Response to acp-2021-883 reviews for RC1

We thank Reviewer 1 for their positive and constructive comments on our manuscript ACP-2021-883. In response to the comments, please find our answers and corrections listed below. **Reviewer 1 comments are extracted in bold from original review supplement** and our responses are given directly below in normal font. *The original text in previous manuscript is repeated in red italic* and *corrected text in revised manuscript is typed in blue italic.*

- **General:**

1 The manuscript presents a comprehensive laboratory investigation of IN properties of soot under cirrus conditions simulated in a continuous-flow diffusion chamber. Two types of commercial soot were analysed fresh and after mechanical agitation, which resulted in compaction of soot particles as revealed by electron microscopy. The resulting four kinds of soot material were further separated into four size classes each and investigated by N_2 and Ar sorption measurements regarding differences in their pore size distribution. Soot water interaction was precisely characterised by dynamic vapour sorption measurements. Differences in morphology, in particular mesopore abundance as a factor of compaction and particle size, and the resulting propensity for pore condensation freezing, convincingly explain observed differences in IN activity between the four kinds of soot material and their size classes.

The clear structure of the manuscript makes it easy to follow. There is little I can suggest in terms of further improvements, except perhaps a broader outlook on the implications following from these investigations. For example, what might be the effect of future changes in jet fuel composition, such as additions of biofuel or synthetic fuel, to the IN activity of the generated soot?

R: Thank you for your positive comments and constructive suggestions.

Regarding adding a broader outlook, we agree with this suggestion and now add some new sentences at the end of Sect. 4 as below (L807-816 in revised manuscript):

'*Finally, synthetic aviation fuels, which contain a negligible amount of aromatics and are gaining an increasing share in aviation fuel usage, can lead to a reduction in aero-engine particulate matter mass and number emissions (Duong et al., 2018; Xue et al., 2019) and significant decrease in the GMD (geometric number mean diameter, short as GMD in L86 in revised manuscript) of aero-engine emitted particle population by up to 40 % (Corporan et al., 2007; Corporan et al., 2011). Considering the IN dependence on soot particle size, it implies that the soot emission from aviation activities fueled by synthetic fuels should have a decreasing impact on cirrus formation. Bräuer et al. (2021) reported that low-aromatic biofuel blends considerably reduce aircraft emitted ice by 40 %. However, Kärcher et al. (2021) noted that a small number of active soot particles in aviation contrails may still modulate cirrus cloud microphysics, by decreasing the ice crystal number concentration but increasing the ice crystal mean size. Hence, further constraining the understanding of the IN activity of small size soot particles is necessary to mitigate aviation soot impacts on the climate.*'

- **Technical issues:**

1 L22: consider replacing 'agitation degree' by 'degree of compaction'

R: Agreed (now L22 in revised manuscript):

'*... the IN activity of soot particles with the same degree of compaction, ...*'

2 Figure 1: What does 'xx #/cc' stand for?

R: The ‘xx #/cc’ stands for the particle number concentration measured the condensation particle counter (CPC) in Figure 1. Now, these ‘xx #/cc’ labels are removed from Figure 1, to avoid misunderstanding.

3 L277: replace 'an' with 'a'.

R: Yes, it was replaced as below (now L283 in revised manuscript):

‘*... in addition to a release of N_2 or Ar from pores ...*’

4 Figure 5: Please say in the legend what the red circles in panels a and b are indicating.

R: Thank you for this suggestion. Instead of using a legend to indicate the red circle, we now add a statement in the caption because Panel (a) and (c) in Fig. 5, where the red circles are marked, are busy. Now the caption for Fig. 5 is as below (now L352-353 in revised manuscript):

‘*TEM images for 400 nm fresh and compacted FW200 and PR90 soot aggregates at a magnification value 75k. The red circle is used to indicate intra-aggregate voids with size of tens of nano-meters.*’

5 Line 358: This sentence needs rephrasing. Perhaps in this way: 'However, the rest of the AF curves for small size (60 and 100 nm) are not significantly different for compacted and fresh FW200 or PR90 soot.'

R: Thank you for this comment. We agree that this sentence needs to be rephrased but we think the following correction might be better (now L365-366 in revised manuscript):

‘*However, the rest of the AF curves for small size (60 and 100 nm) particles do not show significant differences between compacted and fresh FW200 or PR90 soot.*’

6 Line 411: replace ‘shown that’ with ‘shown by the fact that’

R: Yes, it was replaced (now L419 in revised manuscript):

‘*...PR90 soot particles are less active INPs compared to FW200 as shown by the fact that the same size fresh or compacted PR90 soot particles...*’

7 Line 459: do you mean ‘still more competitive’?

R: Within context of L459 (in original manuscript) in Sect. 3.2.1, we mean soot-aggregate compaction still can play a role in promoting the ice nucleation ability of 200 nm FW200comp particles at 233 K close to the homogeneous freezing temperature. To clarify this, the statement was changed as below (now L466-468 in revised manuscript):

‘*Particularly, there is also a reduction in onset S_i value at T = 233 K for 200 nm FW200comp soot particles showing a smaller S_i value compared to 200 nm fresh FW200 soot particles, which means that soot-aggregate compaction can even promote, albeit by a small S_i reduction, 200 nm FW200comp to nucleate ice.*’

The original statement is as following:

‘*Particularly, there is also a reduction in onset S_i value at T = 233 K for 200 nm FW200comp soot particles showing a smaller value just out of the error bar compared to 200 nm fresh FW200 soot particles, which means the compaction induced IN enhancement still plays a role, albeit minor at T = 233 K.*’

8 Line 529: ‘the’ instead of ‘THE’?

R: Yes, it was corrected as below (now L538 in revised manuscript):

'… *between adsorptive gas and the solid,* …'

9 Line 632: 'too large' instead of 'too larger'?

R: Yes, it was corrected as below (now L642 in revised manuscript):

'*These pore structures are too large to trigger inverse Kelvin effects …*'

10 Line 773: 'preliminary' or 'primary?

R: Here, we mean mesopore structures relevant to the pore condensation and freezing (PCF) process is the prerequisite property for a soot particle to nucleate ice via PCF and is more important than the particle contact angle (surface wettability). Now, we replace the word 'preliminary' with 'prerequisite', which we think it is better than 'preliminary' and 'primary'. Please see the correction as below (now L782-783 in revised manuscript):

'… *suggesting that the existence of PCF relevant mesopores, as a prerequisite for soot PCF activation, is more important than the contact angle.*'

11 Line 778: replace 'are in favoured for' by 'favour'

R: Yes, it is replaced as below (now L788 in revised manuscript):

'*Our study suggests that compacted soot particles with abundant mesopore structures and a low contact angle favour ice activation via PCF at low T and RH conditions …*'

12 Lines 781-782: I do not understand the sentence starting with 'Some aero-engine soot particles...'

R: Here, we mean aviation soot particles may experience multiple cloud ice activation cycles, i.e. cloud processing (Mahrt et al., 2020b). Now, the sentence is changed as below (now L791-793 in revised manuscript):

'*Some aero-engine soot particles emitted directly in the upper troposphere can act as INPs in a first ice formation cycle and release residuals upon sublimation of this ice. The released residuals can have a compacted aggregate structure (Bhandari et al., 2019; Mahrt et al., 2020b). After such a…*'

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