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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.

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We thank the anonymous reviewer for the 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 reponses appear in **bold** below.

The manuscript discusses the properties of cloud condensation nuclei (CCN) emitted from biomass burning of solid fuels (seven different fuels) in different cookstoves (four

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different stoves). This study covers particle number size distribution, mixing state, particle density, chemical composition, CCN activation and particle hygroscopicity properties at the same time. The measurement results offer valuable insights for field measurements and global models regarding biomass burning particles. Overall, the paper is well written and relevant to ACP. I recommend publication after the following comments are satisfactorily addressed:

Major comments:

1. There were a bunch of measurements clearly explained in the manuscript. But I think it would be nice to have an overview plot or measurement setup sketch. It should include biomass burning setup, chambers, sampling line, and instruments. It will help readers to understand your measurements better. A simple example can be found in Smith et al., (2019).

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

2. Did you consider the particle wall losses and particle loss inside the inlets (diffusion, deposition, etc.)? For example, the emitted aerosols were injected into a chamber for 10-40 minutes. What is the wall loss affection of the size distribution?

When it comes to potential particle losses, then different effects dominate in different particle size ranges.

Supermicron particles

Potential particles with sizes of $\backsim$1 μm and larger will not be captured in our set up, and while such particles may contribute significantly to the emitted particulate matter (PM), they are unlikely to influence the CCN population on a number

basis.

Potential diffusional particle losses
We did not correct for diffusional losses of the very smallest particles in the sampling lines, and there are two main reasons why we would not expect such potential losses to significantly influence the reported CCN results.

I: The fast particle analyser sampled from the flue gas with a high flow rate (8 slpm) to minimize diffusional losses, and II: diffusional losses are more pronounced for the very smallest particles, which are not large enough neither to act as CCN nor to contribute significantly to the estimated PM. Hence, we do not expect any minor particle losses to influence any of the main results presented.

Effects of aerosol storage
Aerosol storage in the chamber did lead to particle losses, but we do not expect that to influence the reported results, since the CCN and the effective density results were not sensitive to the absolute particle concentration of the studied quasi-monodisperse populations, and the associated spectra were obtained rather fast (2-5 minutes). Wall losses and coagulation effects posed a challenge when it came to studying the effect of photochemical ageing – after some time of aerosol storage. Thus, we only discuss the results of photochemical ageing of the soot mode on a qualitative level in the manuscript.

For clarification, we have added the following sentence:
"It is worth noting that none of the listed potential minor effects will have any substantial influence on the main results reported in this study." To the previous L. 277.

In addition, we have changed L.522-523 from:
"The characteristics of the experimental setup related to wall losses and coagulation (dilution rates) may to some extent influence the inferred CCN emission factors on a quantitative level."

To:
"The characteristics of the experimental setup related to losses in sampling lines and coagulation (dilution rates) may to some extent influence the inferred CCN emission factors on a quantitative level."

And a similar change has been made to L. 524-526.

Minor comments:
Line 205: I could not get why "aerosol particles present in the flue gas and initially injected into the aerosol storage chamber as freshly formed or primary, while particulate matter formed in the flow reactor will be considered secondary aerosol.". Could you please give more explanation about your definition of freshly formed and secondary aerosol?

Our definitions of primary and secondary aerosol particles are similar to the definitions applied in the literature (e.g. Obaidullah et al., 2012, Martin et al., 2013, Reece et al., 2017). We find that the inclusion of a schematic of the experimental set up has made our distinction between primary and secondary aerosol particles more clear in the revised manuscript.

Lines 241-242: Why the soot mode particle was unaffected in the storage for up to 60 minutes? Could you find previous studies that also support this? At least from your measurement, the aged 200 nm particles always had different (most probably higher) kappa values than primary particles.

The geometric mean of lognormal modes fitted to the soot mode sampled from the storage chamber did not change significantly over the course of the first 60 minutes after filling in any of the experiments. In some cases we observed a tendency of increasing kappa value of the 200 nm soot particles with time – which we associate with coagulation between ultrafine and soot mode particles occurring inside the chamber. For that reason, the reported kappa values for the

primary particles represent the very first measurements following the chamber filling.

