

## Response to anonymous referee #1

We wish to thank referee#1 for his/her comprehensive review of our manuscript. Modifications made to the paper as a result of the referee's suggestions and remarks had resulted in a significant revision of the manuscript.

The general impression of the referee is encouraging although we feel he/she has missed some novel and important factors, which we highlight here. Although studies have been performed using regional models to study the composition of the troposphere near Equatorial Africa (EA) during the West African Monsoon (WAM), we perform our simulations for the entire year at a global scale using a state-of-the-art chemical transport model taking into account both photochemical activity and transport into and out of the region (i.e. changing boundary conditions). Moreover, we compliment the multi-model comparisons that have already been performed in the literature (Williams et al, 2010) using the recently developed L3JRCv2 (AMMABB) biomass burning (BB) emission inventory (Lioussé et al, 2010) with simulations using the more established GFEDv2 8-day and monthly BB emission inventories. We show that the latitudinal position of maximal concentrations for both O<sub>3</sub> and CO over the tropical Atlantic Ocean is not especially influenced by the choice of BB emission inventory. By varying the temporal variability, vertical distribution and integrated emission flux we also show that although there are increases in both trace species of between ~5-10% in the outflow regions, such parameters are not dominant constraints towards capturing tropospheric composition over EA. Rather it is the meteorological dataset used to drive the CTM that introduces the largest constraint. Finally, the conclusions of the trajectory calculations presented here are relevant to any study associated with inter-hemispheric transport of pollutants around Africa and also those which use backward or forward trajectories to identify the origins of polluted air-masses using the ECMWF operational meteorology analysis without the use of additional chemical signatures.

We also disagree with the referee regarding the quantitative aspects of the paper as we make direct comparisons against a number of different unique measurement datasets in order to show how well a typical state-of-the-art CTM performs above Central Africa and EA. This is an accepted methodology used by the modeling community towards investigating potential short-comings. Moreover, we have addressed one of the main objectives of the modeling component stipulated in the EU-AMMA project towards assessing the short-comings of large-scale models for this particular region. We also feel that this study compliments previous studies for different years involving e.g. the analysis of multi-annual MOZAIC measurements in the same region.

The aim of the study is to investigate the sensitivity of large scale model simulations towards the description of biomass burning that is included in large-scale models and examine the subsequent effects on tropospheric composition in Equatorial Africa with regards to CO and O<sub>3</sub>. We subsequently modify the introduction in the revised manuscript to make this clear. The following text is now included:

*This study investigates the influence of BB activity in southern and Central Africa during JJA on the composition of the troposphere over EA for the WAM during 2006. Here we differentiate the effect that various modelling parameters (temporal variability, "effective" injection heights, model resolution and emission fluxes) used to describe BB in large-scale CTMs have on capturing both latitudinal and vertical variability for this tropical region. Moreover, by performing a set of trajectory studies around EA, we also differentiate the constraints placed on a global CTM by the quality of the meteorological dataset used to drive the model for the African region. Finally we show the effect of assimilating a more statistically robust set of measurements into the meteorological dataset for August 2006 on the origin of specific air-masses in EA for periods where enhancements of MT CO and O<sub>3</sub> were observed.*

We do not find the figures to be too complicated considering the number of sensitivity studies which are included. Although referee#1 complains about too many figures there is little advice as to what should be removed. Moreover, referee#2 expresses the need for more comparisons. Therefore we include additional figures to (i) provide an overview of the Africa Continent, (ii) make weekly comparisons with the MOZAIC data at various altitude levels to strength our conclusions regarding the sensitivity studies and (iii) introduce further comparisons with a composite of the AMMA measurements. In the revised

manuscript we also correct an error in Figs 1 and 2 (now Figs 2 and 3 in the revised manuscript), segregate Figure 3 (now Figs 4a and b in the revised manuscript) to improve visibility (and present percentage differences in the daily mean values) and remove two of the sensitivity studies from Figs 3 and 4 (now Figs 5 and 6 in the revised manuscript) to improve the visualization of the results and further aid the reader.

