Author response to comments by Referee #1

We thank Referee #1 for his/her work on the manuscript, and comprehensive comments and suggestions on revisions how to improve the contents. The detailed responses (written on red font) to comments as they were posted (green font) are listed below – unless the response includes how the text has been revised by itself, the revisions to the text are marked separately on blue font.

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.

We acknowledge the mentioned structural shortcomings and have moved all methodology information to Sect 2. We have also shortened lengthy sections and added subsections, separating the descriptions of the CFDC, ice nucleation experiments (transient and chamber) and particle characterization, each to their own respective section.

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.

We agree and have revised the text, adding a separate section for emission particle properties.

There is a tendency of the two more ice-active samples having a relatively lower ratio. However, the differences between the samples are not pronounced considering the errors, and we do not observe a significant correlation with this limited data set.

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.

Please see our responses to the specific comments relevant to discussion on the upscaling factor.

Specific comments:

- L24: I suggest to specify the sizes that were selected already in the abstract.

A statement that specifies the sizes has been added:

"The soot particles in nm were 250, 260, 300, 350, 400, 450 and 500."

- L25: Please be more quantitative and add the exact temperature range used for SPIN experiments. "freezing conditions" is too vague.

The text has been changed from:

"to water-supersaturated freezing conditions in the SPIN."

to:

"introduced to water-supersaturated freezing conditions (-32 °C to -43 °C) in the SPIN."

- L32: Add "…and the observed increased ice nucleating ability."

Statement added.

- L33: Change to: "…aerosols from ice via immersion/condensation freezing at temperatures only moderately above homogeneous freezing conditions."

Done.

- L36: This statement should be followed by references. Consider adding: Korolev et al. (2017), Mülmenstädt et al. (2015).

References added.

- 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).

A reference to Pruppacher and Klett (1997) added.

- 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…

Symbols checked and corrected throughout the manuscript.

- L38: Please specific what MPC properties you mean. Do you mean radiative properties? See e.g. Matus and L'Ecuyer (2017).

A wide range of cloud properties are influenced by introducing an ice phase in a liquid phase supercooled cloud. For example, the formation of ice releases latent heat, the presence of an ice phase influences the vapour pressures, and mixed-phase clouds are not in thermodynamic equilibrium. The growth of ice crystals on the expense of liquid droplets influence the cloud optical properties. To clarify this, we have modified the text (L38) from:

"MPC properties and lifetime are very sensitive to the formation of solid ice,"

To:

"A wide range of MPC properties including radiative properties and lifetime are sensitive to the formation of solid ice (Matus and L'Ecuyer, 2017),"

- L41: Add: "number concentrations at the…"

The text has been changed from:

"but there are significant gaps in our knowledge regarding these important ice formation processes."

to:

“but there are significant gaps in our knowledge regarding INP concentrations within the MPCs and these important ice formation processes.”

- 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.

Suggested reference added.

It is outside the scope of our study to review all experimental ice nucleation studies involving soot particles because there are too many of them and it would only be of value providing information about the particles studied. Hoose and Möhler (2012) provided a nice overview of many studies including positive as well as negative ice nucleation results for soot particles, and the reader can find further information there. Hence, there is no conflict in using that citation in both contexts: soot particles showing no or a high heterogeneous ice nucleating ability in the immersion/condensation mode. We have chosen to cite three specific studies with positive results in L43-45, and quite many additional studies in other parts of this manuscript.

We have added the Levin et al. (2016) study to L65, and thus we cite all relevant immersion/condensation mode ice nucleation studies on biomass burning aerosol including soot particles, which we know of in L. 62-67.

We do find that the references included in the revised manuscript are balanced according to the aim and context of our study – without citing all ice nucleation studies on soot particles directly.

- 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.

Added “immersion and condensation freezing” according to suggestion.

- L47: Change to: “…parameterisations used to estimate the soot-INP number concentration…”

Done.

- 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” ERFaci along with the uncertainty bounds and then make the statement that this is uncertain.

What we wish to emphasize in the context of our study, is that the current climate models are highly sensitive to the ‘choice’ of the ice nucleating ability of ambient soot particles. The range in RF (0.111 to 1.059Wm−2) reported by Yun et al. (2013) reflect the result of a low versus a high ice nucleation impact of ambient soot particles on MPCs. We have modified the text from:

”The radiative forcing associated with the impact of soot particles on MPCs has been reported to be very uncertain and potentially up to about 1 Wm-2 (Yun et al., 2013).”

To:

“The radiative forcing associated with the impact of fossil fuel soot particles on MPCs has been reported to range from about 0.1 to about 1 Wm^{-2} depending on their ice nucleating ability, which is uncertain (Yun et al., 2013).”

- L51: You also might want to cite: Bond et al. (2013)

Reference added.

- 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)

The text has been changed from:

Soot particles are produced from incomplete and ambient soot particle properties are highly variable and influenced by the combustion conditions and atmospheric ageing (Corbin et al., 2015).

to:

Soot particles are produced from incomplete combustion (combustion with insufficient oxygen supply) and ambient soot particle properties, such as morphology and chemical composition, are highly variable and influenced by the combustion conditions and atmospheric ageing (Ferry et al, 2002; Popovicheva et al., 2008; Koehler et al., 2009; Corbin et al., 2015; Mahrt et al., 2018; Bhandari et al., 2019).

- 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).

Bhandari et al., 2019 added to references. Please see previous response regarding L53.

- L54: Add: Ferry et al. (2002)

Ferry et al., 2002 added to references.

