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Atmospheric
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Discussions

Interactive comment on "Emission sources contributing to tropospheric ozone over equatorial Africa during the summer monsoon" *by* I. Bouarar et al.

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We thank the reviewer for the comments which are addressed as follows:

General comment:

a) A bit surprising is the fact, that according to the authors soil NOx and biogenic emissions have similar impact on ozone at 240 hPa.

Both soil NOx and biogenic VOC emissions are influenced by convective uplift over West Africa and this could explain their similar impact on O3 in the UT which extends from West Africa to the Atlantic Ocean and central Africa as well. However, as shown

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in Figure 8 and 9, while the maximum O3 changes due to soil NOx occur at around 240 hPa, the highest changes due to biogenic VOCs occur at around 140 hPa.

1. p.13774, l.24 (also p13779, l.20, also. section 5.1): The effect of switching emissions off versus a sensitivity has been invested in Grewe et al., GMD, 2010. Note that the total sensitivity of ozone to a perturbation of the emission still does not cover the total effect compared to a full tagging scheme.

The relevant reference has been added.

2. p.13775, l.15-20. The model resolution of 2.5x3.75 (L19) is relatively coarse when looking at convection. How many levels are below 100 hPa and how many vertical levels represent the TTL or the UT, which is quite important for the later manuscript?

Details about the vertical resolution in LMDz_INCA have now been added in section 2. There are 13 levels below 100 hPa and 7-9 levels are located in the stratosphere. The TTL is represented by 2-3 levels. We agree that the actual standard resolution of LMDz_INCA is coarse when looking at mesoscale events like convection. In a recent model version, horizontal resolution has been somewhat improved (1.875x3.75). The LMDz_INCA model development team is currently working on a new version with 39 vertical levels.

3. p.13776, l.16: For Africa a special high resolution biomass burning emission data set is used. How does it compare with GFed in terms of global numbers? How does long range transport of GFed emissions from other parts of the world (in particular Asia) affect the results?

As recommended by the referee, we added Table 3 showing integrated biomass burning emissions totals for CO and NOx over Africa in the L3JRC and GFED inventories. The following text was added in section 2:

“Table 3 shows integrated biomass burning emissions totals for CO and NOx over Africa in the L3JRC and GFEDv2 inventories. These emissions are higher in the L3JRC inventory by around 47% and 61% respectively in 2006 (and 70% and 75% respectively during the summer (JJA) period) compared to GFEDv2". Concerning the CO and NOx biomass burning emissions over Asia, they represent 37% and 36%, respectively of the total Asian emissions in 2006 and 42% and 10% in JJA. In this study, we focused on the impact of long-range transport of all Asian emissions. The objective is to show that import of these emissions should be considered in studies of the chemical composition of the African troposphere. We agree that assessing the impact of long range transport of each emission category (in particular biomass burning) from Asia (or other parts of the world) would be interesting. This would require further investigation and model sensitivity studies and could be the subject of another paper.

4. p.13787/13788: The conclusion here is that the CO is overestimated due to an overestimate of JJA BB-emissions in the L3JRC data. How would a further reduced emission affect ozone, which is underestimated in Fig.4b)? To me it looks as if convection is too strong enhancing CO and reducing ozone compared to MOZAIC. I don't see the consistency of the conclusions from Figure 4a) and 4b). What's the role of transport from emissions other than biomass burning? What's the role of scavenging and potential long range transport of NOx-reservoir species from other source regions? Which process is less realistic in the model: dynamics(i.e. convection), chemistry, or the emission data set? How is the occurrence of lightning NOx coupled to the NO_CONV experiment? Does NO_CONV imply much less lightning and LNOX as well?

