Review to “Ice nucleating ability of particulate emissions from solid biomass-fired cookstoves: an experimental study” by Korhonen et al. ACPD, 2019

The manuscript by Korhonen et al. presents a laboratory investigation of the ice nucleation ability of particles emitted by burning different biomass types (fuels) in different types of cookstoves. Both the cookstoves and the fuel material represent typical material used in sub-Sahran Africa.
The ice nucleation ability of the emitted soot particles was tested in a commercial continuous flow diffusion chamber (SPIN). Ice nucleation experiments were performed in two different sets of experiments, that the author refer to as transient and chamber experiments.
In the transient experiments, RH-scans were performed at T = -32 °C and -28 °C in both deposition and immersion/condensation freezing experiments. In the chamber experiments, T-scans were performed at constant RH_w = 115 %, measuring the ice nucleation activity in the immersion/condensation mode within a temperature range from approximately -32°C to -41 °C, using size-selected aerosol particles of different sizes. The setup further involves various instrument for particle characterization, such as an SP-AMS.
The main conclusion from the manuscript is that most of the soot types form ice at conditions slightly above those required for homogeneous freezing. Nevertheless, the statement in the conclusions that such particles emitted from biomass material in cookstoves can constitute a globally important source of INPs remains speculative in my opinion. A systematic comparison between the ice nucleation ability of the different soot particles sizes, fuel types and cookstoves types further remains difficult, given that not all fuel types and/or particle sizes were tested for all cookstoves. The authors also investigated different physico-chemical properties of the soot particles, but could not identify an evident correlation between those and the observed ice nucleation ability.
Overall, I find that the topic of the manuscript fits in the scope of ACP, given that the ice nucleation ability of anthropogenic aerosols such as soot remains an ongoing debate. Nevertheless, the manuscript in its current suffers from some inconsistencies (mostly technical), unnecessary repetitions and some issues that could be discussed in a clearer manner (see below). Overall, I suggest reevaluating the manuscript for publication in ACP after the points listed below have been critically addressed and incorporated.

General comments:

- In particular, the results sections contains some information that are required in the methods part. This hinders saliency of the most important findings when reading your Sect. 3. I encourage the authors to critically evaluate and improve the structure of the manuscript during revisions. For instance, a clearer separation of your transient and chamber experiments within Sect. 2.3 could be achieved by using subheadings. I would also encourage the authors to include a brief description of the auxiliary measurements made to characterize the particle physico-chemical properties in Sect. 2 and move the corresponding description of these there, which is currently in Sect. 3.2.
- Your current Sect. 3.2 is rather lengthy. I would consider breaking it up and have a separate section where you discuss e.g. your Fig. 7 (physico-chemical properties). If possible, please also add the particle characteristics for all your chamber experiments listed in Table 2. At the same time it remains unclear whether the “correlation” (L32, L412) is based on a visual inspection of the particle properties and the ice nucleation results, or whether it was quantified by e.g. calculating a correlation coefficient between the individual particle properties and e.g. the ice nucleation onset conditions discussed in the manuscript.
- It remains largely unclear to me how you estimate your “upscaling factors” based on your AS experiments, described in Sect. 3.3. This entire section needs to be improved to warrant a proper discussion of the biases of your SPIN results.

Specific comments:

- L24: I suggest to specify the sizes that were selected already in the abstract.
- L25: Please be more quantitative and add the exact temperature range used for SPIN experiments. “freezing conditions” is too vague.
- L32: Add “…and the observed increased ice nucleating ability.”
- L33: Change to: “…aerosols from ice via immersion/condensation freezing at temperatures only moderately above homogeneous freezing conditions.”

