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Discussions

Interactive comment on "Impact of mixing state and hygroscopicity on CCN activity of biomass burning aerosol in Amazonia" *by* Madeleine Sánchez Gácita et al.

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Response to referee #1

We thank the referee #1 for the careful review of the manuscript and for providing helpful comments on how it could be improved. General comments of referee #1 on the article's form are accepted and will be considered in a manuscript to be submitted for reconsideration. Replies to specific comments and questions raised can be found below.

"Study on nearly the same subject done by Roberts et al (JGR, Vol 108, 2003 doi:10.1029/2001JD000985) is not used and referenced at all and it can provide good

observational and modelling basis for the sensitivity study in current manuscript, especially with respect to uncertainty, variability and error analysis."

Roberts el at (2003) should indeed be referenced as an important precedent to this work and this will be corrected in the manuscript to be submitted for reconsideration. In particular, it was interesting to note that Roberts et al found also discussed the effect of kinetic limitations, reporting a reduction in the droplet concentrations of up to 35% for the dry season and updrafts of 0.1 ms-1, when comparing with the value estimated assuming equilibrium Köhler theory. Our results for low hygroscopicity values agree reasonably with this estimation, considering the differences between both studies (specially, size distribution and the representation of the aerosol), even when we found the overestimation to be much larger (up to $\sim$100% for internal mixings and up to $\sim$250% for external ones) for larger values of hygroscopicity. We thank referee #1 for pointing this out since it will certainly enrich the discussion.

"Detail comments: Chapters 2.1 -2.3 covers summary of basic textbook equations reported in numerous publications in past. I suggest to move these chapters to Appendix or Supplementary material and reduce it with proper references to paragraph or two in paper itself. Chapter 3 should be reduced significantly. It is not aim of this paper to make an overview of the past experiments. Data from each experiment used in this study can be properly referenced and briefly described in one paragraph. Chapter 3.1 is irrelevant for this study and should be removed completely. Chapter 3.2 should be significantly reduced and combined with paragraphs describing individual experiments, which provided observational basis for this study."

The manuscript to be submitted for reconsideration will be modified accordingly.

"P1L23: why original reference to Köhler paper from 1936 is not included?"

This will be corrected in the in the manuscript to be submitted for reconsideration.

"P15 L24-26: underestimation with respect to what? External mixing state? P16 L1:

overestimation with respect to: : :.?"

In both Ext1 and Ext2 situations, it is assumed that the aerosol particles are externally mixed. Therefore, the external mixing is the reference case. Assuming internal mixing typically leaded to an underestimation of the maximum supersaturation reached, and to an overestimation of the aerosol activated fraction. The sections when this is not clearly specified will be corrected.

"P16 L15-25: How close to reality are selected externally and internally mixed fractions? It is not clear to me if it is based on observational evidence or just assumed for test purposes."

This specific case was selected to illustrate graphically the impact of mixing state. Observational data for Amazon biomass burning is better described by the Ext1 externally mixed population, and the impact of mixing state in Ext1 was much lower than what is showed in figure 5, with average overestimations below 6% (P17 L8-18).

Response to referee #2

We thank the referee #2 for the careful review of the manuscript and for providing helpful comments on how it could be improved. General comments of referee #2 on the article's form are accepted and will be considered in a manuscript to be submitted for reconsideration. Replies to specific comments and questions raised can be found below.

"Furthermore, I would expect the manuscript to provide some recommendation for how the findings may be able to inform the treatments in regional coupled models, general circulation models or earth system models, given the diversity of representations of size and composition resolved aerosol and parameterisations of droplet activation. Some model treatments (e.g. the M7, GLOMAP or MOSAIC aerosol variants with Abdul-Razzak and Ghan, Fountoukis and Nenes or Barahona et al. activation parameterisations) are reasonably close to being able to capture the effects mentioned in the

paper and do not make such coarse approximations as the base case assumptions, so it is not clear which models will have problems of the magnitude identified."

We agree with referee #2 in that models are able to capture the effects of hygroscopicity and internal/external mixing state. Most of them also can consider to some degree the impact of kinetic limitations, although variants of Abdul-Razzak and Ghan parameterizations are widely used and do not consider these effects. The choice of to use two separate aerosol populations to account for the externally mixing character of the biomass burning population will increase the computational burden of the model. The modeler might choose instead to consider biomass burning aerosols as only one population internally mixed and externally mixed with other aerosol populations, unless given sufficient evidence that the overestimation derived from this choice is significant (and, in the case of amazon biomass burning aerosols, it seems that it is not). In a similar way, most global models or regional models over a large domain can allow for the specification of the aerosol hygroscopicity for different regions, but it is much simpler to choose a single value for all biomass burning. The choice of a parameterization that accounts for kinetic limitations, typically more demanding in terms of computational resources, needs to be similarly justified. Thus, our work did not aim to suggest improvements of the parameterizations themselves, but rather to guide the modeler choices. This topic will be further explored in the manuscript to be resubmitted.

"Indeed it is unclear whether such a scale of uncertainty is significant given the other sub-grid difficulties such as representation of updraughts."

We agree with referee #2 in that there are another number of factors that also increase the level of uncertainties. Yet, to improve the representation of the aerosol processes in GCMs is of great importance to adequately simulate aerosol-cloud interactions and their impact in the climatic system. In this case, the suggestions for the modeling of biomass burning aerosols that arrive from our work are, for the most part, easy to implement, without requiring improvements in the existing parameterizations.