- L56: Please specify what you mean with “nanostructure”.

The nanostructure refers to characteristics on a ‘nano-scale’ such as fringe length and structural order. Häusler et al. (2018) show that the ordering of graphene on a nanoscale influences the ice nucleating ability. We have modified the text (L56-57) from:

“Chemical groups on the soot particle surface with the ability to form hydrogen bonds with water molecules are likely to be of importance (Gorbunov et al., 2001) as well as the soot particle nanostructure (Häusler et al., 2018).”

To:

“Chemical groups on the soot particle surface with the ability to form hydrogen bonds with water molecules are likely to be of importance (Gorbunov et al., 2001). In addition, the soot particle nanostructure may also be of importance, with highly ordered graphene structures being more efficient in supporting ice nucleation relative to lowly ordered graphene structures (Häusler et al., 2018).”

- L60: Please quantify “significant source of particulate matter”

The text has been changed from:

“Biomass combustion for cooking is an important global source of energy, being the major environmental health risk worldwide, as it is a significant source of particulate matter (PM) and soot particles on regional to global scales (Lim et al., 2012; Bonjour et al., 2013).”

to:

“Biomass combustion for cooking is an important global source of energy, being also the major environmental health risk worldwide (Lim et al., 2012). It has been estimated that approximately 2.8 billion people depend on solid fuel combustion for daily cooking worldwide, mostly in the developing part of the World (Bonjour et al., 2013). Therefore, solid biomass combustion is a significant source of particulate matter (PM) and soot particles on regional to global scales.”

- 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.

DeMott et al. (2009) reported all freezing events to be indistinguishable from homogeneous freezing. Hence, we do not find direct evidence in that study of such a link. However, the findings presented in that study do not exclude potential heterogeneous freezing e.g. for the aged aerosol biomass burning particles.

- 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)

References added to respective relevant contexts.

- L65: Change to: “has been observed to be associated…”

Done.

- L66: Change to: “…produced particles that acted as INPs at T = -35 °C, but not at T = -30 °C…”

Done.

- L68: Delete “they” and specify what you mean with “potentially significant climate impact”.

The text has been changed from:

“Huang et al. (2018) modeled the indirect climate impact of solid fuelled cookstove aerosol emissions, and they reported a potentially significant climate impact from emitted soot particles on MPC conditions and thus climate.”

to:

“Huang et al. (2018) modeled the indirect climate impact of solid fueled cookstove aerosol emissions and reported a potentially significant climate impact in increase of high clouds in their model runs where black carbon (BC) particles acted as INPs. Therefore, soot particle emissions can potentially affect MPC conditions and thus climate.”

- 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?

Such estimates are presented by Huang et al. (2018), to which the statement refers to. According to their study, the black carbon (BC) emissions from biomass-fired cookstoves are globally 2.31 Tg yr^{-1}, which accounts for 23.7% of total BC emissions. Primary organic matter (POM) emissions from cookstoves account for 21% of total global POM emission budget. We consider this an appropriate quantification of global cookstove emissions.

- L71: What do you mean with “climate-forcing”? Do you mean a positive radiative forcing (warming)?

Yes, the statement is corrected from:

“However, an efficient utilization of modern biomass fuels in efficient biomass cookstoves can constitute an alternative to mitigate climate-forcing aerosol emissions and household air pollution (HAP) worldwide (Carvalho et al. 2016).”

to:

“However, an efficient utilization of modern biomass fuels in efficient biomass cookstoves can constitute an alternative to mitigate household air pollution (HAP) worldwide, and aerosol emission effect on radiative forcing (Carvalho et al. 2016).”

- L80: Add comma after cookstoves

Done.

- L89: Do not use capitalized letters for “water boiling test”, to be consistent with L82 and change to “…(WBT, version 4.2.4), …”

Done.

- L91: Change “minutes” to min for consistency

Done.

- 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.

Yes, the statement is corrected from:

“A typical simulation time was 60-85 min in total, including two phases of the WBT process: cold start (15-40 min) and simmering (45 min).”

to:

“A typical simulation time was 60-85 min in total, including two phases of the WBT process: cold start (phase duration 15-40 min) and after that simmering (duration 45 min).”

- L93: Please specify how this “height” affects your sampling. Does this height correspond to the orange squares in your Fig.1?

Figure 1 shows the Three-stone fire as an example, and the orange squares do represent bricks on which the cooking pot was placed during the boiling tests. For other cookstoves, the cooking pot was normally placed on top of the cookstove: this type of operation represents usage of the cookstoves in the way they have been designed to, by manufacturers of commercially available cookstoves (Rocket stove, ND gasifier and FD gasifier). The three special experiments revealed that changing the height from designated one can increase the particle emissions, as L311-315 stated later in the text.

- L95: Change to "..diluted by 1:100.."

Done.

- L96: Change to "…of the analyzers."

Done.

- L104: Replace + by "and"

Done.

- L105: Please add the neutralizer type and the aerosol to sheath flow ratio you have operated your SMPS at.

The sentence describing the SMPS system has been changed from:

"An SMPS system (classifier, TSI 3082 + condensation particle counter, TSI 3775) measured particle size distribution and concentration in chamber experiments at scan range from 15.7 nm to 615.3 nm, with continuously repeated 180-second scans during aerosol injection and sampling."

to:

"An SMPS system (classifier TSI 3082 equipped with aerosol neutralizer TSI 3088, and condensation particle counter, TSI 3775) measured particle size distribution and concentration in chamber experiments at scan range from 15.7 nm to 615.3 nm, with continuously repeated 180-second scans during aerosol injection and sampling at an aerosol to sheath flow ratio of 0.3 vs. 3 litres per min, respectively."

