

Interactive comment on “Importance of the organic aerosol fraction for modeling aerosol hygroscopic growth and activation: a case study in the Amazon Basin” by M. Mircea et al.

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Answers to Referee 2:

We would like to thank the Referee for her/his constructive comments on the manuscript. We have integrated most of our responses to her/his comments into the revised version of the manuscript.

1) In our study we assume an internally mixed scenario for the various organic and inorganic aerosol chemical compounds, since we rely on the analysis performed on impactor samples. The more complex mode characteristics of the HTDMA measurements (discussed in the companion paper by Rissler et al. (2005)) indicate that purely

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inorganic particles were not present during the experiment since the highest measured DGF was only about 1.3. Moreover, these relatively more hygroscopic particles represented a small fraction of the total during the dry period and the hygroscopic growths of the less and more hygroscopic particles were relatively close in growth during the wet period. Our study suggests that the less hygroscopic particles, the most common particles type with respect to the hygroscopic properties, have a chemical composition dominated by organic compounds with limited solubility. The HTDMA modes discussed in the companion paper by Rissler et al. (2005) may derive from a partial external mixing of some organic and inorganic components. However, without any direct information on the chemical composition (and not only on the hygroscopic properties) of single particles, any redistribution of the 9 model organic compounds and of the 4 inorganic salts into separated populations of particles in our model will be simply artificial and will not bring more light on the importance of chemical composition in modeling aerosol hygroscopicity and ability to activate. In particular, the low concentration of aerosol particles with moderately hygroscopic behavior will not affect visibly the predicted CCN number concentrations. Also, intuitively, the DGFs predicted with other aerosol chemical compositions (invented but constrained to the impactor and HTDMA data) will be contained between the two extreme simulations already presented in the MS: insol (insoluble organic species) and IS_d (infinitely soluble and completely dissociated organic species). As the Referee pointed out, simultaneous knowledge of the chemical composition and of the state of mixing of aerosol are important issues and we have tried to address here a part of the story.

On page 5266, the line 26, after (Fig. 1) the following statement will be added: “In all the calculations of this study, the aerosol particles were assumed to be an internal mixture of inorganic and organic species because we rely on the chemical and physical analysis performed on impactor samples. The assumption is in reasonable agreement with the HTDMA measurements that showed a quite homogeneous hygroscopic behavior during wet period and only a minor fraction of moderately hygroscopic particles during dry period (Rissler et al., 2005).

In the References section, the following will be added: J.Rissler, A.Vestin, E.Swietlicki, G.Fisch, J.Zhou, P.Artaxo, M.O.Andreae, 2005: Size distribution and hygroscopic properties of aerosol particles from dry-season biomass burning in Amazonia, Page(s) 8149-8207. SRef-ID: 1680-7375/acpd/2005-5-8149.

2) The diameter of the insoluble aerosol residue, DN, was computed from the total insoluble volume contained in a drop or in a wet aerosol particle at a given relative humidity (RH) or supersaturation. The total insoluble volume was assumed to be the sum of the insoluble volumes (Pruppacher and Klett, 1990) remaining in that drop/particle for each species. The insoluble volume of each species was computed considering the solubility of the specie and the amount of water present in the drop/particle. In all calculations, the particles were considered spherical. As already mentioned on the page 5268 of the MS, “the solubility of each species was considered to be unaffected by the presence of the others” species.

On page 5262, after the line 18, the following explanations will be added: “The diameter of the insoluble aerosol residue, DN, is computed from the total insoluble volume contained in a drop/wet aerosol particle at a given s. The total insoluble volume is assumed to be the sum of insoluble volumes (Pruppacher and Klett, 1997) remained in that drop/particle for each species. The insoluble volume of each species is computing considering the solubility of the specie and the amount of water present in the drop/particle. In all the calculations, the particles are considered spherical.”

In the References section, the following will be added: Pruppacher, H.R. and Klett, J.D., 1997: Microphysics of clouds and precipitation, Kluwer Academic Publishers, Dordrecht, 955pp.

3a) The concentration of 0.1 mol per liter was the highest concentration obtained for the Amazonian aerosol samples that still allowed laboratory measurements of surface tension. The extrapolation of surface tension measurements below this concentration needs more knowledge on the nature of the surfactants and on their interaction. How-

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ever, in various samples, we have observed surface tensions modifications of the film structure due to phase changes of the surfactants at high concentrations. Therefore, the assumption that surface tension is constant above a limit concentration is more plausible than its continuous decrease with the increase in the concentration.

On page 5265, after the line 15, the following explanations will be added: “The extrapolation of surface tension measurements at high concentration needs more knowledge on the nature of the surfactants and on their interaction. However, in various samples, we have observed surface tension modification of the film structure due to phase changes of the surfactants at high concentrations. Therefore, the assumption that surface tension is constant above a limit concentration is more plausible than its continuous decrease with increase in concentration.”

3b) In our opinion, the addition of symbols on Figure 2 will not bring any new information on surface tension of Amazonian aerosol, but it will only enlarge the caption. The symbols belong to aerosol sampled over all three periods. All the useful information on the surface tension is already reported in the manuscript (page 5265).

4) The maximum number of ions for each compound will be added in Table 2.

On page 5267, the lines 9 and 10 will be changed into: “In the following sections, the observed DGFs are shown as a function of the aerosol mobility diameter and the predicted DGFs are shown as a function of the aerosol aerodynamic diameter (Figs.4 and 6).” The same change will be made in the caption of Fig.4. This explains the differences in dry diameters between the measurements and the calculations.

5) We apologize to the Referee, but we do not see any inconsistency between the results and the discussion in the text. Table 3 actually shows that the insol simulations give “good” estimation of DGF only during the wet and transition periods. During the dry period, the best agreement is obtained for LS_u simulation. As already mentioned in the manuscript (page 5269), during the wet and transition periods the insoluble fraction of aerosol was underestimated due to the lack of data on the same impactor samples.

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Thus, the good results of insol simulations are obtained for a wrong reason during these periods and we use them only as a reference (control case) for the other simulations. The best DGF predictions, as is written in the manuscript, were obtained for the dry period when the mass closure was performed for the same impactor samples, knowing the molecular composition of WSOC and their solubility.

6) The effect of “more” or “less” soluble sets of model compounds is qualitatively the same for any aerosol sample used in this study, but quantitatively can be different according to the representation of WSOC and the amount of WSOC in the sample. Unfortunately, the present uncertainties associated with the organic molecular representation of WSOC do not allow us to quantify in a more accurate way the effects of WSOC representation on aerosol growth and activation. However, we aim to show that the WSOC representation can be an important source of errors in modeling aerosol growth and activation and, therefore we consider appropriate to show the effects of “more” or “less” soluble sets of model compounds only on one sample (the TD aerosol sample) for which both DGF and CCN measurements were available.

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