- L36: This statement should be followed by references. Consider adding: Korolev et al. (2017), Mülmenstädt et al. (2015).
- L36: "…Cloud droplets freeze…". This statement should be followed by a reference. You might want to give reference to standard textbooks, such as: Pruppacher and Klett (1997), Lohmann et al. (2016), Lamb and Verlinde (2011).
- L37: Please check the symbol of your degree Celsius units (°C), it seems off here and at other places in the text, e.g. L45, L64…
- L38: Please specific what MPC properties you mean. Do you mean radiative properties? See e.g. Matus and L'Ecuyer (2017).
- L41: Add: "number concentrations at the…"
- L43: You might want to add some never references giving overviews of INPs, e.g.: Kanji et al. (2017). At the same time, I think you should be more specific and give references of those studies that find soot to be a source of INPs, because citing Hoose and Möhler (2012) in this context seems to be in conflict with your citation on L46, please clarify.
- L46: Why do you only refer to immersion freezing? I think it would be better to say "immersion/condensation freezing" and more consistent with your usage further down, e.g. L64, and also with your statement on L224.
- L47: Change to: "…parameterisations used to estimate the soot-INP number concentration…"
- L49: It is not clear to me what you mean. Is the uncertainty 1 Wm^{-2}? Is this just the uncertainty of the indirect effects of soot particles on MPC, or are indirect effects on other cloud types (e.g. cirrus) also considered? Is this the *effective* radiative forcing (ERF), i.e. are fast adjustments included? It would be better practice to state the "best estimate" or "mean" ERF_{aci} along with the uncertainty bounds and then make the statement that this is uncertain.
- L51: You also might want to cite: Bond et al. (2013)
- L53: Specify "… from incomplete combustion of fossil fuel or biomass burning. Properties of ambient soot particles…". Also add: Koehler et al. (2009), Mahrt et al. (2018)
- L53: Please further add references for how soot particle properties can change upon atmospheric aging. A study of interest could be: Bhandari et al. (2019).
- L54: Add: Ferry et al. (2002)
- L56: Please specify what you mean with "nanostructure".
- L60: Please quantify "significant source of particulate matter"
- L62: There is also evidence that particles from biomass burning are a source of INPs in cirrus clouds (e.g. DeMott et al. (2009)), which might be worth mentioning, given that there is increasing evidence, also from your paper, that any sort of combustion particles are an important source of INPs mainly (or only) in the cirrus regime at T < -38 °C.
- For the discussion of soot immersion freezing, you might want to add the following references to your discussion: Popovicheva et al. (2008), Brooks et al. (2014)
- L65: Change to: "has been observed to be associated…"
- L66: Change to: "…produced particles that acted as INPs at T = -35 °C, but not at T = -30 °C…"
- L68: Delete "they" and specify what you mean with "potentially significant climate impact".
- L69: This goes back to my comment on L60: Can you quantify the amount of PM emitted from biomass used for cooking vs. that emitted from natural forest fires, or at least give estimates of the order of magnitude?

- L71: What do you mean with “climate-forcing”? Do you mean a positive radiative forcing (warming)?
- L80: Add comma after cookstoves
- L89: Do not use capitalized letters for “water boiling test”, to be consistent with L82 and change to “…(WBT, version 4.2.4), …”
- L91: Change “minutes” to min for consistency
- L92: It remains unclear whether the 45 min is the duration of the “simmering phase”or the time when the simmering phase starts, please clarify.
- L93: Please specify how this “height” affects your sampling. Does this height correspond to the orange squares in your Fig.1?
- L95: Change to “..diluted by 1:100..”
- L96: Change to “…of the analyzers.”
- L104: Replace + by “and”
- L105: Please add the neutralizer type and the aerosol to sheath flow ratio you have operated your SMPS at.
- L118: Change “chapter” to Sect.
- L122: Change “experimentation” to experiments
- L124: Change to “…was approximately 2h before…”
- L124: How do you keep the particles suspended in the 15 m^3 chamber? Is there a fan at the bottom?
- L129: The mobility diameters of size selected particles for the IN experiments differs to those used for the CCN experiments (L109). Is there a reason for this?
- L134: Delete “simulated”
- L137: Change to: “Africa, which are commonly used…, and that represent regionally…”
- L138: Add: “For instance, the Casuarina…”. Also, this statement should be followed by a reference.
- L148: Add: “located in the sub-…”
- L149: What do these “agricultural residues” encompass? Tree branches and leafs? Please clarify.
- L150: Consider rephrasing to: “…and investigations of using these residues to support the sustainable development of sub-Saharan are ongoing.”
- L162: Change to: “, when forming cereal products”
- L165: Are you trying to say that the comparability results from comparing the African biomass fuels to something that has previously been studied and characterized, such as SW? Please clarify.
- L171: Change to “… (3S), which is a three…”
- L180: Replace version by gasifier stove
- L189: “more than five“ is pretty arbitrary, I suggest to give a typical range here.
- L192/93: INP is already defined. I suggest to reformulate to: “Ice nucleation experiments where conducted with the spectrometer for ice nuclei (SPIN, DMT Inc., Colorado, USA), described by Garimella et al (2016).” There is no need to say since when it is operational in my eyes.
- L195: Please elaborate to what extend the temperature control was improved and how this affects your RH-uncertainty during your RH-ramps.
- L196: CFDC is already defined on L83.
- L196: Delete “with an IN-chamber design”
- L198/99: The abbreviations PINC and ZINC should be given in brackets.
- L200: Replace “ca.” by approximately here and at other instances in text please.
- L203: Replace “before” with upstream
- L203: Add: “Therefore, the temperature of the…”