- L118: Change "chapter" to Sect.

Done.

- L122: Change "experimentation" to experiments

Done.

- L124: Change to "…was approximately 2h before…"

Done.

- L124: How do you keep the particles suspended in the 15 m3 chamber? Is there a fan at the bottom?

Yes, there was a fan at the bottom of the storage chamber to ensure mixing. Gravitational settling was not a significant issue for the stored aerosol populations within the time frames of these experiments of typically less than 3 hours.

- 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?

Yes. The CCNC used a fixed scheme for electrical mobilities 65 nm, 100 nm, 200 nm and at times 350 nm when available in sufficient number concentrations. The IN experiments always aimed for sampling the largest particle size that was available in number concentrations larger than 30 cc^{-1}, which was stated on L322.

- L134: Delete “simulated”

Done.

- L137: Change to: “Africa, which are commonly used…, and that represent regionally…”

Done.

- L138: Add: “For instance, the Casuarina…”. Also, this statement should be followed by a reference.

Reference added. The text has been changed from:

“The *Casuarina equisetifolia* (CAS), commonly known as Horsetail tree, is a species which is commonly used in agroforestry systems in the southern part of the African continent.”

to:

“For instance, the *Casuarina equisetifolia* (CAS), commonly known as Horsetail tree, is a species which is commonly used in agroforestry systems in the southern part of the African continent (Potgieter et al., 2014).”

- L148: Add: “located in the sub-…”

Done.

- L149: What do these “agricultural residues” encompass? Tree branches and leafs? Please clarify.

The statement is corrected from:

“During processing, agricultural residues form up to 50% of the total weight of coffee products (Oliveira and Franca, 2015) and the usage of these residues as biofuels is under research for potential sustainable development in sub-Saharan Africa.”

to:

“During processing, agricultural residues such as tree branches and leaves form up to 50% of the total weight of coffee products (Oliveira and Franca, 2015) and the usage of these residues as biofuels is under research for potential sustainable development in sub-Saharan Africa.”

- L150: Consider rephrasing to: “…and investigations of using these residues to support the sustainable development of sub-Saharan are ongoing.”

Done.

- L162: Change to: “, when forming cereal products”

Done.

- 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.

Yes, the SW pellets are a standardized product whose characteristics, such as composition and origin of raw material, are known.

- L171: Change to “… (3S), which is a three…”

Done.

- L180: Replace version by gasifier stove

Done.

- L189: “more than five“ is pretty arbitrary, I suggest to give a typical range here.

The statement is corrected from:

“A typical number of re-fueling events was more than five.”

to:

“A typical number of re-fueling events was between five and ten.”

- 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.

Statement reformulated according to suggestion and mentioning about the operational period has been removed.

- L195: Please elaborate to what extend the temperature control was improved and how this affects your RH-uncertainty during your RH-ramps.

The main improvement was that the ice nucleation chamber was re-designed to reduce temperature variation on different spots on its walls, and thus improve control of the lamina temperature and RH. Also additional thermocouples that monitor the temperature on different were installed, which enabled optimized temperature and *RH* control during the scans.

The statement is corrected from:

“The main difference relative to the older version used by Ignatius et al. (2016) is improved temperature control.”

to:

“The main difference relative to the older version used by Ignatius et al. (2016) is improved temperature control via e.g. by an increase in the total number of thermocouples from 8 to 32 in the main chamber (growth section).

- L196: CFDC is already defined on L83.

Corrected.

- L196: Delete “with an IN-chamber design”

Done.

- L198/99: The abbreviations PINC and ZINC should be given in brackets.

Corrected.

- L200: Replace “ca.” by approximately here and at other instances in text please.

Corrected throughout the manuscript.

- L203: Replace “before” with upstream

Done.

- L203: Add: “Therefore, the temperature of the…”

Done.

- L205: Write out: “…temperature, relative humidities and the path…”

Done.

- 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?

The following specification has been added to the text:

“Here, the background signal refers to unwanted OPC counts in the ice crystal size range, which may be due to break up of frost on the ice-covered plates or alternatively due to tiny leaks in the instrument chamber.”

Typical background signal counts due to cracking ice layer represent particles that are larger than 6 µm in optical size, therefore the chamber was re-iced when the background signal exceeded 10-15 particles per litre, as is stated on L211.

- L216: Delete “therefore”. Also, equations should be used along normal/main text punctuation.

Done. “:” added.

- L220: Add: “by the OPC, taking into account the dilution resulting from aerosol to sheath flow within SPIN…”

Done.

- 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.

Yes, the background check was carried out just before and just after each RH scan, respectively. Linear interpolation was used to estimate the background during scans.

- 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), …”

Done.

- L224: Change to: “… is expected prior to freezing.”

Done.

- L229: Diluted from what initial number concentration?

Typical number concentrations after ejector dilution by 1:100 were on the order of 10^5-10^6 cm^{-3} that would had been excessive for a CFDC instrument.

- L230: This type of experiments is commonly referred to as “RH-scans” in the CFDC community.

Mentioning about RH-scan added.

- L231: Why do you say “scanning” here? Rephrase the sentence to: “were sampled at constant T= -32°C, scanning the RHi…”. 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.

