

Dear Anne Swank,

Thanks a lot for your comments on our work. I have to say it was a surprise to have this comment! When ACP was created, the novelty (compared to other peer-reviewed journals) was to have comments apart from 'official' reviews. But if you are looking at the numerous papers currently submitted, you can see that the counter is often 0. So, this is a good thing to have this additional comment and we are happy and proud that our work, only submitted (and thus open to improvements), may be used for courses. Of course, we consider this is just a comment and I don't consider the fact that you have a judgment of what has to be published or not. You will find here some answers to your most important questions.

Anne Swank text (in italic):

"The study of Menut et al. quantifies the relative contribution of biomass burning emissions in Central and South Africa on the surface concentrations of CO, O₃ and PM₁₀ in urbanized areas in Southern West Africa. This is done with CHIMERE model simulations of biomass burnings and comparison with satellite and ground measurements data. Several (tracer) model simulations are performed and show that the biomass burning emissions do indeed have an impact on the surface concentrations in urban areas in Southern West Africa. This study contributes significantly to scientific research since it includes the air pollutants in the previously studied air masses transport in Africa, which is an important attribute since (anthropogenic) air pollutants are increasing and have an impact on human's health. The simulation of the model are well tough of and are compared properly to available data. Hence this study fits to the reader's interest of Atmospheric Chemistry and Physics. However, the answer on the main research question about the relative contribution of biomass burning emissions cannot be found in this paper, which can be attributed to the absence of background information and clear explanation between the connection of observed and simulated air pollution values. These quantified relations are sometimes missing and some figures could be shown in another way. Therefore I recommend some major changes in this paper, at least to answer the main research question and on some other aspects, prior to publishing this paper in ACP."

As you write, the main topic of the paper is to quantify the "relative" contribution of biomass burning and this is done in the paper. During this period and over the studied region, there was no available measurements data (except those used in the paper): compared to studies done in Europe, China or South-Americ, there is no air quality network and no surface measurements of pollutants such as ozone, nitrogen oxides. Thus, we consider that the model is able to simulate the background concentrations, knowing that it was used over several other regions, with the same amount of information (HTAP anthropogenic emissions, Apiflame biomass burning emissions, Megan model biogenic emissions etc.). See for example the recent publications of [Marecal et al., 2015], [Real et al., 2015], [Menut et al., 2015], [Mallet et al., 2016], [Bessagnet et al., 2016], [Menut et al., 2016], [Vivanco et al., 2017]. These references are reported at the end of this answer. But, this is right, we have to make the hypothesis that, in absence on local surface measurements, the background concentrations have the correct order of magnitude and, thus, we can consider that the differences between the two simulations (without and with biomass burning emissions) also have a correct order of magnitude.

Anne Swank text (in italic):

Major arguments: The paper does not describe clearly how the biomass burning emissions are contributing to the total air pollution in the simulations and observed concentrations, where the research question is how the emissions contribute relatively. The results of the CHIMERE model simulations as illustrated in Figure 14, clearly with the simulation differences, show the maximum increase in surface concentrations of CO, O₃ and PM₁₀ of about 150 $\mu\text{g m}^{-3}$, 10 to 20 $\mu\text{g m}^{-3}$ and 5 $\mu\text{g m}^{-3}$ respectively. Concluded from these values is that the exceedance of pollution alerts will not be influenced hereby, but the impact on human exposure is not negligible. In the study of Adon

et al. (2016) the concentration of O₃ in West African urban environments ranges from 5.5 to 7.7 ppb (equal to µg/m³), which indicates that an increase of 5 µg/m³ would have an influence. From the study of Antonel & Chowdhury (2014) the PM₁₀ concentration are in range from 60 to 140 µg/m³. I believe that the increase in PM₁₀ due to biomass burning will therefore not have a large influence on the pollution alert. But, from my view it is necessary to include these reference (background) values of air pollutants in the urban areas in SWA, if necessary include observations from previous WAM years, and concentrations when the air pollution alert will be exceeded. The same accounts for the air pollution concentrations which have an impact on human exposure, when certain air pollutants do have a large impact more attention could be paid to these in the study, serving the purpose of the DACCIWA project. With the reference values included the relative contribution of biomass burning emissions on air pollution and its impact can be determined.

