Atmos. Chem. Phys. Discuss., 6, S5072–S5084, 2006
www.atmos-chem-phys-discuss.net/6/S5072/2006/

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.

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Received and published: 5 December 2006

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 referee Mark Lawrence. The referee's original comments are in italics.

Final response to referee Mark Lawrence

Major comments

1) The title of the manuscript is "influence ... on tropospheric chemistry"; however, the

focus of the analysis is exclusively on changes in ozone due to removing various emissions. Considering other important components of atmospheric chemistry, such as CO, NOx, and OH, would strengthen the manuscript considerably and fit better with the title.

We will add discussions and figures of tropospheric CO, NOx and OH due to each of the African emissions, where necessary. We will also add the evaluation of surface CO concentration calculated by MOZECH (see response to comment 3).

2) Throughout the paper, the term "contribution" of various emissions to the total ozone burden is used. This is not a correct interpretation of these types of simulations. This does not at all devalue this type of study, it just means a more careful interpretation and more accurate discussion is needed. When emissions are removed, or even slightly perturbed, in a nonlinear chemical system, feedbacks result in a change in other gases which can be either larger or smaller than the actual contribution of the individual emissions. The only way to properly assess actual contributions is through tagged tracers, but that is known to be difficult for ozone. These simulations in which emissions are removed show the net influence of those emissions within the full chemical system (thus, the sum of influences from all emissions could be more or less than 100%). This is still very informative, for instance politically, for understanding how the chemical system would change if certain emissions were increased or decreased, though there are secondary non-linearities which also influence the interpretation. A careful discussion of this with examples for NOx and a quantification of two types of nonlinearities for India and its outflow is given in Kunhikrishnan, T. and M. G. Lawrence, Sensitivity of NOx over the Indian Ocean to emissions from the surrounding continents: nonlinearities in atmospheric chemistry responses, Geophys. Res. Lett., 31, doi:10.1029/2004GL020210,2004. This should be cited in section 3, in the paragraph "We are aware ...", along with the seminal works on this by Prather and colleagues, and used to help with properly formulating the discussion.

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The aim of this study is to investigate “the influence of each of African emissions categories on tropospheric chemistry”, therefore we have replaced all the occurrence of the term “contribution ... to total tropospheric ozone burden” by either “influence of ... on total tropospheric ozone burden” or “global (or African) tropospheric ozone burden ... is most sensitive to ...” according to the suggestion. The citations will be included.

3) Although I am a strong supporter of the need for proper evaluation of models (just like the characterization of measurements instruments) before they are put to substantial scientific use, the short model “validation” given in section 4.1 is not very convincing of the quality of the model for this study. The free troposphere, as indicated in section 4.1, is indeed often in reasonable agreement. However, the surface, which is the topic of section 4.2, but which is not mentioned in 4.1, disagrees substantially at several locations, including those in Africa (Johannesburg, Cairo, Lagos...), and even has the wrong lower tropospheric gradient over Johannesburg in July. There is no indication given of how all the precursor gases (NOx, CO, etc.) behave, so it would not be possible to diagnose the cause of these deficiencies. I would therefore suggest to remove this brief comparison with the MOZAIC data, and replace it with a short summary of the salient points from the more substantial evaluation which is indicated to be currently in preparation by Rast et al. Note also that the term “validation” is a misnomer which is frequently used in this context; it implies that one believes the subject of validation (in this case, the global chemistry-climate model) is actually correct, and comparisons with observations are being sought to demonstrate that this is so. The more proper term is “evaluation”, which implies that one is determining the strong and weak points of the model, indicative of the current state of the science and model development.