The 'aged 200 nm particles' were actively aged via sampling through the oxidation flow reactor. Those aged particles always had higher kappa-values than the 200 nm particles sampled directly from the aerosol storage chamber in all the cases when ageing was simulated. Those observations are presented in Table 5. The effect of photochemical ageing was generally observed to dominate over potential coagulation effects for the reported ageing experiments. In order to clarify the type of ageing investigated, we have in the manuscript specified that we "simulated photochemical ageing" with the oxidation flow reactor.

Lines 269-270: It is good to adjust the PNSD for dilution rates and normalized to the corresponding consumption of dry fuel mass. I would suggest including the error bars in Fig. 1. The error bars could be 25th and 75th percentiles (with median lines) or one stand deviation (with mean lines). This will help us to understand the fluctuation of PNSD during the experiments.

When it comes to the particle number size distributions presented in Fig. 1, we have optimized it for easy intercomparison between different combinations of stoves and fuels. We find that essential, and it is not possible to obtain that with inclusion of percentiles for the many particle number size distributions presented.

Instead, we provide the requested information by addition of the following paragraph to Section 4.1:
"We have studied the particle number size distributions over the chamber injection time windows to provide some semi-quantitative information about the variability. The particle number concentration for the diameter size bin where maxima on average were observed for the ultrafine modes varied relatively by $\pm$28%, $\pm$17%, $\pm$9% and $\pm$6% for the 3S, the RS, the NDS and the FDS, respectively. The

reported variability ranges correspond to the range from the 25th to the 75th percentile. It was not possible in general to report similarly derived variabilities for the soot-mode-maxima due to less well-defined modes in many experiments. Instead we have looked into the variability in number concentrations for the size bin centered close to 150 nm. Those variabilities were about ±35%, ±50% and ±10% for the 3S, the RS and the NDS, respectively, again with the ranges corresponding to the 25th to 75th percentile range. The relative soot-mode particle number variability for the FDS-sw experiment was about 14%, while it was significantly more pronounced (20%-100%) for the other FDS experiments with very modest absolute soot mode particle number concentrations. Comparison between stoves should be carried out with precaution in this context, since the relevant time intervals, fuels and number of experiments with each stove varied. Nevertheless, we observed indications of more constant ultra-fine particle number emissions with increasing combustion temperature and improved combustion temperature-stability from the 3S to the FDS. Furthermore, it is worth noting that the soot mode variability appears to be more pronounced for the RS relative to the 3s, but additional studies are needed for a robust investigation of such potential differences. The pronounced variability in aerosol emissions in particular from the 3S and the RS illustrates the need for studying average emissions over a full combustion cycle for those stoves in order to assess the general aerosol emission features. Due to the described variability in emissions, we prioritised repeats of the 3S and the RS experiments as indicated in Table 1."

Lines 305-308: Did you see relative higher slopes of the CCN activation spectra for the FDS, sw and NDS, sw-ch? Clear ultrafine and soot modes were observed for these two types of biomass burning particles. There is an overlap of ultrafine and soot modes in the size range around 100 nm. If we assume the ultrafine and soot mode particles have different chemical compositions, a relatively higher slope would be expected.

We optimized the CCN measurements towards obtaining CCN spectra for quasi-

monodisperse aerosol populations very fast - in parallel with the APM measurements. We found that such an approach was essential in this study, and this approach does not provide a solid foundation for detailed studies of the CCN spectra for the complex aerosol populations studied. We invested a lot of time and effort into quality control of the CCN spectra, as described in the manuscript. This in order to ensure, that reported kappa values represented the majority of any given quasi-monodisperse aerosol population studied. We have attempted to extract additional information from the CCN spectra as described in the text. However, we do not find our data set adequate for such detailed analysis. We have added the following to the end of section 4.2: "However, it should be noted that the CCNc experimental approach was optimised for fast scans in parallel with the APM measurements. Hence, nor the DMA transfer function neither the CCNc operation were optimised for extracting information about the mixing state from the CCN spectra."

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