- L205: Write out: "…temperature, relative humidities and the path…"
- L210: Please specify where the background number comes from. I assume you refer to the OPC counts, right? Which optical particle size does this correspond to and how does this compare with the optical particle size used for calculating your activated fraction?
- L216: Delete "therefore". Also, equations should be used along normal/main text punctuation.
- L220: Add: "by the OPC, taking into account the dilution resulting from aerosol to sheath flow within SPIN…"
- L221: Usually the background measurement in CFDC measurements using RH-scans take place before the RH-scan, i.e. at low RH and after the RH-scan, i.e. at high RH. Is this how you corrected your sample? Please specify.
- L222: I would delete this statement here and simply add the Vali et al. (2010) reference to L223: "… is referred to as immersion freezing (Vali et al. 2010), …"
- L224: Change to: "… is expected prior to freezing."
- L229: Diluted from what initial number concentration?
- L230: This type of experiments is commonly referred to as "RH-scans" in the CFDC community.
- L231: Why do you say "scanning" here? Rephrase the sentence to: "were sampled at constant T= -32°C, scanning the RH_i…". Furthermore, it remains unclear why you chose -32°C for your RH-scans. This should be added in the text, along with a statement on the atmospheric relevance.
- L233: Why do you say "approximately" here but not before? If you have a fixed T and you know your RHi, then you can give "exact" RHw boundaries.
- L234: Delete: "and the only difference was in higher lamina temperature." This is clear the moment you state your T is -28 °C.
- L234/235: Delete statement or give reference to other studies.
- L237: Add : "combustion aerosol"
- L237: Specify "highly diluted" and give number concentration instead.
- L241: "T-ramp"
- L243 Why is the timer period for the first and the second background different (5 vs. 3 min)?
- L244: Do you mean -37.9 °C?
- L244: Why do you not show the activated fraction in the color bar of your Fig. 1b? This would be easier, as you use the activated fraction in the text. Also, having the x-axis of your Fig. 1b as lamina temperature would be more intuitive and would save you Fig. 1a, which in my eyes does not add much to the main findings of your study.
- L251: Replace "upward" by ascending for consistency
- L252: Change to: "Sect. 3.4"
- L236-252: It does not become entirely clear in this paragraph, but when looking at your Figs. 4, 5, 6 it looks to me that you fixed the lamina RH_w of SPIN and then scanned different T, right? This can be referred to as "T-scan". This must become much clearer for the reader.
- L258: See my comment above: Showing your activated fraction or OPC counts as a function of lamina T, rather than time would be more appropriate.
- L245: How does this threshold of 4.5 microns compare to theoretical condensational growth calculations? Also, I do not understand why you need to discriminate these particles types, when using an evaporation section, as you describe on L203, please clarify. Do you refer to water droplet survival beyond the evaporation section?

- L259: Why are you using 6 microns as size threshold for ice crystals, if you argue above that the cloud droplets do not grow larger than 4.5 microns? Do you not introduce another bias then to your reported activated fractions? Please clarify in the text.
- L265: “since the ice crystal mode…” You might want to refer back to your Fig. 1 here.
- L269: Please specify “in a later section”
- L275: Delete “in real life”
- L276: Please clarify whether your ice counts are corrected by this factor. Which?
- L280-283: “Ice onset is defined..” This statement should be moved to the description of the methods. Moreover, I find your terminology of “negative” and “positive” results quite misleading. For your T-scan experiments (fixed RH): In case you observe ice formation at T > -38°C (or your experimentally defined homogeneous freezing threshold), refer to it as heterogeneous ice formation. Similarly, in your RH-scan experiments that take place at T = -32 °C, you are above the homogeneous nucleation temperature, so any ice formation you observe should be heterogeneous.
- L286: Please define “ultrafine”, also for usage further down, e.g. L329.
- L289-L291: You need to elaborate on various statements made here:
    - The number concentration introduced into SPIN seems very high. Considering your aerosol to sheath flow ratio, you still have around 250 #/cc entering the OPC downstream of SPIN. What is the upper number concentration that can be sampled without co-incidence error?
    - It remains unclear how and why the detection limit depends on the particle number concentration introduced into the chamber, as you write in the caption of your Tab. 1. Could it be that at higher number concentrations introduced into SPIN you have relatively more particles > 250 nm (> 5%, as stated on L287) that become detected in your OPC? Please see also my comment to L210.
    - Your detection limit of the order $\alpha = 10^{-6}$ seems very low. It would be helpful to add an example plot for one of these experiments, showing α as a function of lamina RH in SPIN to the appendix. That way, the reader could more easily see at what RH you just sample noise and/or aerosol particles and at what point you actually start to see an ice signal. If $\alpha = 10^{-6}$ is your true detection limit, I suggest to more clearly write this in your statement on L211.
- L291: “Ice crystals…”. This statement is not true looking at your “FD gasifier SW” experiment, if your threshold is $\alpha = 10^{-6}$.
- L291-294: Improve statement by saying: “”… four stoves do not nucleate ice in neither deposition nor immersion/condensation mode…”
- L297-318: This description should all be moved into your Sect. 2, where you describe your experimental methodology. Also, avoid unnecessary repetitions such as the mass concentration in the chamber (L304 vs. L122) to improve readability.
- L323: Your statement on the soot sizes selected and your typical number concentrations differs from what is given on L129, please clarify. Furthermore, the number concentration of 50-200 #/cc significantly differs to the number concentration used for the transient experiments (see L290), please comment.
- L323: Please quantify the “minor contributions of multiply charged particles”, so that the reader can judge on how your reported activated fractions are affected. See Wiedensohler (1988). I suggest adding sizes and fractions of double charged particles to your Table 2.

