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Interactive comment on "Analysis of particulate emissions from tropical biomass burning using a global aerosol model and long-term surface observations" *by* C. L. Reddington et al.

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General comments

The paper evaluates multi-annual (2003-2011) monthly mean values of near-surface aerosol mass concentration (PM2.5) and aerosol optical depth (AOD) simulated by the GLOMAP global aerosol model against corresponding measurements conducted at four ground stations in the Amazon region and at 27 AERONET stations located in tropical regions worldwide. The simulations were done with three different datasets of biomass burning emissions (GFED3, FINN1, and GFAS1). Additional numerical experiments involved scaling the biomass burning emissions by a factor of 1.5 or 3.4. The model performance is evaluated in terms of the Pearson correlation coefficient and the normalized mean bias factor (NMBF). It is found that the model considerably

underestimates both PM2.5 concentrations and AOD, with a greater underestimation of AOD than PM2.5. The paper is well written and the presentation quality is good. However, the scientific significance and the overall scientific quality of the study are questionable.

Indeed, the fact that models tend to underestimate AOD over regions affected by fires (including the Amazon region) is well known. This is acknowledged by the authors: some earlier studies reporting the underestimation of AOD are mentioned in the paper, although the list of such studies is certainly not complete (see also, e.g., Petrenko et al., 2012; Konovalov et al., 2014). The use of ground measurements of surface aerosol mass concentration together with AOD measurements is a relatively novel point. However, the parallel analysis of PM2.5 and AOD measurements, as well as the use of three different emission datasets also did not help to fully explain the mismatch between the model and measurements of AOD. Note that Petrenko et al. (2012) has presented a much more extensive analysis of the impact of biomass burning emission uncertainties on simulated AOD values.

A serious drawback of the analysis is that it is based on an obsolete / simplistic understanding of organic aerosol processes. In particular, the authors disregard the well established facts that organic aerosol (which is a major fraction of biomass burning aerosol) is formed by organic compounds featuring a broad distribution of volatilities (see, e.g., May et al., 2013) and that a part of them, while in the gas phase, can provide a major source of secondary organic aerosol (SOA) (see, e.g., Grieshop et al., 2009; Hennigan et al., 2011) as a result of oxidation processes. Meanwhile, these facts have direct implications for biases in biomass burning aerosol emission inventories and for the mismatch between simulations and measurements of AOD (Jathar et al., 2014; Konovalov et al., 2015; Shrivastava et al., 2015). Although it is briefly mentioned that the SOA formation in biomass burning plumes can contribute to the difference between the simulations and observations, any quantitative estimates of such a contribution are not provided. A study could benefit from simulations of biomass burning aerosol by

using the volatility basis set framework and available parameterizations (e.g., Hodzic at al., 2010; Shrivastava et al., 2013, 2015). In any case, simplifications made in the model in regard to organic aerosol processes (such as an implicit assumption that organic part of biomass burning aerosol consist only of non-volatile material) and their implications for the results of this analysis had to be carefully described and discussed in light of earlier findings from relevant laboratory, field and modeling studies.

Specific comments

1. Page 6. The PM2.5 measurements made by the gravimetric filter analysis method that is known to be associated with large uncertainties (Malm et al., 2011). The authors estimate uncertainties of such measurements to be 15%. But how was the loss of organic aerosol mass due to desorption estimated? Available volatility distributions of fresh biomass burning emissions (e.g., May et al., 2013) imply that the loss of the organic aerosol mass from samples taken inside biomass burning plumes (POM$\sim$1000 ug/m3) after equilibration to ambient conditions (POM$\sim$10 ug/m3) could be as large as 40 percent.

2. Page 9, line 12. The fire emissions were injected into the model by using a set of fixed ecosystem-dependent altitudes. Meanwhile, it is known that the injection height depends on the fire intensity. If, for example, the injection height for major fires was underestimated in the model, the surface PM2.5 concentration during fire seasons could be overestimated. The study could benefit from using one of more realistic parameterizations of the injection height (e.g., Sofiev et al., 2012; Paugam et al., 2016). And, anyway, it would be important to ensure by means of a sensitivity analysis that the discrepancies between the results obtained with PM2.5 and AOD measurements are not due to biases in the injection height. The adequacy of the injection heights could further be evaluated by using surface measurements of CO concentrations and satellite observations of CO columns (see, e.g., Konovalov et al., 2014).

3. Page 10, line 13. "The water uptake for each soluble aerosol component is cal-

culated on-line in the model according to ZSR theory". Was hygroscopicity of organic components of biomass burning aerosol taken into account in the simulations? If so, what were typical values of the hygroscopic growth factor for the organic fraction?

4. Section 3.3. The paper could significantly benefit from an analysis of inter-annual variability of fire emissions and of corresponding PM2.5/AOD values in the Amazon region during the fire season. Has such a variability been predicted by the different inventories consistently? Can the model reproduce the observed inter-annual variability in PM2.5? Which of the inventories considered does enable the best agreement between the inter-annual variations in the simulations and measurements of PM2.5?

5. Page 15, line 17. "This suggests that the negative model bias in the dry season is largely due to uncertainty in the biomass burning emissions rather than anthropogenic emissions, SOA or microphysical processes in the model." Please see above a general comment about the potential importance of SOA formation in biomass burning plumes.

6. Page 20, line 20. "Uncertainties exist in the calculation of AOD that may contribute to the negative bias in simulated AOD." Did the authors try to validate their AOD calculations with other independent data? For example, it would be interesting to see if the model calculations are consistent with available measurements of the mass scattering and absorbing efficiencies (e.g. Reid et al., 2005). A bias in these parameters would indicate a similar bias in the AOD calculations.

Minor comments

1. Page 11, line 17. Daily GFED3 fire emissions were implemented in GLOMAP for the period 2003–2011, with monthly emissions implemented for the period 1997–2002. Were simulations analyzed in this paper really extended to the period 1997–2002?

2. Several papers cited in the text (Chin et al., 2009; Randerson et al., 2012; Zhou et al. 2002 . . .) are missing in the references.

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