A sample temperature of -32°C is commonly applied in CFDC studies (e.g. Levin et al., 2016). Measurements at relatively low temperatures improve the signal to noise ratio, and when no INPs are detected for such conditions – we can rule out significant INP concentrations at higher temperatures relevant for MPCs.

The text has been changed from:

“The measurement sequence used in the transient experiments was as follows:”

to:

“*RH*-scans were used for the transient experiments at constant *T* = -32°C, scanning the *RHi* from 100 % up to 160 %, which is close to the procedure used by Petters et al. (2009) and Levin et al., (2016).”

- 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.

Word “approximately” deleted.

- L234: Delete: “and the only difference was in higher lamina temperature.” This is clear the moment you state your T is -28 °C.

Done.

- L234/235: Delete statement or give reference to other studies.

Added references to Petters et al. (2009) and Welti et al. (2009), who both applied the RH-scan in their CFDC experiments.

- L237: Add : “combustion aerosol”

Done.

- L237: Specify “highly diluted” and give number concentration instead.

The term ‘highly diluted’ refers to the ammonium sulfate concentration in the formed cloud droplets (diameters in super-micron range with a seed diameter of 350 nm) in the upper part of the chamber. We suppose that the reviewer suggests including the sample particle number concentration.

The text has been changed from:

“This procedure is illustrated in Fig. 2, where a homogeneous freezing experiment on highly diluted ammonium sulfate (AS, dry mobility diameter of 350 nm) droplets is used as an example.”

to:

“This procedure is illustrated in Fig. 2, where a homogeneous freezing experiment on highly diluted ammonium sulfate (AS, dry mobility diameter of 350 nm) droplets were introduced to the SPIN in number concentration of approximately 150 cm^{-3}.”

- L241: “T-ramp”

Added.

- L243 Why is the timer period for the first and the second background different (5 vs. 3 min)?

The reason for this was that before the T-ramp, the CFDC was running on initial settings and sampled filtered air prior to start for at least 5 minutes before the T-ramp was initiated. The statement has been changed from:

“First, the internal background was checked via sampling filtered dry air for 5 minutes before the T-ramp began.”

to:

“First, the internal background was checked via sampling filtered dry air for at least 5 minutes before the T-ramp began.”

- L244: Do you mean -37.9 °C?

Yes. We observed that there is a difference in ice onset *T* between ascending and descending *T*-ramps, and possible reasons for this are discussed in paragraph.

- 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.

We believe the reviewer refers to Fig. 2. It is not possible to include the activated fraction in Fig. 2b in a meaningful manner. In the figure, we show particle counts in bins without any ice crystals, as well as size bins dominated by ice crystals. Normalisation to the total particle concentration would neither provide an ice-activated fraction – nor more information to the figure.

We agree that Fig 2a alone does not add anything to the main findings of the study, but it shows very clearly why the upward temperature ramps provide significantly better control of the lamina conditions. That is of importance to present, since we are not familiar with similar operation of CFDCs with increasing T-ramps from previous studies.

It would indeed be possible to present fractions of Fig. 2b as a function of average lamina temperature instead of time – but then it would not be possible to include the background checks in a meaningful way, as the lamina temperature is close to constant during those time windows.

Since we operated our CFDC in an untraditional way, we do find it highly relevant to openly show this in Fig. 2, and we find that the relevant information is available from the figure in the current version.

- L251: Replace “upward” by ascending for consistency

Done.

- L252: Change to: “Sect. 3.4”

Done.

- 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 RHw of SPIN and then scanned different T, right? This can be referred to as “T-scan”. This must become much clearer for the reader.

Yes, that was the experiment procedure. References to T-scan added to the paragraph, for instance the ramp description has been edited from:

“All experiments were carried out on similar automated ramps containing the following steps: First, the internal background was checked via sampling filtered dry air for 5 minutes before the lamina T-ramp began.”

to:

“All experiments were carried out on similar automated *T*-scan ramps containing the following steps: First, the internal background was checked via sampling filtered dry air for at least 5 minutes before the *T*-scan began.”

- 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.

Please see our response to comment for L244.

- 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?

We assume that the reviewer initially refers to the statement in L255 (and not L245). A subset of the SPIN experiments were intentionally carried out with water droplet survival as indicated e.g. in Fig. 2. With that operation mode, the mode of droplets surviving the evaporation section never grew larger than ~4.5 micrometers, which allowed us to consider any optically larger particles as ice crystals. The motivation for this mode of operation was to ensure that the vast majority of size selected soot particles in the lamina were exposed to a supersaturation with respect to water sufficient for cloud droplet activation, which allowed for investigation of condensation/immersion mode freezing. As shown by Garimella et al.(2016), the evaporation section of SPIN does not allow for operation at high supersaturations with respect to water before droplet breakthrough is observed. This ‘new’ operation approach (allowing droplet breakthrough) leads to the ice-activated fraction being biased low to a degree, which can be estimated. This be discussed in more detail below. The alternative of operating at supersaturations low enough to ensure no droplet breakthrough would most likely also bias the immersion freezing ice-activated fraction low. This is because an unknown fraction of the sample aerosol potentially would not be exposed to supersaturations high enough to ensure activation into cloud droplets inside SPIN.

It is not straightforward to apply condensational growth calculations to an aerosol population inside a CFDC. Garimella et al. (2017) showed significant discrepancies between modeled and measured size distributions, which they ascribed to only a fraction of the sample being focused in the lamina.

