M_a = molecular mass of air

It is known that:

Y_s = n_s / n_a ..... (1)

X_s = m_s / m_a ..... (2)

m_s = $n_s \times M_s$ ..... (3)

m_a = $n_a \times M_a$ ..... (4)

Using Eqs. (3) and (4) in Eqn. (2) yields:

X_s = $(n_s \times M_s)$ /$(n_a \times M_a)$ .....(5)

Therefore:

X_s = $(M_s/M_a) \times Y_s$ ..... (6)

Hence,

m_s = $(M_s/M_a) \times Y_s \times m_a$ ..... (7)

We know that:

Air Pressure, P_a = $(m_a \times g)/A$ .....(8)

Therefore, m_a = $P_a \times A/g$ ..... (9)

Where g = acceleration due to gravity (9.8 m/s^2) and A = Area of the grid box containing m_s. Substituting Eqn. (9) into Eqn. (7) produces:

m_s = $(M_s \times Y_s \times \mathrm{P}_a \times A)/(M_a \times g)$ ..... (10)

If one start from mass mixing ratio, Equation 10 will be :

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$m_s = X_s \times P_a \times A/g$ ..... (11)

We will include only the discrete form (i.e. specifying latitude, longitude and levels) of equations 10 and 11 in the paper. Rather than discussing this in the introduction, we will discuss it in section 4.3.

Using Equation 11, we calculated the total amount of ozone found in the troposphere over the respective regions caused by emissions originating from Africa. To achieve this, we first calculate the difference of ozone MMR in our reference experiment and the respective sensitivity experiments. Using the region mask, we then convert whatever MMR is found in a certain region to burden (i.e. g (O3)) according to Equation 10. Therefore, each of the entries in columns 2 – 6 of Table 3 represents this value.

On the other hand, column 7 of Table 3 gives MOZECH total tropospheric ozone budget over the respective regions. That is, as calculated from the reference experiment. Therefore, column 7 gives the total tropospheric ozone burden over each region as simulated by MOZECH, whereas the summation of column 2 – 6 gives the amount of this total tropospheric ozone budget that can be attributed to African emissions. Hence, the big difference tell us how much of tropospheric ozone over the regions are coming from other sources (such as transport from the stratosphere and ozone produced by emissions of other regions apart from Africa). For example, calculating this difference shows that only about 28% of tropospheric ozone over Africa can be attributed to African emissions. This means that about 72% of African tropospheric ozone is due to transport of ozone and its precursors from other continents and transport from the stratosphere.

We have redefined the tropopause to 150ppbv O3 level as used in Stevenson et al., 2006. Our earlier calculation indeed excludes the high ozone enhancement due to biogenic and lightning emissions at the upper troposphere (which extends to higher

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levels above 200hPa). Therefore the biggest change (ranging between 10% - 25%) in Table 3 occurs at the tropical regions (e.g. Africa, Latin America and South-east and south-central Asia) and for biogenic and lightning emission entries. The global tropospheric ozone burden due to biomass burning, biogenic VOC, biogenic, lightning and anthropogenic emissions is now 9.52 Tg, 15.1 Tg, 19.59 Tg, 9.0 Tg, 4.67 Tg, respectively. Global tropospheric ozone burden calculated by MOZECH is 384.32 Tg. While the total tropospheric burden over Africa also changed from 29 Tg to 33 Tg.

Concerning the distinction between biogenic VOC and other biogenic emissions, see our response to ML major comment 5.

II-) Specific points:

Introduction: Page 5800, Line 1 to 3: This sentence from Marufu et al. (2000) is surprising as the 16% contribution from biomass burning is twice the amount calculated in this study (from Table 5). Such a statement should be discussed below, maybe in the conclusion. TOB needs to be defined here. Page 5800, Line 24: This sentence is quite ambiguous with the above arguments stating that the lightning emissions should be close to the lower limit of a few Tg(N)/yr. Remove it or clarify its use.

Marufu et al., 2000 calculated the influence of **global biomass burning emissions** (i.e. 287 Tg (C)/yr CO, 52 Tg (C)/yr NMHC and 8 Tg (N)/yr NOx, note that their biomass burning emissions includes biofuel emissions, whereas biofuel emissions are included as anthropogenic emissions in our study) on African tropospheric ozone burden. They found that this contributes about **4.16 Tg to Africa tropospheric ozone burden (i.e. 16% of 26 Tg)**. We calculated the influence of **African biomass burning emissions** (this is 93 Tg (C)/yr CO, 8 Tg(C)/yr NMHC and 4.6 Tg (N)/yr NOx) on African tropospheric ozone burden and in Table 3 we show that they contributes **2.5 Tg**

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to African tropospheric ozone burden , so this should not be a surprise, given that African biomass burning emissions is about 44% of global biomass burning emissions. Our clarifications here answers the questions raised by the referee on Table 1 and Table 2. Please note the words in bold.
We have assumed the referee is referring to Table 3 and not Table 5.
We cannot quite understand the ambiguity of the statement in line 24 of page 5800. The statement corroborates the fact that increasing lightning NOx emissions increases tropospheric ozone, but it also shows that doubling global lightning NOx emissions does not lead to a doubling in the tropospheric ozone produced. Therefore revealing that there is a non-linear relationship between precursor emissions and photochemical tropospheric ozone.

