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Atmospheric Chemistry and Physics
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.

Anonymous Referee #2

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The paper presents a sensitivity study of parcel-model simulated droplet number to mixing-state and potential kinetic limitation to droplet growth. The study is extensively stated as being motivated by a number of S. American biomass burning experiments and the model parameter range inspired, though not tightly constrained, by observations from such experiments. I think there are some interesting results that are potentially worthy of publication - the unimportance of kinetic limitation and the roles that assumptions of mixing state and of kappa based on non-Amazonian observations are of note. However, I don't think they are done very good justice by the current form of the paper, which I found rather meandering and over-long. I found the abstract to be too discursive and not terribly useful in not providing a quantitative precis of the key

findings to make it easy for a reader to digest the study. I similarly found the main messages were buried in long discursive sections throughout the paper. A more tightly focussed presentation of the key results, conclusions and recommendations is needed to bring out the novelty and significance of the study. There are also a few places where the paper seems to be imprecise and appear to convey a lack of understanding that should be clarified (see specific points below). I won't try to rewrite the paper, but sections 3 and 4 are very long and could conceivably be replaced by a couple of paragraphs referencing tables 1 to 5 to state exactly how measurements were used to define the parameter range for the model sensitivities, simply referencing the relevant observational publications. Such a concise presentation would better allow a reader to gauge exactly how much observational constraint is provided for the sensitivity analysis without having to dig out the material. 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. Indeed it is unclear whether such a scale of uncertainty is significant given the other sub-grid difficulties such as representation of updraughts. Some discussion of these aspects should be included at the expense of concision in the more superfluous material.

Below I've listed a few specific areas which the authors should also pay attention to in any revised submission.

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. However, some of the things they don't cover appear quite relevant to the study and should be addressed by the authors: 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. 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. I do not necessarily expect the authors to agree with these claims, but think it must be considered a first order sensitivity in BB dominated aerosol to be folded into the mixing-state and kinetic limitation discussion.

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.

If the authors were to provide a tighter and more focussed version of the manuscript addressing the points above, I think it would be a useful contribution to the literature.

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