Impact of biomass burning on pollutants surface concentrations in megacities of the Gulf of Guinea

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Dear Editor and reviewers,

We acknowledge the reviewers for the time spent to evaluate our work. We also acknowledge the Editor and we made all proposed changes in the revised manuscript. Please note that our answers are in blue in the text and after each reviewer's remark.

Best regards, Laurent MENUT December 21, 2017

1 Reviewer #1

Summary:

This paper presents modeling results from the DACCIWA project. Model results are performed for the period May - July 2014, with and without biomass burning emissions. The model results have been compared to a variety of observations. The model simulations appear to be of high quality, and the authors do compare the model to observations and not the weaknesses/strengths of the simulation.

Paragraphs were added in the revised version to better describe the weaknesses/strengths of the simulation.

However, the paper is tediously long and very un-focused. There are 15 Figures, many of which could be moved to SI. There are also randomly short sections (e.g. Section 7.1 and 7.2 which contain only a few sentences each). Many of the Figures are model output for specific days, but the logic behind the choice of day is hard to follow.

The article was reorganized and the English was completely checked. Some material was moved in several Appendices. Some subsections were reorganized to have more homogeneous sections. Text was also added to have better explanations about the choice of the days presented as examples. The selected days correspond to: (i) the end of the modelled period and (ii) the days where CALIOP data were available and interesting in term of biomass burning plumes. This was also added in the text.

Thus I can't recommend that this paper be accepted for publication in ACP as it currently is formatted. I see that there has been a class exercise devoted to reviewing this article, and they note several grammar issues.

Three comments were received during the discussion phase of this paper. They were all posted by students from the same class. They considered that it was an "exercise of review". But they are not reviewers. The ACP principle is to have (i) scientific reviews from professional, selected by the editor, or (ii) short comments. The short comments have to be "short" and "comments". The goal is to promote a dialog and improve the paper. And it was not the case with these comments. A long answer was written for the first one. The others were considered as not constructive and after discussion with the Editor, it was decided not to answer the second and third comments.

I also see quite a few grammar issues, but I have not pointed them out specifically because I think re-structuring is necessary. Here is my recommendation for re-structuring.

We acknowledge the Reviewer for these remarks. Following his/her recommendations, we simplified the article, with material in several Appendices, less figures in the main text and a Section devoted to the presentation of observations and a Section focusing on new model developments only .

Recommendations:

1. Begin with a single large map (similar to Figure 1) that shows the locations of the urban areas of interest, and biomass burning emissions during this period of time. Remove unnecessary figure clutter, and label the legend. This is currently not done in Figures 2 and 3. The current versions of Figure 2 and 3 can be omitted or moved to supplemental.

The Figures for the emissions are now in an Appendix, with the corresponding description of the model.

2. Separate the observations from the modeling. It does not makes sense to have sites with no measurements listed in Table 1.

We moved the description of the sites of interest that did not reflect actual measuring sites to the Table in the 'tracers' section.

3. Describe the essential components of the modeling in the methods, and move some of this information to SI. This section is currently 5 pages. The documentation is good for reproducibility, but can be moved to SI.

A large part of the model description is now in a Appendix.

4. Begin the paper by showing the observations, rather than the model results. Figure 9 would be a good place to start. This Figure shows large areas of high AOD corresponding to fire and dust emissions. The legend should be labeled (not with a green highlighted text, but rather next to the legend in plain English). Then I would move to describe Figure 10. It would be good to focus on specific events that are simulated well and those that are not simulated well. Highlight those events with colors. From here, the authors could present Figure 8, which shows the maps of surface tracer concentrations for a specific day (27 July). This Figure should be clearly linked to Figure 10 and potentially to Figure 11 and 12. Without this link, the choice of model output seems very random.

There was a logic to present the results as it was. First the meteorology, then the tracers, then the full simulation. CALIPSO data can be compared only at the end, after the full simulation. Nevertheless, we have decided to comply with the referees suggestion and we have restructured the paper accordingly.

5. Push most of the model validation to the SI.

Many figures and text are now in an Appendix.

6. End the discussion by noting the key points of Figure 14 and Figure 15. Please put the gas phase species in ppbv rather than ug/m3. It is unclear if Figure 15 is the amount of PM10 attributed to fires. The caption is confusing.