We agree with the referee that the section on comparison with MOZAIC CO and O3 data was not very clear and needed to be improved. It has been replaced by the following text:

"Switching off the convective transport in the Conv_off simulation leads to a significant decrease of CO in the UT confirming that the modelled CO distribution in the African UT is sensitive to convective uplift. BB emissions are an important source of CO in this region although they are not directly uplifted by convection over central Africa but are transported in the LT northward to the convective region over Sudan/Chad before

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being uplifted into the UT and redistributed southwards over central Africa by the large-scale Hadley circulation. The northward shift in simulated CO maximum in the UT compared to the observations appears to be due to the location of convection in the model. Analysis of meridional wind speed and vertical velocities from ECMWF and KE_AMMA in the UT showed strongest ascending winds in the model over West Africa and north of 5°S over central Africa. Moreover, Barret et al. (2010) showed that detrainment occurs over a larger meridional region in KE_AMMA simulation compared to other models. Comparisons with results from TI_AMMA are in better agreement with MOZAIC compared to KE_AMMA, due to weaker uplift into the UT, although simulated values are still higher compared to MOZAIC. The overestimation of UT CO in KE_AMMA could therefore be due to strong convective updrafts and detrainment in the Emanuel scheme leading to rapid uplift of CO from the LT and redistribution over a broad region. However, as shown by Barret et al. (2010), other global models based on the Tiedtke scheme (TM4 and p_TOMCAT) also tend to overestimate CO compared to MOZAIC. Another possibility is that biomass emissions are too high in the L3JRC dataset. Reducing the African BB emissions by 20% (i.e. -31 TgCO in BB_red) during the JJA period leads to better agreement with MOZAIC CO data. Therefore, the overestimation of UT CO could be due to an overestimation of BB emissions. Convective uplift may also be too strong over central Africa since it has only been possible to validate vertical profiles further north over West Africa where convective uplift of CO seems to be reasonably well simulated.

Low O3 concentrations (less than 50 ppbv) were observed by MOZAIC over the convective region between 0 and 15° N. The highest values occur north and south of this region as a result of O3 formation in the upper level branches of the Hadley circulation as already reported in previous studies (Sauvage et al., 2007c; Barret et al., 2010). The latitudinal distribution of O3 observed by MOZAIC is fairly well reproduced by the model simulations although the shape of O3 transect south of 5° N is not reproduced correctly. However, KE_AMMA underestimates O3 by up to 20 ppbv south of the Equator at around 10° S. Barret et al. (2010) showed that other models (except MOCAGE) also fail to capture the observed maximum and that KE_AMMA also underestimates O3 relative to MLS observations at around 10° S. As noted in the comparisons with MOZAIC CO, these discrepancies may be due to a combination of factors linked to convective uplift and emission sources. Modelled O3 in the UT over central Africa is rather insensitive to surface BB emissions with results from the BB_red run showing only small changes in UT O3 relative to KE_AMMA (1 ppbv decreases south of the Equator). Results from emission sensitivity runs (see section 5.4 and Fig. 9) confirm this low model sensitivity to central African BB emissions, despite significant convective uplift of these emissions. UT O3 is also sensitive to available NOx and so, it is possible that there is insufficient modelled photochemical production in BB air masses or too much production of HNO3, which is then washed out during convection leading to too little O3 in the UT. Modelled UT O3 is also rather insensitive to other surface emissions (BB, biogenic VOCs, soil NOx and anthropogenic sources (< 1 ppbv O3 changes)). On the other hand, model results are most sensitive to lightning NOx emissions (see section 5) which, following the analysis presented in the previous section, are underestimated. This direct injection of NOx leading to photochemical O3 production downwind is a significant source of O3 in the UT. Increasing lightning NOx production by 50% over Africa in the XLiNOx simulation during JJA leads to higher O3 concentrations (up to 6 ppbv) south of the Equator. The influence of lightning NOx, which is mainly produced over West Africa during the monsoon season, extends up to central Africa due to southward redistribution of such emissions in the UT by the Hadley circulation (see section 5.1 and Fig. 6). This is also confirmed by the Conv_off experiment, where both lightning NOx emissions and convection were switched off. In this run, the underestimation of UT O3 in LMDz_INCA is more pronounced compared to KE_AMMA.

Thus, while overestimation of convective uplift and BB emissions may explain the overestimation of MOZAIC CO data, a combination of weak lightning NOx emissions and weak photochemical production in uplifted (BB) air masses may explain the underestimation of MOZAIC O3. Enhanced washout of NOx reservoir species, linked to strong convection, may also impact the amount of NOx available for O3 production over downwind regions over central Africa in the model."

