

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.

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We thank the anonymous reviewer for the many detailed comments and suggestions. We find that the revised version of the manuscript has improved due to these comments, and we thank the anonymous reviewers in the revised version of the manuscript. Our references to line numbers and figures generally refer to the ACPD version of the manuscript – unless specified differently. There appears to be an offset in line numbers of about 1 between the manuscript version the reviewer refers to, and the ACPD version of the manuscript, which we refer to. Our reponses appear in **bold**

below.

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 is part of the Salutory Umea Study of Aerosols IN biomass cookstove Emissions (SUSTAINED) to study sustainable approaches to residential cooking in Sub-Saharan East Africa. The experiments were conducted using 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-modal 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 nm diameter). The ultrafine mode was moderate to highly hygroscopic (κ between 0.1 and 0.8), with higher κ associated with higher combustion temperature and soluble salts such as potassium, while the soot mode was much less hygroscopic (κ 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 instru-

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mentation 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.

A schematic of the experimental setup has been included as the first figure in the revised manuscript.

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

We made lognormal mode fits to the SMPS soot mode before, during and after sampling through the oxidation flow reactor. In a few cases, we observed very minor increases in the soot-mode-diameter during parts of the simulated ageing experiments, and in most cases we did not. The experiments included in the ‘aged-CCN-emission factors-figure’ mainly belong to the latter category. The fact that the CCN and effective density measurements were more sensitive to minor SOA formation on the soot particles than the SMPS measurements is not surprising. The initial kappa and effective density levels were significantly below the expected corresponding values for SOA, so addition of a relatively small SOA volume can be detected in those properties.

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We have added the following comment to L247: “This is most likely due to the initial soot particle CCN activity and effective density being very low compared to those properties of SOA. Hence, soot particle CCN activity and effective density are very sensitive to minor additions of SOA.”

The ageing effects reported by Li et al. (2015) are all based on a rather different aerosol stored in an environment with significantly higher RH than in our study. Furthermore, we generally report results obtained shortly after chamber filling. We cannot rule out effects of coagulation, which is discussed in detail in the paper – but as mentioned, that will be the case in any type of experimental set up – and also real-life cookstove emissions.

We mainly include the effects of photochemical ageing on the CCN properties on a qualitative level, and we do not find it adequate to go further into detail with such ageing effects in this study. The experimental approach was simply not optimized for studies of photochemical ageing.

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 ($\kappa=0.004$). 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.

The calculations behind the curve was described in L. 220-224, and the relevant κ values for 100 nm and 200 nm particles were provided in Table 2. The reviewer assumes the κ value for the soot mode to be constant with chang-

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ing mobility diameters, which is not a reasonable assumption. Nevertheless, the reviewer is both right and wrong here.

It is not meaningful to assume a constant apparent kappa value of 0.004 for the entire soot mode. Nevertheless, this reviewer comment led us to reprocess a lot of data, and we found a mistake in the data processing influencing this particular CCN emission factor curve. We will describe these aspects in more detail below.

In this study, we report kappa-values calculated from the mobility diameter as described. There are 3 reasons why we would expect such a kappa value to decrease with increasing mobility size over a soot mode, which is evident from the kappa values reported in Table 2.

1. If we assume that the chemical composition of the soot mode does not vary significantly with the particle size, then one may assume that the kappa-value calculated from the volume equivalent diameter could be close to constant (Dusek et al., 2011). We observe pronounced decreases in the effective density with increasing soot particle mobility diameter (Fig. 3). Hence, for that reason, the reported kappa values should decrease with mobility size over the soot mode. However, that shape-effect is unlikely to explain the entire difference in reported kappa values between the 100 nm and the 200 nm soot particles in our study.

2. Kappa is a volume-based property as mentioned in the manuscript. We observe strong indications of the ultrafine particles contributing to increasing the kappa-values of the soot modes. The kappa value of the 100 nm soot particles will increase ‘faster’ with coagulation with the ultrafine particles relative to the much larger volume of the 200 nm soot particles. We expect this effect to be of importance in this study, but it is not straightforward to estimate the magnitude of this effect.

3. There is a third effect leading to expected decreasing kappa with mobility diameter for a soot mode, which is described in more detail by Wittbom et al.

(2014). For low-hygroscopicity soot particles, the cloud droplet activation may occur on just a fraction of a soot particle since the critical droplet size at activation is relatively small. That effect will be more pronounced with increasing soot particle size and lower kappa value, and it depends on particle morphology. Whether this effect plays a role for the CCN activity when comparing kappa values of 100 and 200 nm soot particles in our study is not entirely clear, but based on the work by Wittbom et al. (2014), it cannot be ruled out.