The caption was corrected. For the units, it is better to have concentrations in $\mu g m^{-3}$. ppbv is a unit mainly used in climate modelling, and related to the upper troposphere and lower stratosphere, since this is a unit designed to expressed very low quantities, with no dimension and independent on the air density (even though dependent on the altitude: 5 ppbv at altitude 5km is not the same amount of pollutants than 5 ppbv at 10km). For atmospheric pollution, as in this study, and in the boundary layer, the unit is $\mu g m^{-3}$. This unit is the one for all surface stations and of many research papers (see atmospheric composition papers in ACP or GMD).

Many parts of this paper were changed or moved at the end of the manuscript in several Appendices. We hope that, now, the structure of the paper and content of the paper are clearer to the reviewer.

2 Reviewer #2

Menut et al. present a comprehensive evaluation of the influence of biomass burning emissions in central Africa on atmospheric composition and air quality in cities around the Gulf of Guinea. They clearly show that their model represents the atmospheric state in good agreement with different observational datasets. Their evaluation of the model sensitivity to parameters related to the injection height of biomass burning emissions is very welcome and clearly explained. I recommend publication of this work in Atmospheric Chemistry and Physics subject to addressing the following comments.

Thanks a lot for these positive comments.

General comments

I recommend that the authors check through the manuscript for the consistent use of some expressions. In particular they exchange "Gulf of Guinea" with "Guinean Gulf" quite regularly and it would read more clearly if they chose one and use that. Also the terminology of the different experiments (e.g., NoFIRE and FIRES) is a bit mixed up through the manuscript after it has already been defined - they can use the experiment names without having to describe them again.

The acronyms and grammar was completely checked and corrected. For example, this is now "Gulf of Guinea" in the whole manuscript. A complete check was also done for the simulations names.

The manuscript touches on a couple of key issues in biomass burning and atmospheric composition on which some further comments or recommendations would be useful. Firstly, some concluding comment on injection heights for biomass burning emissions would be of interest, particularly on whether it improves the estimation or not.

About this point, the following paragraph was added in the revised version and in the conclusion: "In order to reduce the uncertainty in the simulation due to the way to inject biomass burning emissions in the atmosphere, two simulations were performed with different vertical profiles. It was shown that modelled results were not sensitive to the shape of the profile. The reason is that, during a fire, the pyroconvection induces a strong and fast mixing of the surface flux. Whatever the shape of the injection profile, the pollutants are finally quickly vertically mixed before a long-range transport."

Secondly, the CHIMERE model underestimates column CO compared with observations which is commonly seen across different models - can the authors make some comment on why this is the case

with CHIMERE? In particular the injection height of the fire emissions doesn't seem to make up the difference, could the OH field in the model be playing a role?

We agree this is an important question. In fact, we tested the injection profile shape and not the injection height. The injection height is parameterized fire per fire and using the [Sofiev et al., 2012] scheme. We consider this scheme to be validated and we used it without changes. About the CO underestimation, the following paragraph was added in the revised version: The differences between observations and model may be due to several factors: First, the boundary conditions used for the simulations are global and 'climatological' in model outputs. The transition from 'mean' values and this real test case may induce biases. For long-lived species such as CO, these biases may be transported inside the model domain. Secondly, underestimated CO may be due to overestimated OH or to an underestimate of the production of CO from the oxidation of VOCs. [Zeng et al., 2015] showed that this last process results in a large variability in model results. However, without complementary observations it remains difficult to disentangle different contributions.

I don't expect them to answer these questions fully but some comments in the context of the presented work would be a welcome addition.

All corrections presented below were done.

Specific comments

- Page 1, line 7-8: replace "to be" with "being". OK corrected.
- Page 2, line 2: replace "have evidenced" with "show". OK corrected.
- Page 2, line 27: "groud" should be "ground".

• Page 2, line 28: replace "Satellites data provide" with "Satellite data provides".

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OK corrected.
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• Page2, line 32: define the AERONET acronym - it is given in the caption for Table 1 but should be included in the text and removed from the Table caption.

OK corrected.

• Page 4, line 1: the authors mix up use of "modelled" and "modeled" - please check consistency throughout the manuscript (ideally using "modelled").