Next we answer the referees general comments:

*The use of the trajectory model feels a bit as a different part, which is not very well integrated in the paper. I have the impression that the trajectory analysis does not really fit in the paper; the model is also not described in the experimental setup. Shouldn't it have been better to test this new meteorological forcing in the CTM, rather than introducing a different tool (a trajectory model)?*

We now introduce a new section 2.3 (Trajectory model) where we provide a brief description of TRAJKS. We do not agree with the comment about the relevance of showing the results as the trajectory analysis allows us to differentiate whether the deficiencies are principally due to the parameterizations used in the CTM (e.g. convection, advection, chemistry) or related to the ECMWF meteorology used to drive the model. Without the use of the trajectory model we would not have been able to categorically show this, as the CTM is Eulerian and therefore does not follow individual air masses. At the time at which the study was performed the new meteorological datasets were not included the operational data which we use to drive our model, with the new measurements being assimilated in a special re-analysis. It is beyond the scope of this study to pre-process the meteorology at global scale for use in the CTM.

*In addition, the use of the trajectory model is not an "independent way" for me.*

Considering this is principally a modeling study then the two models can be considered to be entirely independent methods of investigating the same issue, where one is based on Lagrangian transport of air parcels and the other uses a Eulerian grid model.

*I have the impressions that you used a lot of things which were available (from other studies, in the model, ...), but maybe are not optimal for this study :  
Two different types of models*

The results compliment each other so we do not agree with this point. By using different modeling tools we strengthen our findings by being able to differentiate between transport, parameterizations for introducing BB into large-scale models and chemical effects.

*You used as base simulation FULL while the HIGH\_ANTH simulation as base simulations would have been better*

We performed this sensitivity study to investigate whether potential deficiencies in the anthropogenic emission inventory as a result of rapidly expanding urban centers could partly explain the low O<sub>3</sub> between 600-800hPa simulated in TM4\_AMMA. Our method was to use a space based estimate of the decadal growth in emissions for Cairo as a best guess across the entire continent, which does not necessarily improve the description of anthropogenic emissions throughout the continent but rather introduces a crude correction. The result was rather trivial ( $\pm 2\%$  differences in mixing ratios for e.g. O<sub>3</sub> in the lower layers above the most populated regions) therefore does not impact on our main findings and conclusions that we present here. To improve clarity we now remove the HIGH\_ANTH sensitivity study from the paper.

*Part of the reasoning is based on results which are not present: the impact of temporal resolution of the meteorological forcings and horizontal resolution (just by mentioning personal communications of TM5 results); trajectory calculations showing more transport to the equator are mentioned but not shown or proven*

For brevity we cannot show all results, especially when the differences between those that are shown are rather small. Showing many 'non-results' detracts from the more interesting findings. We still wish to include the general conclusions though to support our argument that the horizontal resolution used in the model did not significantly degrade to quality of the simulations i.e. it is not a major constraint on modeling the region. However, we remove all mention of the seasonal differences which are not related to JJA. We

do not wish to show additional trajectory calculations as the trajectory analysis is performed to differentiate the reasons as to why the CTM fails to capture the enhanced concentrations of CO and O3 in the middle troposphere on the specific dates selected for the comparisons (which compliments other papers in this ACP special issue).

*Emissions budgets and burdens in regions like "34N-34S" and over all longitudes are not very meaningful for this study*

Unfortunately no 3D chemical budget including all individual grid cells on the global domain are available in the CTM. However, it is well established in the literature that the long range transport of biomass burning plumes significantly impacts tropospheric composition in regions far away from the source regions therefore we do not see the benefits of focusing solely on the African domain, especially where integrating over a 3 month period and considering the Westerly transport out of the region above the tropical Atlantic. We focus on the tropics as this is the region where a large fraction of pollutants are oxidized (e.g) CH4 due to high photochemical activity. Finally, the benefits of a global model are that we do not have a restricted horizontal domain as with a regional model and therefore we want to take advantage of this.

*sampling at 650 hPa, at 670 hPa, ... : why not sampling all the time at the same altitude?*

These are the average pressure values calculated for each particular subset of data. By the nature of the model, a grid cell contains pressure values at both the top and bottom of the cell calculated using sigma hybrid co-ordinates, with the average being used throughout the grid cell. Thus values change with respect to latitude depending on changes in the surface pressure. Therefore, the average of all pressures along the 2D transect at a particular model level shown in the Hövmüller diagram is not necessarily the same as those from a single latitudinal transect.