As you say, the quantification is presented in Figure 14 and in the text. The main question of the article has thus its answer. And there is not in our text the "total air pollution" but "pollutants". This is not the same thing. We are giving an answer for ozone, CO and PM₁₀, three of the main pollutants, but, of course, not all pollutants. The model takes into account all possible emissions (and this is not the case of many regional models currently used in the community). The differences presented at the end of the paper are thus really representing the additional amount due to fires, in addition to all other sources.

About the background values, even if we are studying the same SWA region, this is not reasonable to compare different areas, different periods. The meteorology and the biomass emissions are very changing with time, from day to day and the pollutants plumes may be very different from a country to another one. So, this is not correct to directly use previous studies done for other period or locations and use the values to directly conclude something. About exceedances, there is periods where surface concentrations exceed threshold, but we can not extrapolate past values on our period.

Anne Swank text (in italic):

"In the explanation for the TRC experiment in paragraph 3.5 is stated that the tracer emission flux is injected constantly without including the diurnal cycle, which produces the results as shown in Figure 7 and 8. However, in the study of Parker et al. (2005) is stated that the diurnal cycle has implications on the mixing of trace gases and aerosols between the surface layer and free troposphere. The vertical mixing occurs to be most efficient at night, which indicates that for the tracer experiment the diurnal cycle should be taken into account. This is confirmed by the study of Gilge et al. (2010), which indicates that an increase in the vertical mixing within the free troposphere could influence the air pollution levels in the lower free troposphere, with implications to the boundary layer. As consequence different vertical mixing profiles due to the diurnal cycle will probably have an influence on the tracer emission fluxes and transport. However, in section 3.4 of this study is indicated that the two different vertical mixing profiles provide the same results for the transport of the tracer and that only the profile of PR2 will be discussed. Wouldn't the transport be affected by different vertical mixing profiles when the tracer emission experiences a diurnal cycle? It might be good to discuss this in the article with results of the tracer experiment including a diurnal cycle for the two different vertical profiles."

You probably missed this part in the paper: "The fluxes being daily estimated, a diurnal profile is applied where 30% of the daily is redistributed during the night (18:00 to 8:00 LT-local time) and 70% during the day, close to values usually chosen in biomass burning model studies, (Zhang et al., 2012)."

Biomass burning emissions are estimated using satellite data. But this is not a geostationary satellite and there are no measurements every hour. We thus have to make hypothesis about the diurnal cycle. Even if our hypothesis is not perfect, this corresponds to the state of the art for these emissions.

Anne Swank text (in italic):

"The simulations in this study to quantify the biomass burning emissions are performed with the CHIMERE model. In the article is stated that comparison between satellite data, ground observations and simulations of the CHIMERE model show that the output model results are robust. This is illustrated for instance as time series in Figure 11 for the CO concentrations. What I miss in this article is why the air pollution concentrations are simulated with the use of the CHIMERE instead of another air chemistry model. In the study which Solazzo et al. performed in 2017, but with the CHIMERE model version of 2013, is stated that the CHIMERE model as its performance is studied in Europe, not always simulates the air pollutant concentration correctly. When the ozone lateral boundaries are changed, a shift is visible in the ozone diurnal cycle of the CHIMERE model with significant impact, which could indicate a flaw in the PBL dynamics. A positive bias in the ozone concentration simulated by CHIMERE is also concluded from the study of van Loon et al. (2017). Besides, the error of NO₂ impacts influence the ozone error significantly. These problems do not seem to be solved as I read from the article of Mailler et al. (2017) which describes the 2017 updates of CHIMERE. Due to the flaws in the model on ozone specifically I am not convinced that CHIMERE is the model to use in this study at this moment. An explanation about the simulated ozone concentrations or the relation between the ozone simulations and observations would help me to understand the choice to use CHIMERE. Besides, including some (necessary) correlations between observations and CHIMERE simulations for at least O₃, aerosol subtype and CO would convince the reader of the robustness of the model, similar as Table 2 and 3 for AOD and PM₁₀, next to only the quantitative differences in Figure 11 and 13 for instance."

There are several reasons to use CHIMERE. Some are subjective, the others objective. CHIMERE is a research model and, as all models, it is not perfect. The first subjective point is we are the developers of the model, therefore we are using it. The studies you are citing (Solazzo, Van Loon) were done by users of the model (there is about 300 persons using the model we are developing at the lab). A part of our work is to improve the model constantly and offer to the scientific community a new version each year. The second subjective point is that, being the developers, we can test parameterizations, schemes and modifying the code as we want. The objective points is that we are confident in our results. This is proved by many other studies you are not citing: see the results in the references cited at the end of this letter. You can also see the CHIMERE web page with all publications and you will see that for a large part of studies, CHIMERE is robust for the ozone concentrations modeling. And this is for this reason that the model is used for forecast in many Air Quality Networks in Europe, for the French official Air Quality Forecast called PREVAIR and is one of the 8 models selected to run daily the air quality forecast in the framework of CAMS Copernicus.