"Modelled" is the British spelling, "modeled" is the US spelling. So, it seems that the ideal is British. We agree and the word was changed accordingly.

• Page 5, line 19: "consist in" should be "consist of". Also check rest of manuscript.

OK done (three times).

• Page 5, line 27: it isn't clear what is meant by "monthly databases".

It means that HTAP propose maps of anthropogenic emissions mass on the basis of one month. Thus, there is 12 global maps available, one per month. Depending on the modelled period, we are using the maps corresponding to the month we are modelling. All this part is now in a Appendix, following the request of Reviewer #1. And more explanations were added in this Appendix about these emissions.

OK corrected.

• Page 5, line 32: the last sentence isn't very clear - are Abidjan and Lagos the only megacities? Or are there more?

The sentence was corrected. Abidjan and Lagos are two megacities, there is others along the coast.

- Page 6, Figure 2 caption: check the units are consistent with those given in the main text. There was an error in the caption and it was corrected. This is $(g m^{-2} day^{-1})$.
- Page 6, line 15: replace "were done" with "have been made".
- Page6, line 29: replaces "source" with "sources".

OK corrected.

• Page 7, line 10: "area burned" should be "burned area", "daily estimated" should be "estimated daily".

OK corrected.

• Page 7, line 11: clarify that daily refers to the daily emission.

OK corrected.

• Page 7, line 12: "CO for the month".

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OK corrected.
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- Page 7, line 14: clarify if "South-Africa" refers to SWA. Yes this is SWA and it is corrected.
- Page 8, line 16: "numerically cost consuming". OK corrected.
- Page 9, line 11: "fires" should be "fire".

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OK corrected.
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- Page 9, line 32: clarify if "vegetation emissions fluxes" refers to biogenic emissions or fire emissions. OK corrected. This is 'biomass burning emissions'
- Page 9, line 34: use "concentrations" rather than "content". OK corrected.
- Page 10, first paragraph: the explanation of the different experiments has already been given in the previous pages I found the explanation clearer on this page and it would benefit the reader to use just this one, linking to the TRC and FIRES experiments as already described.

OK the paragraph was merged with the explanations on the previous pages.

• Page 10, line 11: "information is". The details of the plots in Figure 1 are not necessary in the text and should be removed.

OK, it is right, this part is already in the caption and was removed.

• Page 10, lines 15 and 17: use "south" rather than "bottom"? OK corrected.

• Page 11, Figure 5 caption: not necessary to specify BADC as the source of observations - this is clear in the text.

OK corrected.

• Page 11, line 10: "fire emissions"?

OK corrected. This is 'biomass burning emissions'.

• Page 12, line 1: "dry convection in the lower troposphere".

OK corrected.

• Page 15, line 1: replace "concentrations" with smoke or pollution?

Here, this is the tracers experiment. This is not adapted to add smoke or pollution. This is just concentrations of a tracer.

• Page 15, line 2: replace "catched" with "caught".

OK corrected.

• Page 15, line 11: "continental Central Africa".

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OK corrected.
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• Page 15, Section 5.3: it isn't clear that this section is all that necessary as much of the summary of results has already been made in the preceding points.

This is not mandatory and we suppressed these intermediary conclusions.

• Page 16, line 1: "MODIS AOD product at wavelength of 500nm" - use of the symbol lambda is unnecessary, also using phi for latitude later in the manuscript.

OK corrected.

• Page 16, Figure 9 caption: should "CHIMERE without fires" be NoFIRE and "CHIMERE with vegetation fire emissions" be FIRES? Also it would be of great benefit to have each plot of the figure labelled so that a clearer explanation can be given in the caption - this also applies to the other figures.

OK titles were added in the Figure.

• Page 16, line 11: "fires emissions" should be "fire emissions" - please also change this throughout the manuscript.

OK corrected.

• Page 16, line 13: "shows that the model".

OK corrected.

• Page 16, lines 13 and 14: please quantify the terms "underestimated" and "over- estimated" - it isn't easy to tell from the Figure, and the addition of difference plots would be useful.

A quantification of the differences was added in the text. The difference plots would add a lot of Figures, when the Reviewer #1 considers there is already too many Figures. Here the only message is about the most important differences between the observations and the three simulations. For that, a quantification in the text is enough.