*Figures 3, 4a and 4b : they are small, with too much details, and therefore ask a lot of energy to recognise what is described in the text; the mentioning of colour codes in the caption instead of on the plot is not good*

We now segregate Fig. 3. (now Figs 4a and b in the revised paper) to show larger Hövmüller plots of the FULL run CO and O3 distribution. Then we show the percentage differences for NOSAFR, NOGUIN, FULL\_8day and HIGH\_IH (now omitting the HIGH\_CO run). This will address the point regarding the lack of evidence for our conclusion regarding the magnitude of the differences in long range transport introduced by both temporal variability and effective injection height made by referee #1. We now modify Figures 4a and 4b (now Figures 5 and 6 in the revised paper) to include colour keys up the side of each figure to aid the reader, where the NOGUIN and HIGH\_IH simulations are removed from the plots. We also change the colour key to highlight the differences. The text has also been modified so each figure is introduced sequentially.

*Sometimes new topics arise in the middle of the text: p7516, line 20-22: weather 6-hourly data is sufficient*

We modify the introduction to include all relevant topics, mention the use of the trajectory model and include a section in the methods giving a brief description of the trajectory model.

*I do not find the titles of the sections very well chosen*

These are now modified according to the suggestions of both referee#1 and referee#2.

*I should not talk about "sensitivity studies" in the abstract (that is too technical too appear in the abstract)*

The abstract has now been modified and made more robust at the request of referee#2.

*The trajectory figures always show different parts of Africa, which does not increase the readers comfort.*

We do not feel that including a large part of Africa will improve the visibility of the individual trajectories by increasing the 'dead space'. Moreover, each diagram includes latitude and longitude values which the reader should be able to use to locate the area (western Equatorial Africa). The inclusion of an overview of the African continent in the new Figure 1 also places these trajectories in context.

The referees review is 14 pages long and therefore it is not feasible to reply to every single point. The majority of the suggestions regarding both grammar and wording have been adopted, although there remain certain points associated with the discussion that we retained as this appears to be a style issue.

Here we respond to the more specific questions:

*p7509, line 9: "temporal distribution" - isn't "temporal resolution" better?*

Both the monthly and 8-day GFEDv2 BB inventories are provided at a 1° x 1° global resolution and subsequently coarsened onto the working grid of the model, therefore there is no change in resolution between the inventories. Rather the update frequency is higher in the 8-day inventory which affects the emission flux per grid cell area (thus the distribution of the most intense burning events changes). We now use the term temporal variability throughout the revised paper to address this.

*p7509, line 10: "much more important": isn't this a bit too strong?*

“Much” is now removed.

*p7509, line 17 : "extreme" is some term for some specific phenomenon described in other papers; therefore, I would just write "very high"*

Thouret et al (2009) refer to measurement taken on the 14<sup>th</sup> August as an ‘extreme event’ therefore we retain this definition in the abstract and throughout the revised paper

*p7510, line 2-9: I miss a reason why it is worth studying these fires: is it a health or a climate impact? Which species?*

We add the following sentence regarding the motivation for the study:

*“The transport and chemical evolution of polluted plumes from BB containing high concentrations of e.g. CO, aerosols has been shown to affect the composition of the troposphere at both regional (e.g. \ Real et al., 2010) and global scales (e.g. de Laat et al, 2007), thus influencing both local air quality, visibility and the lifetimes of important greenhouse gases via perturbations in the oxidizing capacity of the troposphere.”*

*p7510, line 14-15: "such events" is too vague: are as well the BB as the natural wildfires assumed to increase?*

An increase in wildfires is linked to a drier climate rather than anthropogenic biomass burning which is more linked to population growth and political incentives such as laws against uncontrolled burning. We have re-written this section including a more robust argument for our statement at the request of referee#2.

*p7512, line 8-9: what is meant by "differences in the monthly variability"? That the mean in one month is very different of the mean in another month? Should "variability" not just be "distribution"?*

The term ‘monthly variability’ is now replaced with ‘seasonal variability’.

*p7512, line 23: shouldn't you give some short information on the model: chemical scheme (stratospheric/tropospheric chemistry), wet and dry removal parameterisations, convection, turbulence?*

All these details are comprehensively described in Williams et al (2010) which is referenced. For brevity we do not wish to include them here.