We do not consider it necessary to carry out theoretical condensational growth calculations in this context, when the experimental reproducibility of single experiments is high. However, we consider it most likely, that even the largest surviving droplets to some extent shrunk in the evaporation section before detection.

We have not applied any changes to the manuscript in this context, since we find that the presence of the droplet mode already is significantly described e.g. in Fig. 2.

- 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.

A small tail of the droplet size distribution detected with the OPC would typically approach 6 microns in size at a sample temperature near -32°C with our operation conditions. Therefore, an ice threshold of 6 microns was applied throughout this study.

The text (L255-257) was changed from:

“Based on the homogeneous freezing experiments and all the other experiments we carried out under these conditions, the liquid droplets do not grow larger than approximately 4.5 µm in the SPIN when the sheath and sample flows of 9 LPM and 1 LPM, respectively, are used and the particle residence time in the IN chamber is 10 seconds.”

To:

“Based on the homogeneous freezing experiments and all the other experiments we carried out with the same operation conditions, the very largest droplets detected with the OPC approached a diameter of 6 µm.”

- L265: “since the ice crystal mode…” You might want to refer back to your Fig. 1 here.

Reference to figure added.

- L269: Please specify “in a later section”

The text has been changed from:

“The potential biases in the ice-active fraction associated with this approach are discussed in more detail in a later section.”

to:

“The potential biases in the ice-active fraction associated with this approach are discussed in more detail in Sect. 3.4.”

- L275: Delete “in real life”

Done.

- L276: Please clarify whether your ice counts are corrected by this factor. Which?

The presented ice-activated fractions are NOT exposed to any corrections. Since the correction factor is uncertain, which is inherent for CFDC experiments in general (Garimella et al., 2017).

- 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.

The statement is moved to Methodology section and mentioned after the definition of the activated fraction.

The latter statement has been changed from:

“A result is considered positive if the ice onset has taken place under conditions when the sample temperature or *RHi* have been significantly higher or lower, respectively, than conditions required for homogeneous freezing. The result is considered negative if ice crystals have not been observed or if there is no distinguishable difference to homogeneous freezing conditions.”
to:

“A result is considered positive if heterogeneous ice nucleation is observed under freezing conditions that are distinguishable from homogeneous freezing, and negative if ice crystals have not been observed or if there is no distinguishable difference to homogeneous freezing conditions.”

- L286: Please define “ultrafine”, also for usage further down, e.g. L329.

The text has been changed from:

“All samples were dominated by ultrafine particles by number,”

to:

“All samples were dominated by ultrafine (< 100 nm) particles by number,”

- L289-L291: You need to elaborate on various statements made here:

o 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?

Throughout the campaign we observed that all cookstove emissions were heavily dominated by ultrafine (< 100 nm) particles which go through the OPC undetected when operating SPIN without droplet breakthrough. Even with droplet breakthrough, only a fraction of the sample particles will be detected by the OPC – the ones focused in/near the lamina while also ‘surviving’ the evaporation section. At these particle concentrations, previous OPC characterisation showed no risk of co-incidence errors. Please see our response to also the next comment.

o 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.

The detection limit is basically defined by the signal to noise ratio, in this context, the ice crystal counts relative to the background noise. Increasing the sample concentration thus may increase the signal and thus the sensitivity. Our operation with sample number concentrations at the order of 10^3 cm^{-3} should ensure negligible vapour depletion inside the CFDC (Levin et al., 2016)

o Your detection limit of the order α = 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 α = 10-6 is your true detection limit, I suggest to more clearly write this in your statement on L211.

We have added a more detailed description of the detection limit to the original statement on L211, which is presented below:

"Typically, the background signal was below 1 particle per liter in the beginning of the experiment and the IN chamber was re-iced when it exceeded 10-15 particles per liter. This protocol, together with averaging the instrument data over 10-second periods as was done in data analysis, allowed detection of the ice-activated fraction from the order of 10^{-6} upwards in experiments where the sample concentration exceeded 2000 cc^{-1}. Data averaging enabled investigation of large droplet/ice crystal populations against their corresponding background signals, thus decreasing the lowest detection limit."

- L291: "Ice crystals…". This statement is not true looking at your "FD gasifier SW" experiment, if your threshold is α = 10-6.

Table 1 states that the detection limit in "FD gasifier SW" transient experiment was higher than 10^{-6} due to lower sample concentration.

- L291-294: Improve statement by saying: ""… four stoves do not nucleate ice in neither deposition nor immersion/condensation mode…"

Done.

- 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.

Description moved to Sect. 2.3.

- 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.

1. Size range and concentrations corrected to be consistent with L129.

2. The difference in number concentrations is due to size selection that was applied in chamber experiments, when only a fraction of the size selected particles makes it through the DMA. On the contrary, the transient experiments were done on polydisperse aerosol that was heavily dominated by ultrafine particles.

- 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.

We have added the theoretical maximum estimate for doubly charged particle fraction to Table 2, following the calculations by Wiedensohler (1988). From there it can be clearly seen that the potential contribution of doubly charged particles account number concentrations that are much below the minimum reasonable sample concentration to the SPIN.

- 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.

The text has been changed from:

“which indicates that soot emissions were present in all studied cases.”

to:

“which indicates that large soot particles were present in all studied cases.”

- L329: Replace “when” by “after”

Done.

- 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.

Done.

- 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.

Yes, Table 3 supports the discussion. References added.

- 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.

The number has been corrected to correspond to the ones in Table 3.

- 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?