In the re-processing of our data, we discovered a mislabeling of a data file. Unfortunately, the reported kappa values for the FDS-sw represented an experiment with an alternative and non-ideal stove operation. Hence, that mistake influenced the CCN emission factor curve of the FDS-sw experiment presented in Fig. 4.b. The correct values for the FDS-sw experiment are now presented in Fig. 2, Fig. 4.b and Table 2 of the manuscript.

In this case, the reviewer was correct that the pronounced step in the CCN emission factor curve previously reported – should indeed not be as steep. But it does not change the fact that significant mobility size ranges of the soot mode often activate within relatively narrow supersaturation intervals due to the reasons described above.

We have replaced:

“The step in the FDS-sw curve near $SS=0.7\%$ is due to activation of the entire soot mode within a narrow range of supersaturations. That soot mode is probably produced due to quenching of the flame on the bottom of the pot, and a more optimal fuel load can potentially significantly reduce those soot particle emissions.”

With:

“The steps observed in some cases (e.g. NDS-sw, NDS-sw-ch) are results of significant particle size ranges activating into cloud droplets over a narrow range of supersaturations due to the strong increase in kappa with decreasing D_p in

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some cases.”

Specific comments:

L11: specify or give a reference for ‘standard protocols’

We do not find it adequate to provide references to standard protocols in the abstract. Specifications are provided in the relevant section (Materials and Methods). The analysis of the fuel composition was carried out in a specialized and commercial external laboratory. The protocols can be purchased online.

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

The mobility diameter (D_p) is defined in L. 88 and used throughout the manuscript. The diameter referred to in L. 43 would typically be the aerodynamic diameter, and that is not of direct relevance to the focus of our study.

L77: change ‘focus’ to its plural ‘foci’

That has been changed as suggested

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.

Those dimensions refer to the base and the height, respectively. The text has been changed to include that information.

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

We find it more appropriate to refer to the 4.2.3 water boiling test described by the Clean Cooking Alliance in 2014. The document can easily be found by use of a web search engine: <https://www.cleancookingalliance.org/binary-data/DOCUMENT/file/000/000/399-1.pdf>

But we do not know whether the document will remain there for a foreseeable future, so in the revised manuscript we provide the main web-address of the

organization.

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

'were operated' has been changed to 'were initially loaded'

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

The range in CCNC flow rates (0.2 to 1.0 lpm) was described in detail in L. 155-160. The associated range in terms of supersaturation was between about 0.1% to about 1.5% depending on the temperature gradient as reported in L. 169-170.

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 that 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?

The two different instruments (FPA and SMPS) used for measuring particle number size distributions did not produce 1:1 identical particle number size distributions (as described in more detail in the text), and we would not expect them to do so. The general particle number size distribution features were rather similar between the two instruments – when comparing the flue gas measurements with the initial emissions into the aerosol storage chamber – as stated in the text. It was not possible for us to tell to which extent the described minor differences were due to instrumental differences and/or diffusional losses/coagulation effects.

The aerosol population in the flue gas originating from biomass combustion

may change significantly over timescales at the order of 10s, so the FPA is the better choice for measurements in the flue gas. In addition, the instrument is optimised to reduce losses of the smallest particles. We have included the following to section 3.3 in order to clarify advantages and limitations to the FPA in this context.

“It was operated with a sample flow rate of 8.0 slpm and the inversion matrix for bimodal spherical aerosol particles from the manufacturer was applied. This matrix is suitable for spherical biomass particles and heavily coated soot, but it may underestimate the sizes of fractal-like soot particles due to the particle shape influencing the charging efficiency. That effect may play a role for fractal soot aggregates with mobility diameters larger than 150 nm and can, when these are present, result in a slight overestimation of particle numbers at smaller sizes (Symonds et al., 2007). The main advantages of the FPA when studying combustion aerosol emissions online are (i) a very fast instrumental response time (<1s), and (ii) minimised losses of the smallest particles (Reavell et al., 2002)”

The main point is that minor biases in the particle number size distributions are unlikely to substantially impact any of the reported CCN results. Thus we have added the following statement in the L276+: “It is worth noting that none of the listed potential minor effects will have substantial influence on the results reported in this study.”

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.

