Dear Reviewers,

Thank you very much for your careful reading and the detailed comments and recommendations on our revised manuscript: *"Enhanced ice nucleation activity of coal fly ash aerosol particles initiated by ice-filled pores"*. Please find our responses to the minor revisions in blue text beneath each comment. We have also marked the corrections with a blue text in our latest submission.

Report 1

Review of revised manuscript by Umo et al., 2019:
General comment:

The manuscript is strongly improved and the readability increased. I recommend publication after the following minor revisions.

Thank you.

Specific comments:

Page 2, line 24: It is questionable whether ice-filled pores should be termed active sites, since active sites are considered to consist of a material different from ice. Also, growth of ice on ice should not be considered a nucleation process. Please revise the formulation.

The sentence has been revised as follows: *"In an ice-supersaturated environment, these ice-filled pores can then initiate the growth of macroscopic ice crystals on the particles."*

Page 3, lines 30 – 32: This sentence needs to be formulated better. It should become clear that the first statement refers to immersion freezing experiments and it should be explained what is meant by "hydratable components" (their origin and composition).

We have reformulated this sentence as: "*Grawe et al. (2018) partly attributed the ice nucleation behaviour of the CFA particles in the immersion freezing mode to the quartz content of the CFA particles. The influence of this quartz content on the particles' immersion freezing ability can be suppressed in a situation where hydratable components form a layer on the particle surface (Grawe et al., 2018). These hydratable components are chemical compounds (e.g. $CaSO_4$) contained in CFA particles that are capable of taking up water at elevated ambient relative humidity. This can lead to the formation of new compounds such as calcite and gypsum."*

Page 4, lines 19 – 20: Can you specify the CaO content of classes C and classes F CFAs?

We have added a sentence to indicate the percentage of CaO contents in classes C and F of CFA particles. *"A typical mass fraction of CaO in Class F CFA particles is ~ 1 – 12 wt.%, whereas Class C has higher CaO contents, sometimes up to 40 wt.%.*"

Page 6, lines 26 – 27: is this a correct definition of ice-activated fraction? Shouldn't the division be through the sum of all particles/droplets/crystals present in the chamber?

Yes, this is true. All ice-activated fractions presented in the manuscript were also computed according to the above definition. The erroneous statement in lines 26/27 has been modified to: *"The fraction of ice frozen (i.e., the ice-activated fraction, f_{ice}) was calculated as the number of ice particles detected divided by the total number of seed aerosol particles present in the chamber (Vali, 1971)."*

Page 6, lines 33 – 37: there are 3 panels in each row. It should therefore read: top panels represent … middle panels show … bottom panels show…

Changed.

Page 6, lines 35 – 36: do you really mean that the freezing experiment was stopped and not just

the expansion?

It was just the expansion that was stopped at that point. We have corrected this sentence to: *"The point where the pressure starts rising indicates when the expansion was stopped."*

Page 7, line 16: "we discuss" instead of "we discussed".

Changed.

Page 7, lines 21 – 22: this sentence reads as if the 0.19 % of the particles all nucleated at 244 K, Do you really mean this or not rather that about 0.19 % of the particles had nucleated ice at 244 K?

We have revised this sentence as: *"However, at T_{start} = 253 K, about 0.19 % of the particles had nucleated ice via the immersion freezing mode in the course of the expansion cooling run until the minimum temperature of 244 K was reached (Fig. 3B)."*

Page 7, line 23: the same issue again: do you mean "had increased…"?

Here, we wanted to compare the detected ice-active fraction to that observed in the preceding experiment started at 253 K. The sentence now reads: *"The ice-active fraction encountered during the expansion cooling run at T_{start} = 245 K was by a factor of 10 higher compared to the run started at 253 K."*

Page 7, lines 27 – 28: can you give the value of the homogeneous freezing temperature that you measured in your experiments?

We have added the homogenous freezing temperature that we observed in our experiments (i.e. referring to Fig. 3C). The sentence now reads: *"The homogeneous freezing threshold temperature observed in our experiments (237.0 K) agreed with previous reports (Benz et al., 2005; Schmitt, 2014)."*

Page 7, line 30: why a difference of 8 K? 245 K – 228 K = 17 K, or to which numbers do you refer here?

Here, we wanted to refer to the homogeneous freezing temperature of 236 K (now specified as 237.0 K, see above). We have revised this sentence as follows: *"This means that within a change of only 9 K from the homogeneous freezing temperature of pure water (237 K) to the expansion run started at 228 K, the ice-active fraction of the CFA_UK particles increased by almost 2 orders of magnitude."*

Page 8, line 3: Either give the abbreviation of ice-activated fraction or write it out but you do not need to do both every time you mention it.

Corrected.

Page 8, line 20: is it only the size of the particles or also their number that is different? In drop freezing assays there are usually very many particles per drop.

Yes, we will add the following statement after line 21: *"Moreover, in a drop freezing assay method, a droplet can contain many particles, whereas each cloud droplet activated in the AIDA chamber only contains a single particle."*

Page 8, line 34: First was the size discussion, the aerosol composition is rather "second", and the measurement techniques "third". Consider to revise.

We have now put the ordinal numbers (first, second, and third) in the right order (P8L38 to P9L1-6).

Page 8, line 36: Isono and Ikebe (1965) and Mason and Maybank (1958) are rather old references for this statement. Consider to add more recent ones.

We have added the following recent studies to the list – *"Fitzner et al., 2015; Harrison et al., 2016; Lupi et al., 2014"*

Page 9, line 5: Consider to replace "behavior" by "activity".

Changed.

Page 9, lines 21 – 31: The sequence of experiments is confusing. Table 2 suggests that the experiment with Tstart = 254 K is a new experiment. What is the history of this experiment? 250 K→ 254 K → 264 K? If you did two such experiments, you might show the results of both in Table 2.