In order to model the theoretical freezing temperature based on classical nucleation theory, we would need to accurately know not only the time development of the droplet size distribution, respective temperatures and residence times for potentially different RH and temperature conditions. That is unknown for this and other CFDCs. And yes, that is why we determine the homogeneous freezing experimentally for the given operation conditions. The motivation for the comment, is that you can find examples in the literature of slightly higher homogeneous freezing temperatures closer to -38°C (e.g. Ignatius et al., 2016) potentially related to larger droplets.

The text has been changed from:

“The observed homogeneous freezing with 10^{-3} ice activation at an average lamina temperature of -38.9 °C may occur at slightly lower temperature than expected. It is not clear at which temperature homogeneous freezing in SPIN can be expected due to the sample aerosol being exposed to a span in temperature and RH, and uncertainties related to droplet sizes and residence times above the evaporation section as discussed by Ignatius et al. (2016).”

to:

“The observed homogeneous freezing with 10^{-3} ice activation at an average lamina temperature of -38.9 °C may occur at slightly lower temperature than expected from previous results reported in the literature. It is not clear at which temperature homogeneous freezing in SPIN can be expected from based alone on theoretical considerations due to the sample aerosol being exposed to a span in temperature and RH, and uncertainties related to droplet sizes and residence times above the evaporation section as discussed by Ignatius et al. (2016)."

- 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.

Discussion on potential biases moved entirely to Sect 3.4.

- L342: Please elaborate how you estimate this correction activated fraction of 10-3 for the temperature interval indicated.

Application of a correction factor is mainly due to only a fraction of the sample particles being focused in the lamina and a potential presence of ice crystals below the threshold of 6 µm. Garimella et al. (2017) investigated the fraction of sample particles being focused in the lamina for SPIN. They reported 10% to 30% of the sample particles being focused in the lamina in most cases. That study indicates potential correction factors in the range from ~3 to ~10 to the ice-activated fraction. In our experiment, we estimate an upper correction factor to be ~8, which is based on the ice-activated fraction (for D>6 µm) reaching ~0.12 with the average lamina temperature being -41°C for the homogeneous freezing experiment. At that average lamina temperature, we would (in the ideal experimental setup) expect an ice-activated fraction approaching 1.

Application of correction factors in the range from 3 to 8 to the homogeneous freezing ice-activated fraction e.g. presented in Fig. 4 leads to a corrected ice-activated fraction of 10^{-3} in the temperature range from about -38.5°C to -38.0°C. Those homogeneous freezing temperatures are close to what we would expect from theoretical estimations as provided by Ignatius et al. (2016).

L342-343 have been changed from:

"We estimate that correction for those biases in the homogeneous freezing experiments will result in 10^{-3} ice activation between -38.5°C and -38.0°C in average lamina temperature."

to:

"Garimella et al. (2017) reported that the fraction of sample particles being focused in the lamina for comparable operation conditions of SPIN to be from 10% to 30% in most cases. In our homogeneous freezing experiment, the 'raw' ice-activated fraction as presented e.g. in Fig. 4 is about 0.12 for an average lamina temperature of -41°C. At that temperature, we would ideally expect an ice-activated fraction close to 1, which would be reached applying a correction factor of 8. For slightly higher lamina temperatures, the correction factor could potentially be lower, so we consider a correction factor in the range from 3 to 8 to be possible for this experiment for the lamina temperature in the range from -41 to -38.0°C. Application of such correction factors for the homogeneous freezing experiment will result in 10-3 ice activation between -38.5°C and -38.0°C in average lamina temperature."

- L349: Replace "as was done by" by "following"

Done.

- L349: "The uncertainty…" This sentence is a direct repetition of the previous statement, please delete.

Done.

- L353: Change “experiment emissions” to “experiments” and “high experiments” by “Good”

Done.

- L354: Change to “Sect. 3.4”

Done.

- L355: This statement needs to be clarified:

o 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.

The α-uncertainty bars that present the combined relative standard deviation (RSD) for the CPC and OPC will be added to the revised manuscript.

o 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?

The AMS analysis shows that the only difference in chemical compositions between the two mentioned experiments is higher BC to OA ratio in the 3S_CAS_300nm experiment, but we consider that it cannot explain the missing size dependency in IN activity. The differences of other studied physico-chemical properties were negligible in these two experiments. We would not expect to be able to identify physico-chemical differences between those experiments explaining the minor differences in ice-active observations – since our detailed analysis linking the results of more ice active samples to their properties presented in Figures 6 and 7, respectively, showed no clear pattern.

- L358: Add “than the ones…”

Done.

- L359: I cannot find the number “-37.7°C” in the RS experiments in your Tab. 3, please check.

Number corrected to -37.9°C.

- 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.

Corrected.

- 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.”

Statement tuned down as suggested.

- 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:

o 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?

Unfortunately, there is not directly comparable SP-AMS data from the FD gasifier and SW fuel on normal operating conditions.

o 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:

The description of the modified experiments has been moved to Sect. 2.4, following guidelines given in general comments. The description of the "*pot height*" type experiments is as follows:

"The "*pot height*" experiments were done using different, less favorable heights of the cooking pot. The three such experiments were done on designated operation conditions of the FDGS and SW pellets, but the cooking pot was intentionally lifted to achieve production of larger particles: the real-time size distribution was monitored on the FPA throughout the experiment and the pot height was adjusted in a way that an increased production of large soot particles was observed. In typical cases, an offset of 8-10 cm above designated pot height affected production of large soot particles significantly."

o 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?

