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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Water uptake and chemical composition of fresh aerosols generated in open burning of biomass" *by* C. M. Carrico et al.

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Interactive comment on "Water uptake and chemical composition of fresh aerosols generated in open burning of biomass" by C. M. Carrico et al.

Anonymous Referee #1 Received and published: 14 February 2010

Comment: This paper describes the results of experimental studies on the hygroscopic growth factors (GF) of fresh biomass burning smoke particles. The paper is well written and the results are presented clearly. The most important result is that particles dominated by organics are less hygroscopic than those dominated by inorganic salts.

Reply: We thank the reviewer for the kind words and for taking the time to review the manuscript.

Comment: Since this was a laboratory experiment on fresh biomass burning aerosols, what is the general relevance of the results? Why would they be expected to apply at remote continental or globally-representative locations. Why would highly aged smoke be expected to behave similarly to fresh smoke generated in the laboratory?

Reply: To address this comment, we have added the following statement to the introduction (P. 3630, Line 26): "Smoke properties undoubtedly evolve after emission as ambient smoke is transported from source regions, ages and mixes with other aerosol types. Fresh smoke emissions, however, are important in source regions and as the starting point for predicting the evolution of smoke properties."

We have added the following statement to reiterate that these results are for freshly emitted smoke (P. 3641, Line 14): "We reiterate that our results apply to freshly emitted biomass burning aerosols, which is only one important subset of all ambient aerosols."

To emphasize that the results from these experiments have a more general relevance though, we have added the following statement in the discussion and conclusions section (P.3642, Line 10): "The general consistency of the fresh smoke properties reported here with ambient observations is important as it suggests that smoke hygroscopicity may be modeled with some general relationships to fuels, combustion conditions, and the inorganic-to- organic composition ratio."

Comment: Dinar et al. (2007, 112, D05211, doi:10.1029/ 2006JD007442) reported a GF (90) of 1.24 for HULIS extracted from samples dominated by aged biomass burning aerosol. How do those results compare with those derived from the laboratory experiments? What would be the kappa for HULIS with a GF of 1.24 at 90% RH?

Reply: We have added the following statement (P. 3641, Line 24) to link these results to the paper by Dinar et al. (2007): "Dinar et al. (2007) report hygroscopicity ranging from $0.07 < \kappa < 0.10$ for extracts of humic-like substances from biomass burning samples, with larger values corresponding to more aged aerosols. These values compare to the least hygroscopic smokes we found in this study, i.e. those that contain the highest

fractions of carbonaceous material."

Comment: While there appears to be a (log-log) relationship between kappa derived from 100 nm particles and the ratio of total carbon/inorganic ions derived from PM2.5 samples, this implies that chemical composition is invariant with particle size. As the authors note in the introduction, this may not the case.

Reply: We agree that the composition and thus hygroscopicity varies with (and within) particle size. We did not intend in the manuscript to claim that composition is invariant with size. Figure 7 is simply intended to illustrate the general relationships (with scatter) that we observed, and not to be predictive. Heterogeneity of composition is mentioned in several places including on P. 3628, Line 14-17: "The bimodal κ distributions were indicative of smoke chemical heterogeneity at a single particle size, whereas heterogeneity as a function of size was indicated by typically decreasing κ values with increasing dry particle diameters," And on P. 3637, Line 12-14: "Comparisons of κ values for Dd=50, 100, and 250 nm particles show that this chemical heterogeneity was a function of particle size. Generally, the number fraction of particles in the more hygroscopic mode increased with decreasing particle size."

We have added the following statement on P. 3641, Line 9, to further emphasize this point: "Variability in chemical composition as a function of particle size is one likely contributor to discrepancies between measured κ and those calculated from bulk PM2.5 chemical composition."

Comment: More detailed chemical characterization as a function of particle size could have been obtained with an AMS, which, if I am not mistaken, was deployed during this experiment. If this is the case, why weren't those data used?

Reply: The reviewer is correct that AMS measurements were made during these experiments. Our observation that the inorganic fraction of the aerosol really drives the hygroscopicity of fresh smoke is important here. Since we expected refractory species, including elemental carbon and mineral dust species, to be present and to play a role

in the hygroscopicity, the traditional filter based collection coupled with IMPROVE analysis protocols (e.g. ion chromatography for inorganic ions) gives a more complete dataset than does the AMS alone, although of course with no time resolution.

Comment: Nonetheless, this paper suggests that hygroscopic growth can be estimated well using a simple chemical and thermodynamic parameterization. This has important implications for modeling aerosol optical and hygroscopic properties on local, regional, and global scales.

Reply: We agree with the reviewer and have added the following statement in the conclusion: "It appears that this simple chemical parameterization is potentially predictive of the bulk hygroscopic properties of smoke, but more data are needed to generalize this finding."

Interactive comment on Atmos. Chem. Phys. Discuss., 10, 3627, 2010.