*p7513, line 7: "using a scaling ratio of 10": do you mean that you use values which are 10 times stronger than in the parameterisation of "Heymfield and McFarquar"?; or do you convert this two physical quantities with different units one into the other just by multiplying?*

We follow the approach given in Heymfield and McFarquar and multiply the Surface Area Density by factor of 10 to obtain the cross-sectional area. We now explicitly state this in the text.

*p7513, line 23-26: does this data set also contains emission data for 2006, or only up to 2000?*

We add an additional sentence:

*'Thus the anthropogenic emissions are for the year 2000 and the biomass burning emissions are for 2006, where any increase in the anthropogenic emissions in Africa is assumed to have a minimal effect on the results presented here'.*

*p7515, line 5-9: how can the coarsening be responsible for this? I don't understand how the interpolation process (which is linear) and for both data-sets (GFED monthly and GFED8-daily) should behave differently. "Cumulative sum" is not nice, it is saying twice the same.*

The 3°x2° emission flux is a cumulative sum (not interpolation) of the individual 1°x1° emission fluxes over the entire area of a grid cell rather than an area weighted average value. Given the variability in a typical month in the 8-day inventory (which is not linear) the integrated flux applied for one month can be different as a result of the update frequency in the 8-day emission inventory depending on the variability in each grid cell. We have ensured that the annual emission sums are identical between both of the monthly and 8-day emission inventories.

*p7515, line 20: "methodology" : a bit vague (I think you mean by which instrument ,how frequent sampling, wavelengths, etc ...)*

We now state: "... methodology (e.g. instrument, sampling frequency, etc) .... "

*p7515, line 26-30: the reason why you do this, is not well explained. I think you use the emissions for the year 2000 from RETRO (which you don't mention, but which I presume) and want to correct them for the assumed increase. The increase rate is not very well known, and you use an increase rate from one city (because nothing better is available). The aim of your paper is studying BB, not the background anthropogenic emissions. I understand that you want the best possible background, but choose than 1 of the 2. This sensitivity study is a bit out of the scope of this paper.*

The sensitivity study related to increasing anthropogenic emissions is now removed from the revised paper.

*p7516, line 20-22: does this analysis allows you to decide whether 6 hourly analysis are sufficient? I suggest that you have to correlate observations with model results to say whether this frequency is enough. Here, one gets the impressions that comparing O3 with tracer distributions undergo the same forcing can explain.*

*p7516, line 21- 22: "whether using 6-hourly updates of the meteorological fields is sufficient": a new topic arises; this should be mentioned in the introduction.*

We now state:" By showing the daily variability in CO and O3 with the respective passive tracers we examine the fluctuations in transport into the region when using 6-hourly updates of the meteorological fields." . The introduction has also been modified.

*p7517, line 11: why 34N-34S (due to your analysis method)? why looking at all longitudes? even looking at all the longitudes, it is not a closed system. So therefore I would suggest studying just a part of the Africa continent + part of the Atlantic ocean, or just the "cross section". By looking to all longitudes and expressing the results in percentage, these number don't tell a lot. Another option is to discuss absolute differences.*

We now adopt the absolute differences in the emissions as suggested by the referee.

*p7517, line 17-19: I don't know in what sense this is clarifying*

This sentence is now removed.

*p7517, line 21-24 : if I read this sentence, I interpret it as : From looking at the figure one should see that the BB extends (1) far inland reach 15\_N, and (2) well into the SAHEL (10-20N). Are you not saying twice the same?*

The text has been changed thus: *“It can be seen that the influence of BB from southern Africa extends far inland over West Africa reaching ~10-20°N affecting tropospheric composition far inland, as well as near the southern coast.”*

*p7517, line 29: why are FULL and FULL\_8day so different? Why giving numbers for 34N-34S and 0-34S, and not 0-34N and 0-34S?*

*p7517, line 28-29: shouldn't you expect that the monthly and 8 daily data sets should be coherent? How can one otherwise say that an impact is caused by the time resolution (while it may also be caused by different emission totals)? Please explain this more.*

