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Atmospheric
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Discussions

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

Anonymous Referee #1

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This paper presents CCN derived hygroscopicity parameter (κ) of aerosol particles from a range of different biomass burning experiments, both before and after photochemical ageing in a smog chamber. A wide variety in κ is observed for primary biomass burning aerosols in experiment with different types of fuel, which is related to effects of chemical composition. This variety decreases after photochemical ageing, and a secondary material from biomass burning is determined to have a CCN activity comparable to secondary organic aerosols from laboratory studies of monoterpenes. The paper is very well written and presents a well analyzed dataset, dealing with highly complex processes of atmospheric interest. I would recommend publication after a consideration of the minor and technical concerns, listed below:

Specific comments:

In general, I miss either error bars in the figures or an estimate/discussion of the experimental uncertainties in determining κ, organic mass fractions and f44.

P. 7524, l. 10: How much was the degree of dilution, and what were the emissions diluted with (purified air, free of particles, trace gasses, water etc.)?

P. 7524, l. 15: Can you characterize the UV lights compared to ambient sunlight?

P. 7524, l. 26: It is described how the emitted aerosols were preconditioned due to their fractal nature, but not what the effects of the treatment are (or at least what they are expected to be), or what the consequences had been if this preconditioning had not been carried out.

Section 3.1: Much of this section describes the practical, experimental operation rather than the theoretical background. You should consider combining it with Section 2.

P. 7526, l. 20: Is the density of 1 g/cm an arbitrary estimate, or is there a scientific reasoning behind this choice?

P. 7527, l. 8: Was a new nucleation mode clearly observable after lights on? It could be interesting to see examples of size distribution before and after lights were turned on, for example as supplementary material.

P. 7527, l. 13-15: What does a ratio < 1 mean (as for example in burn no. 65)?

P. 7527, l. 19: What was the range of κ in the described regression? I would suggest that you made a figure of the paired κ sets and added it as supplementary material, possibly with a table showing for which experiments/fuels the differences were most significant. Have you considered if the variation in κ for different experiments with the same fuel could be due to variations in the dilution system? And have you considered that variation in κ for different supersaturation could be caused by evaporation in the CCN counter, similar to the effect described by Asa-Awuku et al. (2009)?

P. 7528, l. 12: The abbreviation “Q-AMS” has not been defined, and the purpose and concept of this technique should be introduced in Section 2 (Experimental methods).

P. 7528, l. 13: The high volume filter measurements should also be described in Section 2, unless these results have already been described in an earlier study – in which case this study should of course be referenced.

P. 7528, l. 23: This is the first mention of the study's experimental characterization of POA (excluding the abstract). The concept (i.e. that POA is the organic particulate matter present after filling but before photo-oxidation) should be clearly defined, either here or in an earlier section. Be sure to make it clear, that the primary material has both an organic component (POA) and an inorganic component.

P. 7529, l. 2: You should discuss that some studies have observed more complex dependencies between CCN activity and the degree of oxidation, see for example Massoli et al., (2010); Chang et al. (2010) and Frosch et al. (2011), and also that CCN activity seems less sensitive to oxidation than hygroscopicity (compare with e.g. Tritscher et al. (2011) and Duplissy et al. (2011)).

P. 7529, l. 5-8: Although concentrations of aerosol, oxidants and trace gasses are listed in the earlier study by Hennigan et al. (2011), it would be helpful to have a summary of concentrations of the main components available in the present study.

P. 7530, l. 7ff: You discuss very thoroughly how photo-chemical ageing DEcreases CCN activity. However, I miss some discussion on the chemical reactions and processes leading to an INcrease in CCN activity, observed in at least some experiments.

P. 7530, l. 21-25: Have you considered that an organic coating on the primary particles might also hinder water uptake, change growth droplets and delay activation, (similar to for example the effect of b-caryophyllene SOA reported by Asa-Awuku et al., 2009)?

P. 7531, l. 2: Change "ponderosa pine experiment" to "Burn number 40" (to avoid confusion, since there are two ponderosa pine experiments). Have you any explanation for the high value in burn number 40, or of why you could determine κ in only 14 of the

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18 experiments? And why only 14 experiments?

P. 7531, l. 12: It would be good if you could show initial organic mass fractions for each experiment, for example in Table 1. It would also be interesting if you discussed the change in CCN activity caused by photo-oxidation in relation to the change in f44, which is probably also seen due to photo-oxidation and chemical ageing.

P. 7531, l. 12: I am not entirely sure of your meaning: Do you mean that SOA and POA have similar CCN activity/hygroscopicity, or SOA and PA (primary aerosols, meaning both organic and inorganic) have similar CCN activity/hygroscopicity?

P. 7531, l. 13-18: The simple mixing rule for determining κ for a multicomponent system (eq. 3) has been presented in Section 3. Have you considered to actually test it, using for example the κ listed for example the black spruce experiment? You already have κ for the primary material (i.e. before photo-chemical ageing) and κ for SOA. Using SMPS data (or AMS masses with some assumption regarding density), you could determine relative volumes of primary and secondary aerosols.

P. 7532, l. 17-23: The comparison with the studies by Moore et al., Rose et al. and Petters et al. could be expanded to include more details and would fit better in Section 4.

Table 1: You state that aged κ was averaged from measurements from the last 30 minutes of data (footnote c). Was there any temporal variation? And did all experiments last the same number of hours, or could different exposures to oxidants explain some of the experimental variations discussed in the paper?

Figure 3: I wish I could see which types of fuel resulted in the various organic mass fraction – perhaps this information could be included in the figure, or initial organic mass fractions could be listed in Table 1?

Figure 4: SS (in the label on the y-axis) has not yet been defined (supersaturation was defined as S in eg. 1). Make clear that “primary aerosol” is not the same as POA. Why

does the figure only contain data from supersaturation 0.26%?

Figure 5: Throughout the paper, you mainly describe κ. Why do you suddenly choose to display activation diameter on the y-axis, and not κ? And does the shaded area represent an estimate of the experimental error? From the data, it looks like uncertainties increase with time – which one would also expect from the decreasing particle concentration caused by wall losses. However, this is not reflected by the shaded areas.

Figure 6: Consider giving all three subplots the same y-axis (from 0 to 1), which would make them easier to compare. Change "kappa" in the x-axis label to "κ"

Figure 7: As for Figure 4, SS (in the label on the y-axis) has not yet been defined (supersaturation was defined as S in eg. 1). Consider centering y-axis around 0 and marking 0 with a horizontal line. Is "Fresh POA mass fraction" identical to the "initial organic mass fraction", which is mentioned in Section 4? If so, you should use the same phrase – or alternatively describe the differences. Also, it could be very good if you discussed for which experiments/fuel types you observe the largest change in κ.

Technical comments:

You alternate between writing "-values" (e.g. P. 7528, l. 8) and " values" (e.g. P. 7522, l. 12) – please make sure you are consistent.

P. 7525, l. 13: Change "is" to "was".

P. 7527, l. 20: Consider changing "Values range . . . = 0.6" to "κ values ranged from 0.06 to 0.6)

P. 7528, l. 15-18: It is confusing that the sentence starts in the present tense, "There is a clear relationship", and ends in the past "mass fractions increased . . . primary aerosol decreased".

P. 7532, l. 11: Change to: "These new particles nucleated and grew due to. . ."

References:

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