• Page 17, line 8: "BADC stations" - please clarify the source of the measurements (Met Office MIDAS land surface stations?) and not the data centre where they were obtained from.

Yes, this is "Met Office MIDAS land surface stations" as written. The sentence was simplified.

• Page 17, line 10: change "this" to "it", and "not modelled enough correctly" to "not modelled correctly".

OK corrected.

• Page 17, line 14: "biomass burning" (use either fire or biomass burning consistently.

The word "fires" was replaced by FIRE because we were talking about the simulation in this case.

• Page 17, line 20: what is meant by "not well retrieved"? does this refer to the satellite observations? Or how the model represents the atmospheric concentrations? "the Central Africa" should be "Central Africa", and "AODs" should be "AOD" (please change this throughout the manuscripts as well).

OK corrected.

• Page 17, lines 35 and 38: "FIREs" should be "FIRES" or "FIRE" - also throughout the rest of the manuscript.

OK corrected throughout the rest of the manuscript, including the Figures.

• Page 17, line 37: a comment on how difficult it is to reproduce the long-range transport over the ocean due to errors in the model transport would be useful.

The following sentence was added in the manuscript: "The low score in Ascension is related to the location of the site and the fact that the long range transport over the sea is difficult to reproduce: being less turbulent, there is less horizontal diffusion and vertical mixing. The plumes are thinner and more concentrated and the results are more sensitive to a possible model error on the wind direction. The comparison to observations located at one single point over the sea is thus often less correlated than comparisons over land."

• Page 18, line 8: replace "retrieved" with "represented"?

Yes, sure. This was done.

• Page 18, line 14-15: suggest changing to "the hypothesis that the optical properties of the modelled aerosols, and the estimation of the extinction".

OK, corrected.

• Page 19, Figure 10 caption: clarify which column is which - in the text it is described but should also be here in the caption. Labelling the plots will be helpful.

OK, the caption is now: "Comparison of AERONET measurements and model results for the AOD (left) and Angstrom exponent (right). Time series are presented for the Cinzana and Lope stations and for the whole modelled period."

• Page 19, line 6: "expressed" should be "expresses".

OK, corrected.

- Page 19, line 8: "while high values". OK, corrected.
- Page 19, line 11: change "finest particules" to "finer particles". OK, corrected.

• Page 20, Figure 11 caption: "carbon monoxide", no need to redefine the different experiments (i.e., remove "without" and "and with biomass burning").

OK, corrected.

• Page 20, line 6: "consists of three-day averaged".

OK, corrected.

• Page 21, line 4-5: the description of the three model simulation types is not necessary.

OK, corrected.

• Page 21, line 8: clarify that these are CO columns rather than concentrations.

The new sentence is now: "The IASI data show the increase of vertically integrated CO concentrations over Central Africa..."

• Page 20, line 13: change "where are located the studied cities" to "where the studied cities are located".

OK, corrected.

• Page 20, line 14: "has" should be "have".

OK, corrected.

• Page 20, section 6.4, line 6: "colocated" should be "collocated".

OK, corrected.

• Page 20, section 6.4, line 16: remove "correctly" - as described in the manuscript, the model does not 100% correspond to the observations.

OK, corrected.

• Page 21, line 1: the first sentence isn't very clear - is the author referring to modelling the vertical aerosol profile for comparison against CALIOP? If this is the case it can be stated more simply.

The new sentence is: "The CALIOP lidar measurements, on-board the CALIPSO satellite [Winker et al., 2010], are analyzed to obtain an aerosol sub-type classification (CALIOP v4.10 product), as proposed in [Omar et al., 2010] and [Burton et al., 2015]."

• Page 23, line 1: "thresholds of optical characteristics"?

OK, corrected.

• Page 23, line 4: "not being modelled".

OK, corrected.

• Page 24, line 24: change "retrieved" to "reproduced".

OK, corrected.

- Page 24, line 27: "bit" should be "but". OK, corrected.
- Page 25, line 11: "enables us to have". OK, corrected.
- Page 25, line 15: "For the fire emissions". OK, corrected.

- Page 25, line 25: "tree" should be "three"; change "both produced by" to "produced by both". OK, corrected.
- Page 25, section 7.1, line 6: "no pollution peaks".

OK, corrected.

• Page 26, line 1: "a pollution alert"?