The changes between the FULL and FULL\_8day are different due to the update frequency being applied in each of the BB emission inventories. Although the global annual total is the same, analysing the season JJA allows some differences to occur. It should be noted that changes in BB emissions in South America and parts of India and Indonesia are also included in the totals as a result of integrating across all longitudes (no 3D budget is available for the runs). That is the FULL\_8day emission inventory is applied globally so as to change the background and transport into the region. In the revised paper we now only present the absolute differences between 34°N-34°S to allow the direct comparison with those presented for the NOGUIN and NOSAFR runs. An additional sentence is added to further explain this: *“Even though the annual emission totals are the same, analysing specific seasons can result in differences as a result of the more rapid variability in the 8-day inventory for a given period. Moreover, these differences also include the variability in BB for South America, parts of India and Indonesia.”*

*p7518, line 27: can't you quantify this correlation*

The correlation changes along with the emission flux of CO from southern Africa for each month, where JD150-190 shows a much worse correlation than JD 191-240. We do not feel that providing one correlation co-efficient for JJA between the SAFR passive tracer and CO would provide any more insightful information than assessing the agreement by eye. We now mention this change in agreement between the different JD periods in the text.

*p7519, line 2-4: is the "NOGUIN AUG" correct: finding such differences for a source region where there is normally almost no emission?*

We thank the referee for pointing this out. An error occurred with the production of the diagram which has now been fixed in the revised paper. The NOGUIN simulation now has only small differences when compared to the FULL.

*p7519, line 9-10: "where higher (lower) concentrations are seen in the FULL run" : (I don't see this)]*

*p7519, line 8-10: this conclusion is much too strong*

Presenting the percentage differences in the daily mean mixing ratios of the various sensitivity simulations in Fig 3 (Fig 4a in the revised manuscript) now makes this statement more obvious and supports our conclusion. Also, removing the NOGUIN and HIGH\_IH results from Figs 4a and b (Figs 5 and 6 in the revised manuscript) support this statement. We also do not find the conclusion too strong given that the percentage differences shown as part of Fig 3 (Fig4b in the revised manuscript) aid the reader in assessing effects.

*p7519, line 13-15 : from Figure 3, I cannot see that*

Again, presenting the percentage differences in the daily mean mixing ratios strengthens this conclusion.

*p7521, line 12-16: can't you be more specific; there are a lot of possibilities left over*

In order to elucidate the importance of the potential factors contributing to this under prediction by TM4\_AMMA would involve more simulations and detract from the main focus of the study. The MOZAIC comparisons are principally shown to give the reader an indication that the model tends to underpredict CO near the source regions. Moreover, we do rule out the influence of model resolution and the update

frequency of the meteorology in the text (by mentioning a TM5 run performed for the same year using an identical emission inventory).

*p7523, line 19 - p7524, line 5 : what is new in this? your are only mentioning existing literature*

We agree with the referee. These details also appear in the introduction therefore we remove this text in the revised version.

*p7525, line 27 - page 7526, line 2 : any explanation?*

We now include the following explanation: “Examining the NOSAFR run at 8h50 shows that at high altitudes there is an additional source of O<sub>3</sub> compared to CO. This is most probably from long range transport by the AEJ in the MT as discussed in Sauvage et al (2007).”

*p7526, line 11-13 : I don't understand this; for me it is just that CO is mainly governed by regional sources, while this is not especially true for O<sub>3</sub>*

We now include an additional figure showing a comparison between a composite of the CO measurements taken above EA during July and August 2006 during the AMMA measurement campaign and co-located model output. We choose to show both the FULL and NOSAFR comparisons which categorically show that the over-estimation of CO throughout the lower and middle troposphere around 8-15°N is due to BB from southern Africa.

*p7526, line 24: "reproduce much of the large scale variability": isn't it rather the vertical gradient which it represents (it is not large variability, it is because the aircraft changes altitude). I have the impression that the NOSAFR vertical gradient is closest to the observed vertical gradient (see Cotonou profiles in Fig. 6), although there remains still a very large bias. And the gradient is maybe right, but the bias remains very large.*

We agree with the referee that the variability is somewhat governed by the height at which the measurement is taken. We modify the text accordingly in the revised version. Moreover, we also include two extra tables containing Pearsons correlation co-efficients for the lower and middle troposphere between CO measurements taken during July and August 2006 and co-located model output.