Another important point: there is no "good" or "not good" model. A chemistry-transport model is the sum of many processes (meteorology, emissions mixing, deposition) and all existing regional models have strengths and weaknesses... But knowing the status of the other available models, we are not sure to have better results.

Anne Swank text (in italic):

"Minor arguments: The conclusion from this paper is that the increase in air pollutants is mainly related to PMM and POM, indicating biomass burning. This is stated in part 7.2 of results, however I cannot find (neither in literature) whether this statement can be related to a real life scenario. Quantitative argumentation and references should be included on this matter."

You can read the paper explaining how we calculate the biomass burning emissions : the Apiflame model. It was also developed in our research team and you will see the emitted model species, PPM and POM, are some of these species. The reference to Apiflame is already in the paper.

Anne Swank text (in italic):

« In chapter 3 is mentioned that CHIMERE reads (WRF and) the surface emissions to simulate the chemical concentration, it is not described how these are obtained or can I assume that these are obtained by MODIS data? A brief explanation would be adequate. »

No, the emissions can not be obtained from satellite measurements. An emission flux and an atmospheric concentration (even if it is close to the surface) are completely different : between the two, you have the mixing, the chemistry, the deposition. All explanations about the emissions are provided in the section 3.2, with many details in (Menut et al., 2013) about all these processes. For the anthropogenic emissions, explanations and references are at the end of section 3.2, for mineral dust emissions, this is the section 3.3 and for the biomass burning emissions, the section 3.4.

Anne Swank text (in italic):

"In the article the output of the simulation of the CHIMERE model in the urban areas are quantified. However, it is not stated clearly how the values are obtained. Are these point measurements or averages over the whole urban area?"

Yes, you are right, we can add some lines to better explain how we select the location to compare model and observations. This is a good point and we will revise the manuscript with this suggestion. The principle is as follows : when you have a surface stations (such as MIDAS or AERONET), you have the exact location in longitude and latitude. The model has an horizontal resolution of 60km in this study. We use the four model points around the location and we calculate the corresponding concentrations using a bilinear interpolation. This is the same for the cities : but, in this case, having no specific location, we are using the center of the city to know what grid cell to use for our interpolation.

Anne Swank text (in italic):

"It is a bit unclear to me, whether all fires for the PR2 simulations are estimated on the same Hp or are estimated per individual fire (as described in paragraph 3.4 page 9)? I believe that the magnitude of fires differs and results in different Hp which are influencing the transport of air particles. The explanation could be improved."

With this line, you can see that the Hp value is calculated for each fire.

"The calculation of Sofiev et al. (2012) is based on the Convective Available Potential Energy estimation, itself diagnosed using the Fire Radiative Power (FRP) of each fire."

and:

"Hp is estimated, for each individual fire, as"

Laurent Menut
November 6, 2017.

Additional references:

Marécal V., V.-H. Peuch, C. Andersson, S. Andersson, J. Arteta, M. Beekmann, A. Benedictow, R. Bergstrom, B. Bessagnet, A. Cansado, F. Chéroux, A. Colette, A. Coman, R. L. Curier, H. A. C. Denier van der Gon, A. Drouin, H. Elbern, E. Emili, R. J. Engelen, H. J. Eskes, G. Foret, E. Friese, M. Gauss, C. Giannaros, M. Joly, E. Jaumouillé, B. Josse, N. Kadygrov, J. W. Kaiser, K. Krajsek, J. Kuenen, U. Kumar, N. Liora, E. Lopez, L. Malherbe, I. Martinez, D. Melas, F. Meleux, L. Menut, P. Moinat, T. Morales, J. Parmentier, A. Piacentini, M. Plu, A. Poupkou, S. Queguiner, L. Robertson, L. Rouil, M. Schaap, A. Segers, M. Sofiev, M. Thomas, R. Timmermans, A. Valdebenito, P. van Velthoven, R. van Versendaal, J. Vira, and A. Ung, A regional air quality forecasting system over Europe: the MACC-II daily ensemble production, *Geosci. Model Dev.*, 8, 2777-2813, 2015, www.geosci-model-dev.net/8/2777/2015/, doi:10.5194/gmd-8-2777-2015

