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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Properties and emission factors of CCN from biomass cookstoves – observations of a strong dependency on potassium content in the fue" *by* Thomas Bjerring Kristensen et al.

Anonymous Referee #2

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Review of 'Properties and emission factors of CCN from biomass cookstoves - observations of a strong dependency on potassium content in the fuel', T. Kristensen et al., ACPD, 2020

This manuscript presents an analysis of the CCN properties of the combustion of a range of fuels in different types of stoves to assess the impact of residential cooking fires on CCN emissions. This manuscript of part the Salutary Umea STudy of Aerosols IN biomass cookstove Emissions (SUSTAINE) to study sustainable approaches to residential cooking in Sub-Saharan East Africa. The experiments were conducted using

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traditional stoves (3-stone stove and rocket stove) with wood logs comprised of sesbania, casuarina and birch (reference-type). Natural draft and forced draft stoves were used in conjunction with pellet fuel made with different materials, including coffee husk, rice husk or water hyacinth mixed with Swedish softwood, which also served as a reference. The objective of the experiment is to determine the contribution of stove and fuel to CCN emissions and assess the contribution of residential cooking to aerosol-cloud interactions and public health.

Measured particle number size distributions were bi-model with an ultrafine mode that was often less than 65 nm diameter, as well as a soot mode centered on average 150 nm diameter. CCN hygroscopicity and effective density were measured for up to four sizes (65, 100, 200, 350 um diameter). The ultrafine mode was moderate to highly hygroscopic (kappa between 0.1 and 0.8), with higher kappa associated with higher combustion temperature and soluble salts such as potassium, while nthe soot mode was much less hygroscopic (kappa between 0 and 0.15) and comprised of black carbon and organic material.

The correlation between CCN emission factors and PM emissions factors is highly dependent on aerosol hygroscopicity (particularly K content) and combustion temperatures (stove type), respectively. In general, well written, but at some points that need clarification, particularly the calculation of CCN emissions factors.

General comments: The reviewer suggests the authors add a schematic showing the layout of the instrumentation and the sampling configuration. A schematic would help distinguish which instruments are sampling in the flue, which instruments are sampling from the stainless-steel chamber, and how the dilution lines and ageing experiments are implemented. In addition, details such as lengths and layout of sampling lines and chamber residence times are helpful in assessing particle losses. Pertinent details should be included in this manuscript and not just referred to in another publication.

In lines 240+, the authors state that they were not able measure an increase in di-

ameter with ageing from particle number size distribution in spite of such indications from the CCN, APM and AMS measurements. Yet, in the same section, the authors note that that average aerosol number size distributions were larger than flue measurements. Much of the discussion in this manuscript on ageing was centered around chemical changes by the addition of secondary organic material, but there are also physical changes to soot particles, particularly in the first hours after emissions [Li et al., Atmospheric Environ., 2015; doi: 10.1016/j.atmosenv.2015.09.003]. This section would benefit from a discussion of the evolution of particle morphology as well.

In lines 519+, the authors state that the step in the FDS-sw curve near 0.7% supersaturation (Figure 4b) is related to activation of the soot mode within a narrow supersaturation range. The FDS-sw soot mode (Figure 1) appears encompass a size range from ca. 80 to 250 nm diameter, which corresponds to supersaturations between approximately 1.6% to 0.4%, respectively (kappa $\sim$ 0.004). This range of supersaturations for the soot mode is more than an order of magnitude more than the range shown in the step in Figure 4b. The reviewer suspects there is a step in the CCN spectra related to a discontinuity between flow scans and CCN spectra or the use of discrete hygroscopicity values applied to the number size distribution. The authors need to describe their calculations.

Specific comments: L11: specify or give a reference for 'standard protocols'

L43+: it would help the read to specify 'diameter' throughout the text

L77: change 'focus' to its plural 'foci'

L115: triangular cross-section has been described with two numbers (dimensions of 2.6 cm times 2.5 cm . . .). It's not clear to what these dimensions are referring.

L124: provide a reference for the standardized water boiling test 4.2.4

L130: replace 'were operated' with 'were initiated' or 'began'

L170: what was the range of the total flow rate scans of the CCN instrument?

L276+: The authors state that particle number size distribution in the flue (from the fast particle analyser) were similar to the initial measurements taken by the SMPS in the aerosol chamber, which seems to confirm consistency between independent measurements of particle number size distributions. The authors then note than average number particle size distributions in the chamber were larger than those of the flue, which seems logical given that the particles in the chamber are no longer representative of the fresh emissions in the flue. Why do the authors speculate losses, coagulation in the sample lines or offset between instruments, especially given the consistency between the fast particle analyser and the SMPS at the beginning of the experiment?

L302, Section 4.2: The impact of mixing state can also be assessed by using the aerosol size distribution and associated aerosol hygroscopicity to regenerate the CCN spectra and compare cases of different mixing states. The authors inferred a single kappa value, which suggests then that either the aerosol is internally mixed or that external mixtures do not produce significant differences in the CCN spectra in this study. Also, what do the authors mean by CCN spectra of 'appropriate quality'? The reviewer also encourages the authors to add a figure showing the CCN spectra to compliment Figure 1.

L371: Do the authors mean a more spherical aggregate when referring to more compact black carbon particles? The ageing experiments using the PAM and thermodenuder clearly show the impact of the SOA condensing onto fractal aggregates. However, did the authors also observe morphological changes (for example, an evolution of effective density compared to initial measurements)?

L390; Section 4.3.2: A reference to Table 3 needs to be added earlier in the paragraph to orient the reader.

L397: replace 'totally' with a quantitative assessment

L403: replace 'state-of-the-art' with literature references

L407: remove ‘more’ in ‘more refractory species’

L455+: This discussion in this section could be reorganized and main points clearly stated. This paragraph discusses 3S-RS results, the next paragraph discusses NDS and FDS results, and then the discussion returns back to 3S-RS results.

L473: What do the authors mean by ‘variations in kappa for 200 nm particles . . .. correlate with variations in ultrafine kappa values for FDS’? The discussion in this paragraph is not clear. The reviewer interprets the results as 3S associated with a higher organic fraction across the entire size distribution and is consistent with higher relative densities, while RS have higher EC fractions along with lower relative densities.

L500: what ‘special conditions’ are the authors referring to?

L524: Have the authors tried to quantify wall losses and coagulation to assess how much they may impact CCN emissions?

L614: The authors suggest that PM emissions are sensitive to the very large particles, which is not entirely correct. PM emissions are sensitive to the mass size distribution (the product of the number concentration and the particulate mass at a given size). A figure showing the calculated mass size distribution using the effective densities would be useful in illustrating this point.

L664+: The reviewer suggests to integrate the perspectives (wildfires and health impacts) into the conclusions.

L676+: As stated in the text, the optimal scenario would be a reduction in both PM and CCN emission factors. Based on the experiments conducted here, can the authors reiterate what specific combinations of stove / fuel should and should not be used?

Figures 1 and 2: A description of the legend is needed in the figure captions.

Figures 4 and 5: As mentioned previously, the issue with the steps in the emission factors needs to be resolved. It is also not clear what is the purpose of the insets at

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low supersaturation.

Figure 5: ‘For most other PAM experiments. . .’ This analysis needs to be in the main text rather than the figure caption.

Figure 6b: The upper part of the label for gray-scale bar has been cut.

Table 3: Add the chemical analysis used to determine the ash content in the figure caption.

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