*p7527, line 10: these values of 2-4\_N have not appeared earlier in the manuscript, so it is a bit strange that they appear here after a "we have shown"*

All latitudes are shown in Figure 3 for the 2D transect at this altitude (now Figure 4a in the revised manuscript).

*p7527, line 22: does this trajectory model includes vertical diffusion and convection?*

The TRAJKS model contains neither diffusive nor convective processes. To make a first-order correction for pyrogenic convection we started the trajectories 1.5km above ground level. For more details of the trajectory model we refer the referee to Scheele et al. (1996). Some details have also been given in the new section introduced into the model description section.

*p7528, line 10-11: why do you conclude this: first, because fig9a keeps it pretty low? and second because fig 9b needs convection?*

The forward trajectories started between 15-20°E on the 4<sup>th</sup> August travel in the lower troposphere more westerly than the Cotonou launch site therefore do not predict transport into the middle troposphere around 2°E. The trajectories started between 25-30°E do not travel westward but remain in the lower troposphere above southern Africa before rising near the Equator, thus they are too far East to impact on the launch site.

*p7528, line 13-15: this "elevated" gives the impression that you talk about both days, while next you say that it was high in August the 14th but not on August 3rd*

The text has been modified in the revised paper.

*p7529, line 22-25: I would say that Fig.12a is not too different from Fig.10a. Only two trajectories have their origin over the southern Africa continent*

A further two trajectories in Fig 10a also have their origin in the Guinea region, which has little BB activity for August. Thus the new meteorological dataset has the potential to increase the influence of BB emissions at the launch site in the CTM.

*p7529, line 25-27: even if these trajectories pass a certain moment nearer the regions where the BB plumes are transported inland, if you follow the trajectories longer they come from over the ocean (similar to Fig 10b).*

But convective mixing above land can introduce additional pollutants after it has travelled from the ocean whilst the air mass circles near Cameroon.

*p7529, line 27-page 7530, line 1: this is very interesting, why don't you show it*

On reflection only one or two of the 25 forward trajectories from Central Africa enter the middle troposphere when using the improved meteorological dataset, and these impact more south than currently declared in the text. The text has been modified accordingly.

*p7530, line 3-4: why this conclusion about the limitation of trajectory studies?*

Trajectory calculations based on existing meteorological datasets have been used to ascribe likely source regions responsible for the enhanced CO and O<sub>3</sub> observations seen e.g. over the southern coast of West Africa (Andrés-Hernández et al., 2009). Our point is that when assimilating the additional measurements into the forecasting models the origin of air-masses around Equatorial Africa the results for 2006 were quite different (as exemplified by Figs 11 and 12) and the AEJ-S moves a few degrees northwards as a result. Therefore, use of trajectory calculations for EA for deriving the origin of air masses during the West African Monsoon should be treated with caution.

Figures and Tables:

The HIGH\_ANTH scenario is now removed from the paper. An introductory figure has now been included showing the African continent along with the geographical analysis regions and locations of the measurement sites as suggested by both of the referees. Colour keys are now provided for Figs 1 and 2 (now Figs 2 and 3). To improve Fig 3 (now Fig 4) we split the daily mean values in the 2D cross-section into a plot showing the variability for the FULL simulation and an associated plot showing the percentage differences between the FULL simulation and the other sensitivity studies. To improve on Fig 4 (now Figs 5 and 6) a colour key is given at the side of the plots and the NOGUIN and HIGH\_IH comparisons are removed. We introduce a further Figure showing a comparison of a composite of the CO measurements taken during the AMMA campaign with co-located model output for the FULL and NOSAFR simulations. We also provide two additional tables containing Pearson's correlation coefficients between the measurements of CO and co-located model output for the FULL, FULL\_8day, HIGH\_IH and NOSAFR simulations. The biomass burning region in Fig 9a (now Fig 11) is now removed. We re-phrase the individual plots in Figs 9-11 (now Figs 11-13) as top and bottom, rather than a, b and c. All comments regarding the headers and legends are addressed.

*Fig4a/b: Lower right panel: if this plot represents values over the 6-8N region, averaged 3W-6E, the mixing ratio for the Guinea tracer should be maybe closer to 100 ppt.*

We thank the referee for noticing this. Investigation reveals that there was an indexing error by one latitudinal position in the selection of both grid cells. This is now fixed in the revised paper.

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