Rea G, S.Turquety, L.Menut, R.Briant, S.Mailler and G.Siour, 2015, Source contributions to summertime aerosols in the Euro-Mediterranean region, *Atmos. Chem. Phys. Discuss.*, 15, 8191-8242, 2015, *Atmos. Chem. Phys.*, 15, 8013-8036, doi:10.5194/acp-15-8013-2015

Menut L., G.Rea, S.Mailler, D.Khvorostyanov, S.Turquety, 2015, Aerosol forecast over the Mediterranean area during July 2013 (ADRIMED/CHARMEX), *Atmos. Chem. Phys.*, 15, 7897-7911

Menut L., S.Mailler, G.Siour, B.Bessagnet, S.Turquety, G.Rea, R.Briant, M.Mallet, J.Sciare and P.Formenti, 2015, Ozone and aerosols tropospheric concentrations variability analyzed using the ADRIMED measurements and the WRF-CHIMERE models, *Atmos. Chem. Phys.*, 15, 6159-6182, doi:10.5194/acp-15-6159-2015

Mallet, M., Dulac, F., Formenti, P., Nabat, P., Sciare, J., Roberts, G., Pelon, J., Ancellet, G., Tanré, D., Parol, F., Denjean, C., Brogniez, G., di Sarra, A., Alados-Arboledas, L., Arndt, J., Auriol, F., Blarel, L., Bourriane, T., Chazette, P., Chevaillier, S., Claeys, M., D'Anna, B., Derimian, Y., Desboeufs, K., Di Iorio, T., Doussin, J.-F., Durand, P., Féron, A., Freney, E., Gaimoz, C., Goloub, P., Gomez-Amo, J. L., Granados-Munoz, M. J., Grand, N., Hamonou, E., Jankowiak, I., Jeannot, M., Léon, J.-F., Maillé, M., Mailler, S., Meloni, D., Menut, L., Momboisse, G., Nicolas, J., Podvin, T., Pont, V., Rea, G., Renard, J.-B., Roblou, L., Schepanski, K., Schwarzenboeck, A., Sellegri, K., Sicard, M., Solmon, F., Somot, S., Torres, B., Totems, J., Triquet, S., Verdier, N., Verwaerde, C., Waquet, F., Wenger, J., and Zapf, P.: Overview of the Chemistry-Aerosol Mediterranean Experiment/Aerosol Direct Radiative Forcing on the Mediterranean Climate (ChArMEx/ADRIMED) summer 2013 campaign, *Atmos. Chem. Phys.*, 16, 455-504, doi:10.5194/acp-16-455-2016, 2016

Mailler S., L. Menut, A. G. Di Sarra, S. Becagli, T. Di Iorio, B. Bessagnet, R. Briant, P. Formenti, J.F. Doussin, J. L. Gomez-Amo, M. Mallet, G. Rea, G. Siour, D. M. Sferlazzo, R. Traversi, R. Udisti, and S. Turquety, 2015, On the radiative impact of aerosols on photolysis rates: comparison of simulations and observations in the Lampedusa island during the CharMEx/ADRIMED campaign, *Atmos. Chem. Phys.*, 16, 1219-1244, doi:10.5194/acp-16-1219-2016

Menut L., G.Siour, S.Mailler, F.Couvidat and B.Bessagnet, 2016, Observations and regional modeling of aerosol optical properties, speciation and size distribution over Northern Africa and western Europe, *Atmos. Chem. Phys.*, 6, 12961-12982

Vivanco Marta G., Bertrand Bessagnet; Kees Cuvelier; Mark R Theobald; Svetlana Tsyro; Armin Aulinger; Johannes Bieser; Giuseppe Calori; Giancarlo Ciarelli; Astrid Manders; Mihaela Mircea; Sebnem Aksoyoglu; Gino Briganti; Augustin Colette; Florian COUVIDAT; Andrea Cappelletti; Massimo D'Isidoro; Richard Kranenburg; Frederik Meleux; Laurent Menut; Maria Teresa Pay; Guido Pirovano; Laurence Rouil; Camilo Silibello; Philippe Thunis; Anthony Ung, Joint analysis of deposition fluxes and atmospheric concentrations predicted by six chemistry transport models in the frame of the EURODELTAIII project, 2017, *Atmospheric Environment*, 151, pp.152-175, <http://dx.doi.org/10.1016/j.atmosenv.2016.11.042>