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We decided to keep section 4.1 by extending the evaluation in the following ways:
(a) MOZECH vertical ozone profiles comparison to MOZAIC data will be focused 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 by the anonymous reviewer (major comment 3), although no measurements were conducted over Brazzaville in November and December of years 1997 – 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. To explain ozone enhancement recorded over Brazzaville in DJF months, we also include a plot showing seasonal variation of biomass burning CO and NOx emissions in Lagos, Abidjan and Brazzaville.
(b) We will 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) vertical profiles at the 4 stations.
(c) Surface CO concentration of MOZECH will be compared with CMDL CO surface concentration at Ragged Point (Barbados), Terceira Island (Azores), Ascension Island, Tenerife (Canary Island), Assekrem (Algeria), Sede Boker (Negev desert, Israel), Mahe Island (Seychelles), Syowa (Antarctica) and Crozet Island. These 9 stations include 1 continental African station (i.e. Assekrem, Algeria), 1 maritime African station (i.e. Mahe Island, Seychelles) and 7 stations downwind of Africa.
(d) We also include the summary of MOZECH performance in the recent IPCC/ACCENT intercomparison experiment (Stevenson et al, 2006)
(e) Short summary of Rast et al. (in prep.) will be included following the anonymous

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referee major comment 2.
In summary, this detailed evaluation of the model shows that MOZECH overestimates dry season lower tropospheric (surface 800hPa) ozone concentration. The reason for this is that the model ozone maximum during the dry season occurs at the surface – 800hPa, while that of the measurement occurs higher up in the troposphere at 850 – 600hPa, leading to a high bias at the surface which 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 the dry-season overestimation of ozone concentration over Africa may be three-fold: (i) MOZAIC data may have missed the high surface to lower troposphere ozone enhancement due to biomass burning since airports are relatively cleaner. (ii) MOZECH may have underestimate dry deposition over continental Africa, and lastly (iii) lack of aerosol (especially mineral-dust) in MOZECH may have also contributed to this overestimation. However, to fully diagnose the cause of this problem will be beyond the scope of this paper. We have to point out that this problem will not effect the results of this study, since we are basically interested in differences of two simulations, but this overestimation has definitely led to an increase of the currently calculated 33.14 Tg(O_3) African tropospheric ozone burden as we pointed out in our response to major comment 4. We also have to point out that the seemingly "wrong lower tropospheric gradient" recorded over Johannesburg in our former July plot was due to an error in our extraction and interpolation algorithm. Actually MOZAIC has no measurements at the surface to around 850hPa levels over Johannesburg and Windhoek. We have corrected this mistake, and our new plots no longer show this "wrong tropospheric gradient". The surface CO concentration calculated by MOZECH shows a good agreement with CMDL stations data, with a slight underestimation at Barbados, Azores and Tenerife and an overestimation at Negev desert, Israel. Section 4.1 will be changed from "Model validation" to "Model evaluation".

4) In the conclusions the advances of this paper beyond Marufu et al. (2000) along with the points from that study which are supported should be summarized (this is

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mentioned in various places in the text, but it is important to place this in the literature by summarizing this in the conclusions).

Our paper investigates the sensitivity of regional and global tropospheric chemistry to African biomass burning, biogenic, lightning and anthropogenic emissions for the years 1997 – 2001. It provides a one-stop discussion on the role of African emissions in regional and global tropospheric chemistry. On the other hand, Marufu et al. 2000 focused on the determination of the source of tropospheric ozone over Africa, with a priority given to biomass burning emissions. Interestingly, our estimate of 9.52 Tg due to African biomass burning emissions is comparable to 10 Tg calculated by Marufu et al., 2000. Also MOZECH estimate of 33.14 Tg tropospheric ozone burden over Africa (using the 150 ppbv O_3 tropopause, see response to minor comment 9) is higher than 26.32 Tg calculated in TM3 model used in Marufu et al., 2000, perhaps due to dry season overestimation of lower tropospheric ozone in MOZECH. We will include this explanation in the conclusions.

5) Why is only biogenic VOC considered, and not biogenic NOx (soils) in the sensitivity run? Further, In Table 3 it appears that other sensitivity runs considering biogenic CO and NOx were indeed conducted, but not discussed properly in the paper (only alluded to in passing once in the results); this should either be incorporated fully into the discussion, or removed from the table.