Section 4.2 and 4.3: Page 5808, lines 18-20: The colour coding for Figure 3 does not make this comment very visible. Page 5808, last paragraph starting line 21: I think Figure 4 shows a marked seasonal variation on contrary of what is said in the text.

We will re-plot the figure using different colour schemes. We did say it shows seasonal variation, but that it somehow less pronounced when compared to seasonality of biomass burning produced ozone. We will clarify this statement in our revision.

Section 4.5: Page 5815, lines 5-6: Overall this paragraph is very interesting. I would like to read more details on this particular statement. is it due to the particular monsoon dynamics or to the El-Nino events that occurred during the simulation period ? Please say more.

Influence of African emissions on Southeast Asia tropospheric ozone occurs throughout the year, with maximum impact in March-April (MA) and October-December (OND). This influence is dominated by biogenic and lightning emissions, which together accounts for about 77% of the African emissions generated tropospheric ozone burden

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(TOB) over Southeast Asia. The least TOB is found in July September (JAS). MA and OND TOB over Southeast Asia due to African emissions is a factor of 2 higher than that of JAS. Generally January-February and May-June are transition period between these two distinct extremes. In JAS and the transition periods, African biogenic and biomass burning emissions wields the greatest influence on TOB over Southeast Asia. The high inter-annual variability we calculated is driven by the particularly low and high transport from Africa to Southeast Asia in our 1999 and 2001 simulation-year respectively, which causes the TOB over Southeast Asia to be about 18 34% lower and 24 31% higher than the 5-year average TOB. This may be connected with the cold and warm anomaly in the sea surface temperature (SST) in 1999 and 2001 respectively (we will include figures of this anomaly). This warm i.e 2001 (or cold i.e. 1999) induces a weakening (or strengthening) of the African easterly jet (references will be included), which increases (or reduces) the amount of substance transported from Africa eastwards (streamlines plots will be included). This study therefore shows a teleconnection of El Nino and La-Nina on transport from Africa to Southeast Asia. Please note that we have performed an AMIP2 run, therefore the years discussed above may be arbitrary.

We will include this discussion and more discussion concerning the simulated CO and NO2 concentration over this region in response to M. Lawrence major comment 7.

Section 4.6: The schultz et al., 2006 reference is missing in the list.

Schultz et al., 2006 is a paper in review, hence it is included as footnote 1.

Table 1 and 2: The biomass burning emissions in Table 1 are quite different from Marufu et al., (2000). I think that deserves a brief comment. Besides, Table 2 shows that Africa contributes to about 44 % of the global biomass burning emissions

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(43% for CO, 42% for NMHC and 46% for NOx). Isn't it too much ? If it's true, how come the impact is only of 2.4 % ?

We will include an additional row to Table 1 and Table 2 to indicate global and African emission estimates used in Marufu et al, 2000. Note that these two tables will be combined into one according to suggestion of ML minor comment 16. We would like to add here that our emissions are within the same range of the emissions in Marufu et al., 2000, except for their global biogenic emissions, which is about 50% lower than our values. Our African biomass burning emissions are within the range of the current estimates that can be found in the literature. Please also note that these same emissions are used in the just concluded IPCC/ACCENT inter-comparison experiment in preparation for the IPCC fourth assessment report (e.g. Stevenson et al., 2006).

Again we want to stress that only African emissions sensitivity study is conducted in this study. Given that Table 1 and 2 shows that **Africa biomass burning emissions** contributes only 19%, 0.9% and 9% to **global CO, NMHC and NOx emissions**, their 2.5% contribution to global tropospheric ozone should not be surprising. Also see our response to specific comment 1. We would like to point out that there are other sources of tropospheric ozone besides those produced in photochemical reactions involving emissions. Ozone is one of the "normal" tropospheric trace gases; hence it will still be present in the troposphere (though at a lesser amount) if there are no emissions. The best way to interprete our results in Table 5 has been explained in our response to the major concern 4.

References: Sauvage B., V. Thouret, J- P. Cammas, F. Gheusi, G. Athier and P. Nédélec, Tropospheric ozone over Equatorial Africa: regional aspects from the MOZAIC data. Atmos. Chem. Phys., 5, 311-335, 2005. Sauvage B., V. Thouret, A.M. Thompson, J.C. Witte, J- P. Cammas, P. Nédélec, and G. Athier, Enhanced View of

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the "Tropical Atlantic Ozone Paradox" and "Zonal Wave-one" from the In-situ MOZAIC and SHADOZ Data, J. Geophys. Res, January 2006. Interactive comment on Atmos. Chem. Phys. Discuss., 6, 5797, 2006.

These references will be included in the paper, where relevant.

Interactive comment on Atmos. Chem. Phys. Discuss., 6, 5797, 2006.