“Figure 1 is unnecessary to the paper, providing a bit of background context and motivation that can be found elsewhere. At most it is supplementary material or appropriate for an appendix. If it were to remain, I would expect a model sensitivity study to look at the sensitivity of precipitation to mixing state. This would need a much more sophisticated model than used in the current paper.

"Sections 2.1 to 2.3 do not present any new approaches and can be replaced by a much shorter section, relegating the rest to the Appendix or to supplementary material or simply referenced.”

The manuscript to be submitted for reconsideration will be modified accordingly.

Specific points:

“i) there can be a strong sensitivity of predicted droplet number to the initial conditions, in particular the height at which an aerosol population is assumed to be in equilibrium with the ambient RH. Table 5 states that the parcel is initiated at 98% RH. Presumably the aerosol populations are assumed to be at equilibrium here. This RH is very close to cloudbase. A mixture of different hygroscopicity of particles will have very different masses of associated water and may have competed for available water more or less successfully already by this stage and may not be at their equilibrium size, dependent on the number of particles in the population. The dependence on initialisation conditions (80, 85, 90, 95, 98, 99% RH, for example) for different updraughts and size distributions may be particularly important for externally-mixed populations. The authors need to demonstrate that 98% is a justifiable initialisation for the entire range of updraughts and particle distributions in their study.”

We thank referee #2 for raising this concern, and will discuss briefly this choice in the article to be submitted. We found that the influence of the initial relative humidity was very low. To illustrate this, the maximum supersaturation and aerosol activated fraction are shown in Figure 1 for a range of initial values of the relative humidity. The values were calculated for the three size distributions considered, considering the externally

mixed case Ext2 with a population average hygroscopicity of 0.10 and three values of updraft velocities, including the minimum and maximum values considered. We found only a weak dependence (differences between maximum supersaturations obtained initializing at 80% and at 99% below 0.03) of maximum supersaturations with the initial relative humidity for the highest updraft values, and a negligible effect in the activated fraction.

“ii) the surface tension of water dependence on temperature may be of some modest importance as Christensen and Petters claim. However, the current manuscript completely ignores the very extensive literature on the roles of surface tension and bulk-to-surface partitioning that has been backwards and forwards in the literature since 1999. This is particularly relevant for particles heavily dominated by the organic components present during biomass burning. The authors need to justify ignoring any discussion or treatment of this, particularly given the recent claims of the pendulum swinging back towards an extremely strong enhancement of activation of organic-rich particles.“

This is an interesting point, and there is, as referee#2 points out, extensive literature on the topic including laboratory data specific for biomass burning that suggest this could be indeed an important issue (Fors et al., 2010; Giordano et al., 2013). However, it was not within the proposed scope of the submitted manuscript to approach this question, considering both the complexity of the biomass burning particles aerosol particles in terms of organic composition, and the scarcity of data to estimate this effects using, for instance, the methodology proposed by (Petters and Kreidenweis, 2013). We will acknowledge this limitation of the study within the article.

“p7 line 11, it is incorrect to state that "McFiggans et al. (2006) proposed sensitivities of the drop number concentration (CCN)..." and then state equation 7. They did propose the method to state sensitivities, but did so with cloud droplet number (N_d). Clearly CCN are not droplets. This sentence can simply be rephrased, but the implications of the underlying understanding of the problem are worrying.”

We thank referee #2 for noting this. The expression will be rephrased.

Caption of Figure 1. Maximum supersaturation (top) and fraction of particles activated as CCN (bottom), as function of the initial relative humidity, for the MP5,1 (solid line, squares), MP1,5 (dashed line, circles), and HP5,5 (dotted line, triangles) size distributions and external mixing case Ext2 with a population average k_p=0.10. Values refer to updraft velocities W=0.1 m s-1 (red), W=3 m s-1 (green) and W=0.1 m s-1 (blue).

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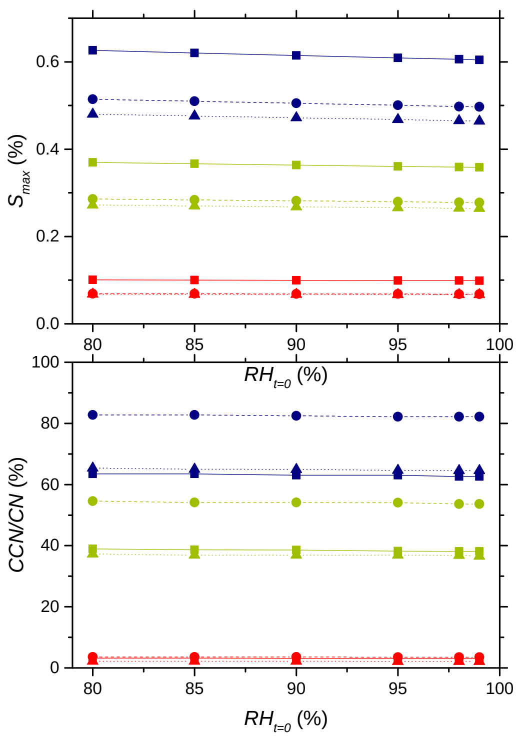

Fig. 1.