A pollution alert is when the concentrations is larger than predefined thresholds. But, in the context of this study, this is not mandatory to speak about that and it was removed.

• Page 26, Figure 14: it would be useful, in the context of pollution alerts to relate the reported CO, O3 and PM10 results to the WHO and/or EU recommended exposure thresholds. Add plot labels.

The plot labels are already present and correspond to the location (top), the aerosol type (legend) and the plotted variables (left). The abscissa corresponds to the time, but it is mentioned "Time series" in the caption, and the x-axis is explicitly with days. The text about 'pollution alerts' was removed here, because the conversion of 'pollutants concentrations' and 'threshold and exposure' is a particular context and devotes probably a specific study.

• Section 7.2: This section would be of great interest if there were any corroborating observations, which does not seem to have been the case here, or if the authors could provide references to where aerosol composition from the model has been compared against observations. In its current form this section seems to be a bit of an unnecessary addition and distracts from the main message of the rest of the manuscript and could be removed.

We think this section is of interest <u>because</u> there is no measurement. In general, observations for surface concentrations are limited to $PM_{2.5}$ and PM_{10} . Some rare measurements are done for chemical species in Europe, but this is very limited and mainly done during specific neasurements campaign. This is also rare to have the information with the models, because it is necessary to have a chemistry-transport model integrating all aerosol types, thus all sources and chemistry. This is the case with the CHIMERE model and this is why we consider this Figure is an interesting added value for this study, since we want to quantify how much and what aerosol are involved in the biomass burning emissions. This aerosol composition was already studied with CHIMERE in [Menut et al., 2016] and this reference was added in the text.

• Page 28, line 12: change "pollutants surface concentrations" to "surface concentrations of pollutants".

OK, corrected.

References

- [Burton et al., 2015] Burton, S. P., Hair, J. W., Kahnert, M., Ferrare, R. A., Hostetler, C. A., Cook, A. L., Harper, D. B., Berkoff, T. A., Seaman, S. T., Collins, J. E., Fenn, M. A., and Rogers, R. R. (2015). Observations of the spectral dependence of linear particle depolarization ratio of aerosols using nasa langley airborne high spectral resolution lidar. *Atmospheric Chemistry and Physics*, 15(23):13453–13473.
- [Menut et al., 2016] Menut, L., Siour, G., Mailler, S., Couvidat, F., and Bessagnet, B. (2016). Observations and regional modeling of aerosol optical properties, speciation and size distribution over northern africa and western europe. *Atmospheric Chemistry and Physics*, 16(20):12961–12982.
- [Omar et al., 2010] Omar, A., Winker, D. M., Vaughan, M. A., Hu, Y., Trepte, C. R., Ferrare, R. A., Lee, K.-P., Hostetler, C. A., Kittaka, C., Rogers, R. R., Kuehn, R. E., and Liu, Z. (2010). The calipso automated aerosol classification and lidar ratio selection algorithm. *Journal of Atmospheric and Oceanic Technology*, 26(10):1994–2014.
- [Sofiev et al., 2012] Sofiev, M., Ermakova, T., and Vankevich, R. (2012). Evaluation of the smoke-injection height from wild-land fires using remote-sensing data. Atmospheric Chemistry and Physics, 12(4):1995–2006.

- [Winker et al., 2010] Winker, D., Pelon, J., Coakley Jr., J. A., Ackerman, S. A., Charlson, R. J., Colarco, P. R., Flamant, P., Fu, Q., Hoff, R. M., Kittaka, C., Kubar, T. L., Le Treut, H., McCormick, M. P., Megie, G., Poole, L., Powell, K., Trepte, C., Vaughan, M. A., and Wielicki, B. A. (2010). The calipso mission: A global 3d view of aerosols and clouds. Bulletin of the American Meteorological Society, 91(9):1211–1229.
- [Zeng et al., 2015] Zeng, G., Williams, J. E., Fisher, J. A., Emmons, L. K., Jones, N. B., Morgenstern, O., Robinson, J., Smale, D., Paton-Walsh, C., and Griffith, D. W. T. (2015). Multi-model simulation of co and hcho in the southern hemisphere: comparison with observations and impact of biogenic emissions. *Atmospheric Chemistry and Physics*, 15(13):7217–7245.