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Yes, in principle, we agree with the reviewer regarding links between CCN spectra and mixing state as described in L. 305-309. However, in this context we find it essential to draw the attention to L. 310-311: ‘However, it should also be mentioned that the size resolution obtained with the DMA in the current study was not optimised for identification of the CCN mixing state’ The CCNC experimental approach was optimized for obtaining CCN spectra very fast in parallel with the APM measurements. A different approach should had been applied, if our focus had been on extracting detailed information from CCN spectra. We have rephrased the statement in L.310-311 to: ‘However, it should be noted that the CCNC experimental approach was optimised for fast scans in parallel with the APM measurements. Hence, neither the DMA transfer function nor the CCNC operation were optimised for extracting information about the mixing state from the CCN spectra.’

Furthermore, it is also worth noting that the reported main findings on CCN activity and associated indications of chemical composition of particle modes, as well as CCN emission factors may be rather sensitive to measurements at 65 and 200 nm, while it is often less sensitive measurements at 100 nm. None of the main findings in our study are sensitive to the reported kappa values for 100 nm particles.

In summary, neither do we find it justifiable to present fast scan CCN spectra in this paper as results in a separate figure, nor do we find such an approach essential for the reported main results.

When it comes to CCN spectra of ‘appropriate quality’, we have added the following to section 3.4:

‘CCN spectra were excluded from the data analysis for four different reasons: (i) if the dT was not constant, (ii) if a full CCN spectrum was not covered within the scan range, (iii) if the CCN counting statistics and signal to noise ratio were too low for analysis, or (iv) in rare cases, if the actual CCNC flow profile over a scan

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differed significantly from the pre-programmed profile.’

As a consequence, L. 308-310: have been shortened from:

‘Regardless of which of the listed options that can explain our observations, we were generally able to infer a single CCN activity from the CCN spectra of appropriate quality in terms of reasonable and stable dT and reasonable CCN counting statistics.’

To:

‘Regardless of which of the listed options that can explain our observations, we were generally able to infer a single CCN activity from the analysed CCN spectra.’

L371: Do the authors mean a more spherical aggregate when referring to more compact black carbon particles? The ageing experiments using the PAM and thermogravimetric analysis 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)?

We have rephrased ‘more compact black carbon particles’ to ‘more compact black carbon aggregates’. In most cases, ‘more compact’ is likely to imply ‘more spherical’ – but shapes may be far from spherical. ‘Compact’ is widely used in context with such soot properties in the literature.

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

The following statement has been added to L390:

“The concentration of the dominant inorganic species in the applied fuels are presented in Table 3”

L397: replace ‘totally’ with a quantitative assessment

The sentence has been rephrased to: “For rh, the fuel ash was dominated by Si (>85% by mass) with a minor fraction of K (4% by mass).”

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

'state-of-the-art' has been deleted, and references have been added at the end of the sentence (Obernberger et al., 2006; Bostrom et al., 2011; Obaidullah et al., 2012.)

L407: remove 'more' in 'more refractory species'

'more' has been deleted as suggested.

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.

We agree, and we have restructured the section for improved clarity.

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.

The discussed correlation is of general relevance to all the studied stoves. We have rephrased to:

"Whenever kappa values for the 200 nm particles were relatively low (or high), they were associated with relatively low (or high) kappa values for the ultrafine mode (65 nm). That relation is represented by a correlation coefficient of 0.76 and a p-value of 0.01 for the results presented in Fig. 3. Hence, it is likely that the ultrafine particles coagulated with the soot mode particles and thus increased the soot mode kappa values. These observations further support, that not only do the potassium compounds appear to play a key role for the CCN activity of the ultrafine particles, they also influence the CCN activity of the soot mode."

L500: what 'special conditions' are the authors referring to? For the RS-ses experi-

ment, the chamber injections did not represent a full combustion cycle. These ‘special conditions’ were described in more detail in L. 295-303.

‘which to some extent is likely to be due to the special conditions for the chamber injections for the RS-ses experiment’ has be rephrased to:

‘which to some extent is likely to be due to the chamber injections for the RS-ses experiment not being representative of a full combustion cycle as described above.’

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

We did not quantify particle losses prior to measurements of the particle number size distributions in the flue gas, but we consider potential losses negligible when it comes to the presented CCN emission factors and PM_{0.5} results. The particle number size distributions presented were measured in the flue gas close to the stoves. Flow rates were high in the flue gas (2.4 m³/min) and in the short sampling line for the fast particle analyser (8 slpm). Hence, we consider diffusional losses to be very minor. There may be non-negligible diffusional losses of the very smallest particles potentially influencing the ‘left part’ of the presented ultrafine particle modes for e.g. the FD-sw and the FD-sw-rh. However, such small particles will neither act as CCN for atmospherically relevant supersaturations nor contribute significantly to the PM_{0.5}. So we do not expect particle losses to influence any of the main results presented.