Answers are given in the text above for most of the referee's questions. Concerning the impact of further reduced BB emissions, the 20% test (BB_red) shows up to 1 ppbv O3 changes. Therefore, further reduced emissions will lead to an O3 change of around 5 ppbv. As discussed above, modeled central African UT O3 is more sensitive to long range transport of lightning NOx emissions compared to the impact of BB, biogenic and anthropogenic emissions (< 1 ppbv O3 changes in sensitivity runs). Concerning the processes which are the least realistic in the model, this study confirms that uncertainties in the emissions are still one of the common problems in chemistry transport models. However, improvements to the convection scheme and model resolution are also needed. An improved Emanuel scheme version is being tested in a new version of the model in order to improve the diurnal cycle of deep convection over Africa. This may lead to more realistic washout and lightning NOx in the model since these schemes are linked to model convection. However, as is the case with many global models, our analysis suggests that improvements to these schemes are needed as well as improvements to the model chemistry. Further analysis of these processes has been beyond the scope of the current study.

5. p.13789, l.25: Comparison is made with MOPITT data, which are carefully used applying the kernel to model data which have the correct overpassing time. Nonetheless the model overestimates the CO column compared to MOPITT. Which role plays the occurrence of clouds? Are the MOPITT data mainly based on cloud free conditions? If so, can this explain systematically lower CO in the observations (due to biased observations towards cloud free conditions)? Is any statement possible about the vertical CO distribution or the relation between model and MOPITT in the UT? Similar for NO2 - what's the role of clouds?

The overestimation of total column CO and tropospheric NO2 columns may be due to uncertainties in both emissions and satellite retrievals. Indeed, model runs using L3JRC emissions show higher CO columns compared with results using the GFEDv2

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inventory showing that model results are very sensitive to the BB inventory which is used. On the other hand, there are also uncertainties associated with satellite retrievals, including, as mentioned by the referee, the sensitivity to clouds. Both MOPITT and SCIAMACHY data are produced for clear-sky pixels (or a small cloud fraction). This can have an influence on the number of observations available for comparison with the model result, especially in winter or over cloudy regions. However, over Africa, the biomass burning emissions occur during the dry season, south of the Equator, over central Africa, far from the convective regions. Some data might also be missed when there is aerosol or smoke being mistaken as cloud but this depends on fire type and phase during the satellite overpass. This is difficult to account for in the case of SCIAMACHY since there is no information available about the vertical distribution of NO2 and also, the presence of aerosols and their type. Concerning MOPITT, even if no analysis has been carried out to date on the impact of clouds over polluted regions, it appears that the retrievals are not affected by smoke (personal communication from Louisa Emmons). Details about uncertainties related to satellite retrievals and the occurrence of clouds or aerosols have been added in section 4.3 as follows:

“Another possible explanation for the discrepancy between KE_AMMA and satellite column data is the fact that both MOPITT and SCIAMACHY retrievals are biased to clear sky scenes. Indeed, this reduces the number of observations available for comparison and thereby increases the uncertainty in the observations. Furthermore, the satellite data may underestimate CO and NO2 from fires if there is a lot of aerosol or clouds obscuring the fire regions. However, this is difficult to account for since there is no readily available information on the presence of aerosols, their distribution and type. Furthermore, the BB emissions occur over central Africa far from the cloudy convective regions associated with the summer monsoon. However, some data may be missed if aerosol or smoke is mistaken as cloud but this depends on fire type and phase during the satellite overpass.”

6. p.13792, l.10: Is it the transport of soil NOx or the transport of soil NOx induced

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ozone?

This is transport of both soil NOx and soil NOx induced ozone. Indeed, the soil NOx sensitivity test, presented in section 5.2, leads to changes in both ozone and NOx concentrations in the LT and UT. The text has been changed as follows in section 5.2:

"The significant influence of soil NOx emissions on O3 in the UT is due to upward transport of NOx-rich air masses and soil NOx induced O3 from the LT by deep convection. . ."

7. p.13797, l.6: add 'global' ozone change

The sentence was modified.

8. Technical: In general for the difference plots: Please indicate in the Figure captions the differences as e.g. Fig 8: KE_AMMA - BIO_red. It facilitates reading the differences.

Figure captions have been changed following the referee's recommendation.

9. Fig13. Wrong y-axis label or numbers.

The plot labels have been corrected.

Interactive comment on Atmos. Chem. Phys. Discuss., 11, 13769, 2011.