- L324: "...which indicates that soot…" So do you only consider particles > 250 nm as soot, please clarify. If so, what are all the other particles? Please see also my comment on L323, where you say that you investigated 200 nm particles.
- L329: Replace "when" by "after"
- L331: Replace "Chapter" by "Sect." Please read the guidelines of ACP here: https://www.atmospheric-chemistry-and-physics.net/for_authors/manuscript_preparation.html
- On the same note of abbreviations, please also use "Fig." instead of "Figure" throughout the text.
- L333: Change to "ice activation", to be consistent with e.g. L336. In any case, please check for consistent spelling throughout manuscript. Moreover, I feel this paragraph seems to refer to your Table 3, right? In this case, having a reference to this table at the beginning of this discussion might be helpful.
- L333: Please be more careful in specifying which of the 3S experiments you talk about. For instance, the 3S on 500 nm CAS (see Table 3), shows that the ice onset is detected at T = -37.8 °C, rather than at -38.1 °C, as you write on L333.
- L337: "It is not clear at which temperature…" This statement seems misleading. Is this not why you do your AS calibration results, to determine homogenous freezing conditions?
- L339-L341: Why do you talk about the biases of the activated fraction here, when above (L330) you say that this is discussed in Sect. 3.4. I suggest to move this to the discussion around L436. Please improve the structure.
- L342: Please elaborate how you estimate this correction activated fraction of 10^{-3} for the temperature interval indicated.
- L349: Replace "as was done by" by "following"
- L349: "The uncertainty…" This sentence is a direct repetition of the previous statement, please delete.
- L353: Change "experiment emissions" to "experiments" and "high experiments" by "Good"
- L354: Change to "Sect. 3.4"
- L355: This statement needs to be clarified:
    - I cannot infer any size dependence from your Fig. 4. In other words, all the experimental curves for the 3S experiments are within T-uncertainty of each other. I encourage you to also add α-uncertainty bars, maybe for every third data point for clarity/visibility.
    - Can you verify a difference in chemical composition between the runs using the SP-AMS, you indicate in your Fig. 1? If there is a chemical difference between the 3S CAS 300 nm and 500 nm experiment, it is meaningful to compare the ice nucleation activity of different sizes of these aerosol types?
- L358: Add "than the ones…"
- L359: I cannot find the number "-37.7°C" in the RS experiments in your Tab. 3, please check.
- L361: "…and conditions for 10^{-3} ice-activated fraction were reached at -37.8 °C…" I think you do not need to repeat your ice nucleation onset activated fraction in every other statement. Just referring to "ice nucleation onset" would be fine (after you clearly defined it) and would improve the reading flow of your manuscript by a lot.
- L365: I would tune this down a little bit and say something along the lines of: "show that the soot particles emitted from burning the various fuels can act as heterogeneous INP at temperatures only slightly higher than those needed for homogeneous freezing of solution droplets."

