

Review of Biomass burning aerosol over the Amazon: analysis of aircraft, surface and satellite observations using a global aerosol model

Carly L. Reddington et al.

Summary:

Biomass burning is a major source of particulate matter pollution, regionally and globally. This has important implications for air quality and climate. Over South America, intense fires occur in August-September typically, providing the dominating source of atmospheric aerosols to the region. Yet considerable uncertainties in the magnitude of fire emissions remain. As such, the paper focuses on 'improving understanding of aerosol emissions from vegetation fires' by considering three different fire emission datasets (namely GFED4.1, GFAS1.2 and FINN1.5) to account for fire emissions in their modelling work. The authors used a global aerosol model (GLOMAP) to study how the simulated particulate matter (PM) concentration and aerosol optical thickness (AOT) are affected by the three different fire emission inventories. These results are compared against a comprehensive set of surface, aircraft and satellite observations collected over the Amazon region during September 2012. The authors have highlighted the spatial and temporal variation in the three different fire emissions and how it affects simulated quantities. Overall, the authors conclude that GLOMAP has skill in predicting reasonable surface concentration and vertical profile of PM over South America despite noticeable differences between the emission inventories. However, GLOMAP simulated AOT is found to be systematically underestimated. The authors therefore recommend caution when evaluating global models using AOTs to constrain particulate emissions from fires.

General comments:

The work presented here shares a lot of its DNA with a previous publication from the same group (i.e. Reddington et al., 2016). In this previous effort, the authors used the same modelling framework to argue that GLOMAP showed better agreement with observed PM mass concentration compared to AOT, potentially suggesting that some of the discrepancy between top-down and bottom-up studies may be connected to the calculation of AOT. In the present work, the authors test this hypothesis further by: *i*) providing a much more detailed evaluation of the GLOMAP model simulations against a comprehensive set of observations collected over South America during the SAMBBA campaign, and *ii*) performing a model sensitivity analysis exploring the assumptions related to the calculation of AOT in GLOMAP.

The paper is well structured and reads easily. The model evaluation is rigorous and convincing. The figures are clear and illustrate the points made in the manuscript. This paper is interesting and has a good potential. The last section of the result (Sect. 3.5) however feels too rushed in its current form and could benefit from the support of more visual material (i.e. show some plots for these results). The results in this section are mentioned too briefly, and do not provide a critical interpretation that would ensure more generally applicable results that could be subsequently transferred to other atmospheric aerosol models. The paper is quite weighted towards model evaluation. A model evaluation is only really useful if it used to interpret observed relationships or processes. As such, section 3.5 fails to convincingly demonstrate the assumption tested in this study.

The diversity in fire emissions highlighted here has important implication for aerosol modelling over this region, and likely in any region influenced by biomass burning. Contrasting the uncertainties from emissions with the uncertainties related to AOT calculation could really improve the scientific strength of the work. I would suggest reworking section 3.5, and perhaps add a discussion section before final publication in ACP.

Specific comments:

- P2, L2 – The authors state “*Our aim is to better quantify particulate emissions from fires over the Amazon basin*”. I would argue that the quantification of the emissions is down to

the groups developing these inventories. Rather, the current paper is investigating how different emission datasets affect modelled quantities (e.g. PM, AOT) and evaluate these outputs against a comprehensive set of data collected during the SAMBBA campaign.