We indeed conducted an experiment where all African biogenic emissions (biogenic VOC, soil NOx, CO, H2) were excluded. For consistency, we have replaced our former discussion of biogenic VOC influence with the evaluation and discussion of this experiment. However we will include the comment that the influence of biogenic emissions is mainly due to biogenic VOC emissions. As we have pointed out in Table 3, this replacement has made biogenic emissions the most important emission source

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influencing both African and global total tropospheric ozone burden.

6) Most of section 4.4 could be dropped in the interest of an improved analysis elsewhere; starting around p. 5812 it is mostly descriptive without much interpretation, and the sizes of the influences (tenths of Tg) are small enough that the seasonal variation in them is not really relevant compared to the other issues discussed in the paper; only the last paragraph of the section becomes more interesting, but it would need to be supported by a meteorological analysis (e.g., vector plots of wind patterns) to make a significant statement. For the rest of the section, there are indeed a few interesting points, but the summary that is given in the conclusions would be enough to bring these across well.

We will remove Figure 8 and its respective discussions (i.e. Pages 5812 (starting from line 14) and 5813). We will add plots of streamlines to show transport pathway outside of Africa.

7) The inclusion of interannual variability to the study is, as pointed out by the authors, an important advance over previous work. This section should be made more in proportion to its importance. In particular, characterizing the reason for the large interannual variability of the influence on southern Asia would be very interesting. I suspect this is related to shifts in the southward excursions of the ITCZ, and to the transport in plumes in the trade winds during the monsoon transition periods, so that this could be nicely related to the satellite observations discussed in Kunhikrishnan, T., M. G. Lawrence, R. von Kuhlmann, A. Richter, A. Ladstätter-Weißenmayer, and J. P. Burrows, Semi-annual NO2 Plumes during the Monsoon Transition periods over Central Indian Ocean, Geophys. Res. Lett., 31(8), doi: 10.1029/2003GL019269, 2004. Also, the last paragraph discussing the interannual variability in emissions is interesting, but given the results in the rest of the study, it would be useful to indicate

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how this might translate into effects on ozone.

The 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 (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 the third year (i.e. “1999”) and the fifth year (i.e. “2001”) of our simulation respectively, which causes the TOB over Southeast Asia to be about 18 – 34% lower and 24 – 31% higher than 5-year average TOB. Note that we have performed an AMIP2 run, therefore the specific years (i.e. 1999, 2001) may be arbitrary. 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. in “2001” (or cold i.e. in “1999”) anomaly induces a weakening (or strengthening) of the African easterly jet (references will be included), which increases (or reduces) the amount of CO and NOx transported from Africa eastwards (streamlines plots will be included).

Our study did confirm that plumes emanating from Africa could be found in Southeast Asia and central Indian Ocean (CIO). We also found that ozone due to African emissions over southeast Asia is located around 700hPa – 70hPa, thus it contributes an insignificant amount to surface ozone enhancement. However our study show different

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seasonality than those discussed in Kunhikrishnan et al, 2004 because we have focus on Southeast Asia and not on CIO in particular. This study shows a teleconnection of El Nino and La-Nina on transport from Africa to Southeast Asia and CIO.
The last paragraph on inter-annual variability of biogenic VOC emissions is indeed irrelevant to this present study because the same globally gridded monthly emissions were prescribed from year to year. We have therefore removed the discussion on the inter-annual variability of biogenic VOC emissions.

Response to the Minor comments:

1) In the abstract, it would be nice to give the relative amounts of African ozone which the changes due to each of the emissions represents (e.g., about 8% for biomass burning, etc.)

The percentage values will be included in the abstract.

2) Abstract: "about 70%" - I calculate exactly 80% for the values in the table

This will be corrected. We meant to say "more than 70%" and not "about 70%". We calculated the difference for each of the emissions categories separately; this also will be clarified in the final version.