Coagulation will play a role in any kind of experimental set up to study CCN emissions from cookstoves – as well as in real-life settings. We find that our approach with significant dilution in the hood and sampling with the fast particle analyser after 2s residence time in the flue gas is optimal for assessment of the primary emissions with minimal impact of coagulation on the particle number size distributions. Coagulation involving the soot particles increases the kappa

value of those particles, which is clearly stated in the manuscript. It is not possible for us to assess the importance of coagulation immediately above the flames versus potential coagulation in sampling lines. Hence, we discuss these effects on a qualitative level in more detail in the manuscript.

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.

In the paragraph referred to, we discuss the contribution of the soot mode to the $PM_{0.5}$ emissions from the FDS-sw and the RS-ses. Based on integration of the estimated particle mass distributions, the particles with mobility diameters >100 nm contributed 97% and 74% to the $PM_{0.5}$ emissions for the FDS-sw and the RS-ses, respectively. Hence, the $PM_{0.5}$ emissions were indeed sensitive to reductions in the soot mode in those cases. We do not find reason to revise the paragraph. Furthermore, we do not find reason to include additional figures with particle mass size distributions, since it was not a focus area of this study. The inclusion of estimated $PM_{0.5}$ emissions was mainly motivated by illustrating the challenge of linking PM levels to CCN concentrations for the studied aerosol. We consider that of importance to the modeling community.

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

We have added the following to the conclusion:

“Overall, we observed high potential to significantly reduce primary $PM_{0.5}$ emissions from biomass cookstoves by (i) applying fuels with low levels of inorganic compounds potentially entering the aerosol phase, (ii) improved stove technology, and (iii) optimal stove operation. Reduced PM and soot emissions are mo-

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tivated from health and climate perspectives. However, our study indicates that biomass fired cookstoves may comprise a very significant and underestimated source of CCN, and substantial reductions of such CCN emissions may potentially lead to warming effects on climate depending on the prevailing meteorological conditions and importance relative to other significant CCN sources. Our study indicates, that with the right combination of stove and fuel, it is possible to significantly reduce the soot and PM emissions while maintaining pronounced emissions of highly hygroscopic ultrafine particles - depending on the fuel. However, stove costs, infrastructures and fuel quality, availability and sustainability have to be considered in the overall guidelines towards improved cookstoves. The observed strong impact of potassium on CCN emissions for a wide range of combustion conditions is likely to be of relevance not only to cookstoves but a wider range of biomass combustion including wild fires.”

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?

We do not state in the text that ‘the optimal scenario would be a reduction in both PM and CCN emission factors’ – it may be more complicated than that. The soot mode dominated the $PM_{0.5}$, and reduced emissions of soot particles can be desired from both a health and a climate perspective. It is less clear to which extent the more hygroscopic ultrafine particles may pose a significant health issue, and it may very well be that such emissions dominate the CCN population in large regions. Hence, significantly reduced emissions of the ultrafine CCN could potentially lead to a pronounced net warming of climate – and that may not be a desired effect.

See our previous response above for the changes made to the conclusion.

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

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The following has been added to the Fig. 1 caption:

‘The abbreviations of stoves and fuels in the legends are: 3-stone fire (3S), rocket stove (RS), natural draft stove (NDS) and forced draft stove (FDS) with combustion of birch (bir), casuarina (cas) or sesbania (ses) wood logs or pellets of softwood (sw), coffee husk (ch), rice husk (rh), water hyacinth (wh) or ses.’

In the Fig. 2 caption, ‘In (a), for the rocket stove (RS) and the 3-stone (3S) stove, in (b), for the forced draft pellet stove.’

Has been rephrased to:

‘In (a), for the rocket stove (RS) and the 3-stone (3S) stove with combustion of wood logs of birch (bir), casuarina (cas) or sesbania (ses). In (b), for the forced draft stove (FDS) and combustion of pellets of ses or softwood (sw) or mixtures of sw with pelletised coffee husk (ch), rice husk (rh) or water hyacinth (wh), respectively.’

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 low supersaturation.

The steps in emission factors are a results of cases where a large fraction of a soot mode activates within a small step in supersaturation. Further above, we provide explanations for such observations. The insets are included to illustrate differences at low supersaturations – supersaturations of atmospheric relevance.

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

The comment has been moved to the main text in sec. 4.6

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

That was changed already in the ACPD paper.

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

The following has been added to the Table caption: “The results were obtained from application of the standard protocols EN 14775, EN 15289, EN 15290 and EN 15297.”

References:

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