- Could you describe the overall methodology behind the GFED, GFAS and FINN products? It would be nice to briefly discuss their strengths and weaknesses which could be used to further support/discuss your modelling results. Were there significant changes between the emissions used in this study (i.e. GFED4, GFAS1.2 and FINN1.5) and those used in Reddington *et al.* 2016 (i.e. GFED3, GFAS1 and FINN1)?
- The Table S1 listing the different optical properties tested is useful and would probably be better located in the main manuscript.
- Are there measurements of aerosol optical properties from the SAMBBA campaign that could be used to further challenge the hypothesis used by GLOMAP in the calculation of AOT?
- Some of the refractive indices listed in Table S1 are derived from Aeronet inversions. Aeronet only provides a bulk column refractive index and cannot artificially separate aerosols into BC and OC components. How do you integrate these values into GLOMAP? Do you apply the same refractive indices for BC and OC when considering the retrieved indices from Aeronet? How does it affect the aerosol absorption regionally? It would be interesting to link that to the different OC/BC ratio from the 3 inventories. Could the simulated Absorption AOT (AAOT) be evaluated against some existing observations then?
- On a similar note, was there any evidence of enhanced absorption from brown carbon during the SAMBBA campaign?
- The sensitivity of AOT to hygroscopic growth constitutes a large uncertainty. It would be useful to show the hygroscopic growth curve response for the two representations considered in this study. The Kohler curve seems to be much more sensitive at higher RH than the GLOMAP parameterisation (e.g. Johnson *et al.*, 2016). Nonetheless, simulated AOT with GLOMAP is much reduced when considering the Kohler model. Could it be due to a lack of representation of subgrid RH in the coarse resolution model? This may be something worth discussing in the model resolution section. In term of meteorological conditions (i.e. RH), was the year 2012 representative of previous years, otherwise could that have an impact on the AOT biases?
- The authors cite the results from Brito *et al.* 2014 indicating that the OA:CO ratios in biomass burning plumes during the SAMBBA campaign suggests limited secondary organic aerosol formation from Amazon fires. The comparison between GLOMAP size distributions and aircraft measurements seems to indicate an underestimation in the Aitken mode. If this is not related to secondary aerosol formation do you have an idea about what causes the discrepancy? Could it be related to the model assumptions on the size distribution parameters and could that have an impact on the calculation of AOT (e.g. moving the accumulation to smaller sizes)?
- It would be good to discuss uncertainties related to the emissions, the measurements and AOT calculation at the end before attributing the modelled AOT underestimation to the way it is calculated. Each measurement technique has its own uncertainties which may vary significantly depending on the observables. In addition, there is additional error that could be related to the sample size and the representativeness of local observations when compared against very coarse model grid-boxes (e.g. Schutgens *et al.*, 2016-2017).
- Please add the definition of NMBF in the main manuscript and explain how to interpretive it. Would the NMBFs listed in Table S3 benefit from being represented graphically to get a better idea of the model skill? (e.g. Figures 3 in Bender *et al.* 2018).
- Page 5, L3 – Figures S2 to S5 are referred before Figure S2 (at L15), reorder.
- Page 5, L29 – Replace Figure 3 by Figure 2. Same at Page6, L9/

- Page 8, L15-17 – “Sulfate concentrations are well reproduced by the model with no fire emissions and are overestimated when fire emissions are included. This suggests that either emissions of sulfate from fires are overestimated or that other sources of sulfate are overestimated in the model”. I’m struggling to see that from Fig 4b. Is it based on absolute numbers? How does the MACCity compares with say CMIP6 inventories for anthropogenic emissions? It could be nice to show how the different emissions (anthropogenic, dust, BVOCs, ...) contribute to the AOT over this region.
- Page 8, L25 – “likely due to a deeper BL over grassland ...”. Is it confirmed by looking at the model boundary layer height diagnostic? Please define BL acronym also.
- Figure 3 – Use a different symbol in the legend for ‘observations’ or a different colour so it stands out from the colour used for ‘Model (GFED)’.
- Figure 5, L8 – Please state explicitly what STP (i.e. standard temperature & pressure) stands for.
- Figure 8, L9 – change purple with grey in the legend.
- A section on data availability and code availability is necessary to comply with ACP requirements https://www.atmospheric-chemistry-and-physics.net/for_authors/manuscript_preparation.html (see manuscript composition).

References:

Bender, F.A.M., Frey, L., McCoy, D.T. et al.: Assessment of aerosol–cloud–radiation correlations in satellite observations, climate models and reanalysis, *Clim Dyn*, <https://doi.org/10.1007/s00382-018-4384-z>, 2018.

Johnson, B. T., Haywood, J. M., Langridge, J. M., Darbyshire, E., Morgan, W. T., Szpek, K., Brooke, J. K., Marengo, F., Coe, H., Artaxo, P., Longo, K. M., Mulcahy, J. P., Mann, G. W., Dalvi, M., and Bellouin, N.: Evaluation of biomass burning aerosols in the HadGEM3 climate model with observations from the SAMBBA field campaign, *Atmos. Chem. Phys.*, 16, 14657-14685, <https://doi.org/10.5194/acp-16-14657-2016>, 2016.

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