This could possibly be because AMS measures the (ensemble) average composition, while the IN constitute a very small subpopulation of the particles. It is not necessary that the size of that subpopulation co-varies with the average composition.

o 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?

Unfortunately, the supportive data does not show which particle properties, or their combinations, could have caused this.

o 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.

Here we do not agree, the B450 and B500 experiments both have high A370/A880 and high Cmid/Call. The difference in OA to BC is rather small (0.18 and 0.23).

- 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.

Yes, the effective density is indeed a function of particle size, so we do not expect the effective densities for 350 nm mobility diameter particles to be identical to those for 400 or 500 nm particles. The effective density was measured for 350 nm particles in all the experiments relevant for the results presented in Fig. 7, but unfortunately, we did not measure the effective density systematically for larger particle sizes.

Rissler et al. (2013) successfully applied 'simple' model fits to the effective density versus particle mobility diameter for different types of soot particles. If we assume a somewhat similar model of effective density vs mobility diameter to be applicable for our studied cases, then qualitative differences in the effective density for 350 nm particles should remain also for larger particle sizes. In other words, we do consider the differences in effective density for 350 nm particles to reflect differences in the effective density for the poly-disperse soot population to some extent.

This hypothesis is supported by a strong correlation (N=5, r=0.98, p=0.004) between the effective density for the 200 and the 350 nm particles within the soot mode for those 5 samples.

- L383: There is no Fig. 8.

Typing error corrected.

- 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.

Information that describes the method moved to methodology section.

- L391: "The values of…" Is this true for any soot aggregate size? Please specify.

No, this is not true for all soot aggregate sizes according to Rissler et al. (2013), to which we refer in that statement. They found that the values we mentioned were typical to soot aggregates larger than approximately 250 nm, i.e. the size range we sampled in our chamber experiments.

- L396: "OA" is not defined. You might also want to use "OA" instead of "Org" in your Fig. 7.

Yes, 'OA' is now defined when first introduced, and applied in figure label.

- L396: Should not K400#1 have a higher UV absorption, given the high Org carbon fraction?

At these low OA/BC ratios the AAE seems to be mainly driven by the components responsible for the refractory midcarbon signals. These have previously been associated with elevated AAE by Malmborg et al. (2019) and Török et al. (2018), which was stated later on L398-399.

- L412: Can you quantify this correlation?

The correlation was quantified on L404, stating that the $C_3O_2^+/C_3^+$ varied slightly between 0.005-0.007 from the least ice-active case to the most active one.

- L416: "Further studies…" I suggest to move this statement to your Sect. 4.

Moved to Sect. 4.

- L418: "The typical…" This is a repetition from L250 and can be deleted.

Repetition deleted.

- L425: Change "Chapter" to "Sect."

Done.

- L426: Replace "to" by "the"

Done.

- L428: This is a direct repetition of L346 and can be deleted.

Repetition 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.

The text has been changed from:

"somewhat similar operation conditions to what we apply in our study"

to:

"approximately similar operation conditions (T ≈ -33 °C at the lamina) to what we apply in our study"

- L445-461: This paragraph need to be significantly elaborated and improved in order to justify a proper discussion of biases in ice crystal detection.

o 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.

We have included α-uncertainty to activated fractions, so the reader can judge uncertainties in both lamina *T* and the particle detection.

o It remains unclear why the "underestimation depends on lamina temperature".

Yes, we agree that this statement may appear unclear. This underestimation depends on whether freezing has occurred in a size range significantly below our ice threshold as we observe strong indications of for an average lamina temperature near or below -40°C as explained in the following statement (L447-449). At such low sample temperatures, we observed more than one size mode to be dominated by ice, so we have also modified the formulation in the previous line (L446) slightly.

For the heterogeneous freezing results obtained well above the homogeneous freezing temperature, the vast majority of the ice crystal size mode appears above our size threshold of 6 microns, so this bias is much more pronounced for the lowest sample temperatures (near -40°C)

The text (L446-447) was changed from:

"only a fraction of the ice crystal mode is included in our reported ice active fractions. This underestimation depends on the lamina temperature."

to:

"only a (varying) fraction of the ice crystals were included in our reported ice active fractions. This underestimation turned significant when the sample temperature was low enough to induce freezing of 'small' droplets and/or possibly hydrated particles appearing at sizes well below the ice crystal threshold."

o How do you arrive at the upscaling factors of 3-5 and 5-10? Please add this to the text.

The lower correction factor estimate of 3-5 for T_{lamina} ≈-33C is estimated by assuming at least a correction factor of 3 due to about one third of the sample aerosol being focused in the lamina (DeMott et al., 2015; Garimella et al., 2017). It may potentially be higher than 3, due to the potential presence of ice crystals with optical diameters <6 µm. In addition, potential losses of ice crystals between the chamber and the OPC have not been quantified. The higher estimated correction factors of 5-10 for T_{lamina} ≈-41C are inferred in another manner. At this sample temperature, particles detected with the OPC in all size ranges (D>~0.7 µm) may be ice crystals, since we observe a clear transition in the average depolarization ratio in all size bins for T_{lamina} going from -41 °C to -38 °C, which is indicative of a phase transition from ice crystals to liquid droplets/hydrated particles. We would also expect cloud droplets to freeze homogeneously at temperature close to -41 °C. However, the inferred ice-activated fractions as shown e.g. in Fig. 6, reach levels from about 0.08 to about 0.2, where we ideally would expect values close to 1. So we estimate the correction factors for T_{lamina} ≈-41C to be at the order of 5-10, depending on the experiment. The statement has been improved from:

“DeMott et al. (2015) found that the ice active fraction should be upscaled by a correction factor of 3 for a CFDC and somewhat similar operation conditions to what we apply in our study. Garimella et al. (2017) reported upscaling factors for SPIN in the range from 1.5 to 10 depending on the operation conditions due to only a fraction of the sample aerosol being focused in the lamina.”

to:

“DeMott et al. (2015) found that the ice active fraction for mineral dust particles should be upscaled on average by a correction factor of about 3 for a CFDC over a range of operation conditions including similar operation conditions (e.g. T ≈ -35 °C and RH_w about 106-108% at the lamina) to what we apply in our study. However, for those specific operation conditions (lamina T=-35°C and just before droplet break through with an expected RH_w about 106-108%) there is quite some scatter in the appropriate correction factor and a larger correction factor is often observed (DeMott et al., 2015). The offset can be ascribed to only a fraction of the sample particles being focused on the laminar flow. Garimella et al. (2017) reported upscaling factors for SPIN in the range from 1.5 to 10 depending on the operation conditions due to only a fraction of the sample aerosol being focused in the lamina. This is also evident in this study at laminar temperatures below -40 °C when all droplets are expected to freeze homogeneously: the inferred activated fractions (see Figs. 4-6) are between approximately 0.07 and 0.2, which suggests that a correction factor range suggested by Garimella et al. (2017) is required.”

o Finally, please be more quantitative when saying “in a neighborhood of -40 °C”

The text has been changed from:

“The considerations discussed above lead us to estimate a likely upscaling factor at the order of ~3-5 of the ice active fraction for temperatures around -33 °C, while the upscaling factor is likely to approach 5-10 for temperatures in a neighborhood of -40 °C.”

to:

“The considerations discussed above lead us to estimate a likely upscaling factor at the order of ~3-5 of the ice active fraction for lamina temperatures around -33 °C, while the upscaling factor is likely to approach 5-10 for temperatures significantly below homogenous freezing, around -40 °C.”

- L468: “This study shows…” It remains unclear whether this statement refer to all different cook stoves tested or not.

The text has been changed from:

“This study shows that even small changes in combustion conditions can significantly affect the IN abilities of emission particles.”

to:

“Based on observations from five special experiments on FDGS, this study shows that even small changes in combustion conditions can significantly affect the IN abilities of emission particles.”

- 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.

The text has been changed from:

“The regional daily usage by at least 500 million people suggests that the solid biomass burning”

to:

“Extensive regional usage of cookstoves suggests that the solid biomass burning”

- 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.

The text has been changed from:

“The CFDC instrument in this study, the SPIN, showed relatively good performance and high reproducibility of experiments.”

to:

“The CFDC instrument in this study, the SPIN, showed relatively good performance in temperature control, which enabled high reproducibility of experiments.”

- L475: It would be good to quantify “poor IN activity” (not INP activity!) by giving a range of observed activated fractions.

The text has been changed from:

“We conclude on the experiments that the fresh, polydisperse emissions from cookstoves have a low INP potential at experimented temperatures (-28 ^{o}C, -32 ^{o}C). All emissions were heavily dominated by ultrafine particles that clearly showed poor INP activity. Moreover, their residence time in the atmosphere is relatively short due to deposition, coagulation and external mixing with other atmospheric particle species. Accumulation mode particles that were present in the transient experiments were not observed to activate heterogeneous ice nucleation at -32 ^{o}C in neither freezing mode, from which it can be concluded that the studied cookstove emissions have low IN activity at that temperature also in the immersion mode freezing.”

to:

“We conclude that the studied polydisperse aerosol emissions from transient experiments with different cookstoves showed no indication of heterogeneous freezing above the detection limit (ice-activated fraction of about 10^{-5}) for the investigated conditions (T=-28 °C and -32 °C and ~75-105% RHw). However, it should be kept in mind that those aerosol emissions typically were heavily dominated by ultrafine particles by number. Hence, the measurement sensitivity with respect to the accumulation mode particles alone was relatively low in those experiments. Therefore, we also carried out a number of immersion freezing experiments focused on size selected accumulation mode soot particles sampled from an aerosol storage chamber."

- 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?

We do not mean that all cookstove emissions end up in the upper atmosphere. However, a notable fraction of these PM emissions can be lifted to several kilometres in altitude in regions where convective draft is strong during daytime (viz. tropical and sub-tropical regions near the equator). The text has been changed from:

"Therefore, we conclude that usage of the cookstoves can emit potential INPs in the atmosphere, and thus affect cloud properties."

to:

"Therefore, we conclude that usage of the cookstoves can emit potential INPs in the atmosphere, and thus affect cloud properties such as the frozen fraction in MPCs and formation of cirrus clouds."

- 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

We have removed the comparison from the text.

- L490: Delete "of"

Done.

- 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.

The text has been changed from:

"Our analysis on physico-chemical properties of the emissions revealed slight differences in studied properties of soot particles that were present in the most ice-active results, yet these properties cannot define the IN efficiency alone."

to:

"Our supportive analysis on physico-chemical properties of the emissions revealed that hygroscopicity, OA-BC ratio, fraction of mid-carbon range fragments and fraction of refractory oxygen species cannot define the IN activity alone; the observed differences of these properties showed no clear trend towards increasing IN activity (see Fig. 7)."

- L761: Change “,” to “.” In front of “The two…”

Done.

- 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…”

Done.

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