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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Ice nucleation abilities of soot particles determined with the Horizontal Ice Nucleation Chamber" *by* Fabian Mahrt et al.

Anonymous Referee #1

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Review for "Ice nucleation abilities of soot particles determined with the Horizontal Ice Nucleation Chamber" by F. Mahrt et al., submitted to ACPD

In the manuscript, the ice nucleation activity of 6 different soot samples is examined with the CFDC-type ice nucleation chamber HINC at the ETH (Zurich). Additionally, a thorough analysis of physical properties of the soot particles is presented. All data are jointly discussed. It is concluded that the examined soot particles do not act as ice nucleating particles in the mixed cloud regime. In the cirrus cloud regime, some soot types were more ice active than others, which was traced back to pore condensation and freezing.

The manuscript is well written. Data is nicely presented. The overall message is clear. Sometimes I felt that the information was a bit too much, but then it is all interesting and

well researched, so I will not suggest so shorten anything. Below, I give a number of (mostly small) remarks / suggestions for improvements. The number of remarks makes me choose "major revisions", but it should all be easily done. Overall I recommend this manuscript for publication in ACP once these suggestions will have been addressed.

Specific comments:

page 2, line 8: Vali et al. (2015) do not introduce any research or their own, but rather summarize knowledge, thus this citation here is a bit misleading. They don't show that soot particles can act as INPs. I suggest removing this citation here.

page 4, line 25: You claim that your sample "LB_RC" is "directly comparable to the lamp black soot purchased from OEC, due to their very similar physical properties" – however, the BET surface areas of these two soot samples is an order of magnitude apart, so I suggest to delete this statement about their comparability, or adjust it accordingly.

page 5, Table 1: It would be interesting to know how many particles were counted in the TEM analysis. This could be given in an additional line.

page 6, line 8: What were these filters used for? It would be nice to get this information here in the text.

page 6, line 10: Does this mean that the mini-CAST samples on the filters were sampled BEFORE this additional coagulation took place? Please give the answer to this question in the text.

page 6, line 18 and 19: You use HINCNE and HINCNE (i.e., once is in italic, the other isn't). Be consistent throughout the text. My personal preference is the one with italic letters.

page 7, line 7-9: It is not so clear to me if you really can assure that the aerosol flow through the DMA is stable – did you check the exhaust air from the mixing chamber regularly? Was it constant? And were the flows in the DMA stable?

page 7, line 22: The phrase “Cloud particles can be formed” is not correct. Ice may or may not form, as you say, and water droplets will only form at RH_w > 100% and therewith at much higher humidities. Therefore, it would be better to say "Ice crystals may be formed".

page 8, line 1-2: “The residence time is the sum of a nucleation and water droplet or ice crystal growth time in the chamber.” – This is strangely formulated. Please reformulate. Maybe “The residence time is the sum of the times . . . “

page 8, line 14-15: How long does it take, until the particles reach the destined temperature after entering HINC? If both sheath and aerosol flow are at laboratory temperature upon entering HINC, there will certainly be a non-negligible delay. This needs to be mentioned and shortly discussed. Also, on the opposite side, could settling of the particles to the lower plate be an issue?

page 8, line 31: Certainly you mean for sizes from 1 ïA■m onwards, and not only in the 1 ïA■m channel? Correct this.

Chapter 2.4: This chapter is an interesting and impressive addition to the ice nucleation measurements - but it would be good to know which of these measurements were performed on the bulk samples and which on the size segregated particles, and for the latter, if then the same set-up described above was used. Please add information on this to the text.

page 9, line 13: Better replace “aid” by “contributed to”.

Figure 2: The “blue dashed line” – is that the one that is almost at the RH_w = 1 - line? This is almost impossible to see - use a different color, e.g., red, or small circles or something else.

page 11, line 11-13: Would this then not mean that every possible ice active material would be outcompeted by homogeneous freezing? Still, heterogeneous freezing is considered to play a role even for the cirrus cloud regime. And if this (second) part of

the sentence can be debated, I wonder if the first part (about 100 nm particles being not relevant) is not a trade of between the HINC detection limit and the abundance of these particles in the atmosphere?

page 11, line 25: Missing space between y and 2.

page 11, line 32-33: Another reason could be that it takes up water better – could be worthwhile mentioning.

page 12, line 4: A new sentence should start between “particles” and “they”.

page 14, line 7: Add a “,” between “increases” and “the” (it took me a while until I got the sentence).

page 14, line 10: Tiny remark, but why “in prep.b” first (before “a”)?

page 15, line 7: There is a space too much before the first letter.