3) Abstract: Rather than listing Russia and other countries (and missing many of the former Soviet states), it would be better to say "northern North America, northern Asia, and Europe" (here and elsewhere)

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Done

4) The choices of what to discuss in the introduction seem a bit scattered to me, e.g., why mention methanol but not other emissions (e.g., butane)? It would help to tighten down the introduction and really focus on what is relevant for Africa (and indicate why where possible).

We have deliberately mentioned isoprene and methanol emissions and their relative importance to tropospheric ozone concentration and burden as found by previous studies. We also found that these two species play an important role in the enhancement of African and global total tropospheric ozone. We will restructure the introduction and add more statements to strengthen the aim of the study.

5) P. 5801, l. 21-22 "is henceforth referred to as Rast et al. (2006)" is not needed, since this is just the normal citation with footnote.

Done

6) Section 2.4: has the lightning distribution been evaluated for these particular runs? (it can change a lot between different horizontal and vertical resolutions, for example in terms of land/sea contrast)

In order to remove resolution dependency, Grewe et al. 2001 parameterization employed in our model is tuned such that global lightning NOx emissions is about 2.7 Tg/yr. This is done for these simulations. A visual comparison of our lightning distribution with that of Lightning Imaging Sensor (LIS) data (available at

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http://thunder.msfc.nasa.gov/data/query/distributions.html) shows good agreement.

7) Section 4.2: "This corresponds with the relatively high contribution of these countrie" - how large is the contribution of each country? Is the relative effect on ozone (and other gases) disproportionately small or large (e.g., due to the concentration of emissions in one region)? This is an example of where deepening the analysis would help make it much more interesting.

Nigeria, South Africa and Egypt together contribute about 35%, 53% and 37% of total anthropogenic CO, NOx, NMHC emitted in Africa, respectively. This is proportional to about 56% contribution of these three countries to African tropospheric ozone budget due to African anthropogenic emissions. We will include this explanation in the final version.

8) Section 4.2: the last paragraph seems to be the opposite of the sentence on p. 5808, l. 21; these should be synchronized.

Done

9) Why assume the tropopause at 200 hPa? This is a bit oversimplified and it is straightforward to do it better (although it might not make too much difference, but with the effect of biogenic VOC being so strong in the TTL on the other hand it might).

We have redefined the tropopause to 150ppbv O_3 level as used in Stephenson 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.

10) Section 4.3: "Over the continental regions outside Africa, the African biogenic contribution to the ozone burden is two times that of the biomass burning" - why is this? Adding analysis of the precursors might give some insight.

See comments on major comment 1. We will add the precursor (NOx, CO) discussions.

11) P. 5811, l. 19-23; these are partially redundant with the previous paragraphs, should be merged.

Will be merged.

12) P. 5816, l. 7: Lawrence et al. (1995) gave a range of 1-8 Tg(N)/yr, not an upper limit of 20; if the citation is moved to after "too high", then the sense of the sentence is accurate; another recent study giving strong evidence of an upper limit nearer to 10 Tg(N)/yr is Labrador, L. J., R. von Kuhlmann, and M. G. Lawrence, The effects of lightning-produced NOx and its vertical distribution on atmospheric chemistry: sensitivity simulations with MATCH-MPIC, Atmos. Chem. Phys., 5, 1815-1834, 2005.

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The citation has been moved to after too high. This is what we meant to say.

13) Grammar (which is generally excellent): replace "at" with "in" anywhere that a region of the troposphere or a hemisphere (NH/SH) is mentioned, e.g., p 5800, l. 6, or p. 5808, l. 11

Done

14) P. 5801, l. 5: "conditions" (plural)

Done

15) P. 5816, l. 17, add "emissions" after "anthropogenic"

Done

16) Table 1 and Table 2 would be nicer merged into one table, with the relative contributions (in percent) of the African emissions to the global emissions being given.

Done

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