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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Cloud condensation nuclei activity of fresh primary and aged biomass burning aerosol" *by* G. J. Engelhart et al.

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(1) *Engelhart et al. present CCN experiments on fresh and diluted photochemically aged biomass burning aerosol. The main finding of the study is that after aging variability of chemical composition is decreased, thereby simplifying the modeling their impact on clouds and climate. The experiments are novel, data appear to be of high quality, the manuscript is well written, and the results are relevant to the community. I therefore recommend publication in ACP.*

We do appreciate the positive comments.

Comments:

(2) *I agree with the comments made by referee 1 and encourage the authors to address those.*

We have done our best to address all the comments. Please see our response to Referee 1 for details.

(3) *I am missing a more quantitative comparison with prior results. If data are available for overlapping fuels, these should be added to Table 1. The main reason for this request is to begin understanding similarities and differences between studies (e.g. what is the variability of pine burns not only within, but between different methodologies). Adding such a comparison to the table will make it easier for future investigators to select fuels.*

We have added the corresponding information and brief discussion to the revised paper.

(4) *I am also missing discussion about heterogeneity within the CCN data. Previous studies showed that it is quite difficult to assign a single kappa value to an entire burn. It varies with particle size and the time/dilution history of the plume. Indeed this point seems to be one of the main motivations for this study. Yet Figure 2 assigns a single "primary" and "oxidized" kappa to the aerosol, with no regard to variability. The authors should not only add variability bars (also suggested by referee 2) but expand the manuscript to discuss metrics of variability. For example, Figure 5 could be a good starting point in computing a mean kappa, delta kappa, and show an increase and/or decrease in the delta kappa with time. In fact, Fig. 5 seems to show that variability within the plume increases 4 hours after lights on, even though variability between fuels decreases.*

The primary aerosol was quite stable during the characterization of the fresh emissions, so we are comfortable reporting a single value. The dilution in the smog chamber was not changing during this characterization period, which may account for the difference

from previous work. Error bars have been added to Figure 2 to quantify the uncertainty of the corresponding measurements. During the oxidized period the kappa was most certainly changing, so the value reported is the average for the last 30 minutes of the experiment. This had been foot noted in Table 1 and has also been added to the caption of Figure 2. The increase in activation diameter measurement variability after 4 hours of photochemistry seen in Figure 5 is largely due to decreasing signal due to the particle loss to the walls of the chamber.

(5) *Another measure of heterogeneity to analyze would be the spread of the activation curves, of which none are shown. It would be useful if the authors added at least examples for each burn to a supplement.*

Examples of each burn have been added to the supplementary information of the paper.

(6) *In the manuscript the authors state that "analysis of the sigmoidal fit was carried out according to the method of Rose et al. (2008)". What does that mean? Does that include the fit to the data? Is a multiple charge inversion used? If so, was it done for both for the CN and CCN? If the method of Rose et al. was used, is it valid for heterogeneous samples and SMPS scans? If no inversion was used, how much does this increase the uncertainty? Please add instrumental parameters: e. g. sheath-to-aerosol flow ratios in the DMA and CCN and potential adjustments made to the data (e.g. if 100 percent activation was not reached due to the presence of non-activating particles), was the resulting activation spectrum scaled to one? If not, what does that mean for the 50 percent activation diameter? In general the methods/data analysis section should be expanded to help the reader understand what has been done specifically to this data set.*

Information about the sheath to aerosol flow ratios for the DMA (10:1.5) and CCN (10:1 with total flow of 0.5 lpm) has been added to the paper. No multiple charge

inversion was used in the beta version of SMCA analysis, although the most recent versions of this software include the ability to do multiple charge analysis. Neglecting this inversion creates a relative uncertainty in the activation diameter of 3-4 percent. Activated fractions were scaled to 1. These details have been added to the paper. For further explanation of SMCA we direct readers to the Moore et al. (2010) reference.

(7) *Much of the analysis is descriptive (regress quantity x vs. y) rather than predictive. Perhaps the data scant do not allow more quantitative analysis? E.g. based on prior correlations of O:C ratio vs. kappa and OA/inorganic fractions, can you predict the evolution of kappa values?*

The inorganic content of the aerosol and more specifically its variation with particle size did not allow more quantitative analysis of the data. While the bulk composition of the particles was measured using filter samples, the composition of the particles in the activation diameter size range was not known. The AMS cannot measure the refractory components of the PM therefore it cannot provide a good picture of the various salts that existed in the activated particles. It became clear from the analysis of the measurements that the organic/inorganic ratio for these particles could be quite different from this of the bulk aerosol. This uncertainty about the size-resolved particle composition made the calculation of the organic aerosol kappa very uncertain.

(8) *Eq. 2 should include "Dd" not "d", "S" should be "Sc", and T and σ_w should be fixed to be consistent with the definition in Petters and Kreidenweis (2007).*

We have made the suggested changes.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 7521, 2012.