Figure 6: One of the screens I used while working at this review did not show this graph well. The "yellow" color shading looked rather pinkish, and the shading of the two other panels was almost the same - maybe there is a better (or additional) way to refer to the panels? (Using a), b) and c) for example?)

page 17, line 19-20: Are you really referring to S16 in the SI, here, so should this rather be S10? But in any case, I have difficulties following your argument – why does the lack of a change in the slope render this interpretation unlikely?

page 17, line 33: Change “of” to “the”

Figure 7: If you used a log-scale for one of the two panels, it would be easier to see the hysteresis and the absolute values for the curves at the lower end. The way it is now, both panels look quite similar, anyway.

page 19, line 21: Are citric acid and glutaric acid organics typical for the atmosphere? Any citation on that? Others might play a more important role (oxalic acid, formic acid,

acetic acid, succinic acid, ...). But there must be a reason why you choose those. Explain that in a few words.

page 20, line 18-21: The last few words “rather than the bulk aggregate size” puzzled me a bit. There is a difference in the freezing curves for FW200 particles of different sizes, and you say, quite correctly, that you observe 1% of particles ice active at similar RHs for a given particle size (BTW: exchange “aerosol size” with “particle size” here). But then why would you refer to “bulk aggregate size” in the end. Would anyone really assume that this plays role? I suggest to delete these few last words unless you feel they are needed.

page 21, line 7: I suggest to tune down “confidently identify” a little.

page 22, line 22: Concerning the use of the word “contact angle”, I realize that you talk about the interaction between the soot surface and water vapor. But on the other hand, it has been shown for other materials that it never only is one contact angle describing the ice nucleation behavior of one substance, but a contact angle distribution instead. This might need to be mentioned.

page 23, line 5: It should not be forgotten that solid fuels (from either biomass burning or wildfires) also produce ashes, so for observed atmospheric ice activities of respective particles, also these ashes could be responsible (Umo et al., 2015, Grawe et al., 2016).

page 23, line 15: In your argument about the pore ice, it was not clear to me right away how it should have gotten there in the first place – after rereading the sentence I now assume that likely you imagine that ice nucleated in a previous cloud cycle, and in the following one, ice crystals can form now more readily? Maybe describe this more explicitly?

page 24, line 2-4: In the framework of “pore condensation and freezing”, indeed liquid water is part of the concept. However, the fact that ice nucleation was observed below

RH required for homogeneous freezing does not necessarily suggest that liquid water is required: Deposition ice nucleation could also happen! I agree with you that maybe pore freezing is the process rather at work here, but still, the reasoning between these two sentences is somewhat flawed. The observation described in the first sentence does not result in the conclusion you draw in the second sentence. Reformulate.

page 25: Why does this start with Appendix B? What happened to A? (Probably an issue for typesetting, anyway.)

Table B1: What do #1 and #2 stand for? And why does "C (#1)" occur twice?

Figure E1: z-values do not fit to what is shown on y-axis - is this only a fraction of the profile that you show here? Please clarify.

Chapter B2: The here presented analysis, and others, were done for bulk samples, while ice nucleation was measured for size segregated particles - possible implications for this should be discussed somewhere (not here, but up in the main text, where you draw conclusions about the connection between soot characteristics and their ice activity.

Table B4: Is there a reason why you give the factor C, but not (or not also) the effective density? The latter could be of interest for the readers.

My only remark for the supplement: Fig. S18: Please mention in the caption, that the length of the bar in the picture is 800nm for all pictures in the left column and 200nm for all others - otherwise this needs blowing up quite a bit before these numbers can be seen, which is a bit annoying.

Literature: Grawe, S., S. Augustin-Bauditz, S. Hartmann, L. Hellner, J. B. C. Pettersson, A. Prager, F. Stratmann, and H. Wex (2016), The immersion freezing behavior of ash particles from wood and brown coal burning, Atmos. Chem. Phys., 16, 13911–13928, doi:10.5194/acp-16-13911-2016.

Umo, N. S., B. J. Murray, T. M. Baeza-Romero, J. M. Jones, A. R. Lea-Langton, T. L.

Malkin, D. O'Sullivan, L. Neve, J. M. C. Plane, and A. Williams (2015), Ice nucleation by combustion ash particles at conditions relevant to mixed-phase clouds, Atmos. Chem. Phys., 15, 5195–5210, doi:10.5194/acp-15-5195-2015.

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