- L368-377: “This indicates that very minor changes in combustion conditions significantly influence the ice nucleating ability of freshly produced soot particles.” I very much agree and I think this is a great finding, however, I think this paragraphs deserves some more clarifications:
    - You say that you block one air supply, so your combustion should become less efficient, i.e. the organic carbon fraction of your soot particles should increase. Do you observe this in your SP-AMS data?
    - On L371 you say that you varied the pot height (see also L92). However, it remains unclear, how this was changed, whether this was a systematic change and how this affects the combustion process:
        - For instance, I find it interesting that you seem to see a very strong change in ice nucleation ability between “FDGS, mod. 400 nm #1” and “FDGS, mod. 400 nm #2” in your Fig. 6 (which you interpret as change in chemical composition due to different combustion conditions), but hardly no change between “FDGS, mod. 400 nm #1” and “FDGS, mod. 500n m”. Can you explain this?
        - On the same note there is a strong size dependence for the 450 nm and 500 nm FDGS samples with blocked secondary air supply, but no size dependence between the “FDGS, mod. 400 nm #2” and the “FDGS, mod. 500nm”. Can you elaborate on this?
        - Looking at your Fig. 7 it is also interesting to note that the FDGS samples with blocked are supply for the 450 and 500 nm case seem to be quite different in their properties, e.g. Org/BC. How reproducible are your soot test points? In the end, what looks like a size dependence in Fig. 6, might also be caused by differences in physico-chemical properties.
- L382: Why is the effective density of 350 nm particles representative for your aggregates of different sizes? The effective density is a strong function of particle size, see e.g. Olfert et al. (2017), as you also note on L392.
- L383: There is no Fig. 8.
- L383-L391: This should be mentioned in your methods section along with a more detailed description and/or reference, how you calculate the chemical properties from the SP-AMS data, not in the results section.
- L391: “The values of…” Is this true for any soot aggregate size? Please specify.
- L396: “OA” is not defined. You might also want to use “OA” instead of “Org” in your Fig. 7.
- L396: Should not K400#1 have a higher UV absorption, given the high Org carbon fraction?
- L412: Can you quantify this correlation?
- L416: “Further studies…” I suggest to move this statement to your Sect. 4.
- L418: “The typical…” This is a repetition from L250 and can be deleted.
- L425: Change “Chapter” to “Sect.”
- L426: Replace “to” by “the”
- L428: This is a direct repetition of L346 and can be deleted.
- L430-434: I think that it would be best practice to report the error corresponding to the largest uncertainty, i.e. the span in T across the lamina (“T profile”) rather than the variation in mean lamina T. See e.g. Garimella et al. (2017)
- L441: What does “somewhat similar operation conditions” mean? You should clarify this.
- L445-461: This paragraph need to be significantly elaborated and improved in order to justify a proper discussion of biases in ice crystal detection.

    - You list a bunch of reasons for underestimating you activated fraction (e.g. choice of 6 µm channel for ice detection), but you lack to quantify the contributions of this to the activated fractions reported here.
    - It remains unclear why the "underestimation depends on lamina temperature".
    - How do you arrive at the upscaling factors of 3-5 and 5-10? Please add this to the text.
    - Finally, please be more quantitative when saying "in a neighborhood of -40 °C"
- L468: "This study shows…" It remains unclear whether this statement refer to all different cook stoves tested or not.
- L469: "500 million people…" This should be followed by a reference and I would move this statement to the introduction, as it does not constitute a conclusion from your study.
- L471: "…showed good performance". This statement is misleading, given that your maximum activated fractions even for the AS experiments are 90% below the theoretically expected values, please be more specific.
- L475: It would be good to quantify "poor IN activity" (not INP activity!) by giving a range of observed activated fractions.
- L481: "Therefore…" Is this really true? Do all the cook stove emission be transported upwards? What about the atmospheric lifetime of these aerosols and what are the cloud properties that you say become changed?
- L487-489: This comparison is very loose and insubstantial and requires a more adequate discussion and comparison of dust/soot emissions, (vertical) burdens, atmospheric lifetimes and many more factors. In the end, your results show that most of the soots can nucleate ice only very close to conditions required for homogeneous freezing. As such, the impact of these soot types on warm or MPCs is very likely absent and/or negligible. In fact, when one takes -38 °C as a "general threshold" for homogeneous freezing most of your soots in Figs. 4 and 5 freeze homogeneously
- L490: Delete "of"
- L499: What do you mean by "slight differences"? Which of the "studied properties" were different? Please be more precise and quantitative here, otherwise it is hard for the reader to take out the main findings of your study.
- L761: Change "," to "." In front of "The two…"
- L767: The dependence of the detection limit on the sample concentration should be discussed in the main text. If this is the case, it would also be meaningful to add the concentrations for the individual experiments listed here.
- L779: Add "are equal…"

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