We performed our first temperature-cycling and freezing experiments with CFA_UK following T_{start} ~ 250 K → 254 K → 264 K. Following the surprising and exciting results, we decided to repeat this experiment and confirm our observations, but this time with T_{start} ~ 251 K → 254 K → 263 K. We have now added the complete data from both experiments in Table 2. Since we obtained similar results from both experiments, we have consistently based our discussion on the data from the first set of our experiments (i.e. Experiment #9, #10 and #11). We have replaced the data in Fig. 4B and 5 with the corresponding data of CAINIC _19 (T_{start}~254 K). For the sake of completeness, we have added a figure (Figure S2) showing the data of the second temperature-cycling process (Experiment #12, #13 and #14) to the Supplementary Information document.

In the revised manuscript, we have deleted the confusing statement on line 22 ("We conducted two independent experiments …") and clarified the experimental procedure by adding the following statement to P9L10:

"Specifically, we conducted two independent series of experiments, each with a fresh load of aerosol particles, following the sequences T_{start} ~ 250 K → 254 K → 264 K (series I, experiment #9, #10 and #11, data shown in Fig. 4) and T_{start} ~ 251 K → 254 K → 263 K (series II, experiment #12, #13, and #14, data shown in Fig. S2). As the results from both series are very similar, we focus our discussion on the experiments conducted during series I."

Page 9, line 28: Here you write that the same processed CFA_UK sample was warmed to 264 K, however, the previous section refers to a different experiment. Please make the history of the samples more transparent.

We have addressed this issue in our response to your previous comment.

Page 10, line 10: could you mark the point where 244 K is reached in the figure e.g. with an arrow? This would increase the readability of the manuscript.

The point where the temperature reached 244 K has been marked.

Page 10, line 27: again, could you mark the point where 246 K is reached in the figure?

The point where the temperature reached 246 K has been marked.

Page 11, line 38: "be plerospheres" instead of "have plerospheres"

Changed.

Page 12, lines 29 – 31: this sentence could be formulated better. Do you mean "processing" instead of "process"?

We have reformulated this sentence to: *"During their residence time in the atmosphere, the CFA particles can be transported through different relative humidity and temperature regimes. If the particles were temporarily exposed to temperatures below 237 K at high ambient relative humidity, their ice nucleation ability might improve by the formation of ice-filled pores."*

Page 13, line 14: "with which" instead of "that"

Changed.

Page 13, line 12: "inherently expected" sounds strange. Try to improve formulation.

Here, we think you are referring to P13L1. We have modified this sentence to: "*By convective atmospheric dynamics, these pre-activated particles could then be released to lower altitudes and trigger ice formation at higher temperatures than expected from their inherent ice nucleation ability."*

Page 13, lines 13 and 14: "than" instead of "that"

Changed.

Page 13, line 24 – 25: "CFA will only show considerable or no ice nucleation potential." I do not understand the logics of this sentence. Please improve.

We have corrected part of this sentence to read: *"…CFA will show very poor or no ice nucleation potential at all."*

Page 13, lines 35 – 37: Relying just on pore volume and specific surface area is dangerous because the diameter is very relevant for pore filling and ice melting.

Definitely, the diameter of pores is a key parameter for pore-filling/ice melting, that is why we need to get the estimate right. Here, we suggest that in a situation where the diameter of pores cannot be directly and accurately measured, models that are used for such estimation should take into consideration both the pore volume and the specific surface area of such aerosol particles. We think that this type of model will be more useful and give a better estimate than current models that just rely on cylindrical- or spherical-shaped pore assumptions. We have modified this statement to read:

"We also suggest that in order to overcome the bias associated with pore models in estimating pore sizes and diameters for natural aerosol particles, a parameter based on the pore volume, pore size/diameter, and specific surface area should be adopted."

Figure caption to Fig. 1, line 5: consider to replace "irrespective" by "despite".

Replaced.

Figure caption to Fig. 2, lines 2 – 5: improve formulation.

Part of the Figure caption now reads: *"A schematic showing the temperature-cycling and freezing (TCF) process adopted in our experiments. The temperatures indicated by the grey circles represent the start temperatures (T_{start}) for the ice nucleation experiments conducted after the warming of the AIDA chamber. For each CFA sample, only a subset of the indicated starting temperatures was chosen to conduct the expansion cooling runs (see Table 2). The start temperature of the successive experiment was individually selected based on the degree of activity observed in the previous freezing experiment. …"*

Report 2

P2L28: Change "fissure" to "fissures".

Changed.

P4L18: Change to "This is the same set of samples…".

Changed.

E.g., P6L28-29, P7L11, SI P2L13, …: In some text passages, "&" is used instead of "and". I advise to avoid the use of the ampersand for consistency.

Changed.

P8L24-26: Please check Parungo et al. (1978). They found an enhanced ice nucleation potential of particles from a coal-fired power plant plume sampled on filters. This observation is reported for a temperature range between 263 and 253 K. Please revise the passage accordingly.

We have revised this passage accordingly: *"In another study, particles in a plume from a coal-fired power plant were not considered ice active at temperatures above 253 K (Schnell et al., 1976). However, when similar experiments were conducted at a higher supersaturation, the particles' ice nucleation ability increased, indicating that CFA particles could act as good INPs even at temperatures as high as*

263 K (Parungo et al., 1978). However, in these experiments, not many details on the exact experimental conditions are available for a direct comparison with our experiments."

P11L15: Change to "but that a network of … is necessary".

Changed.

P11L34: Change to "Another example is CFA Cy, which has…"

Changed.

References

Fitzner, M., Sosso, G. C., Cox, S. J. and Michaelides, A.: The Many Faces of Heterogeneous Ice Nucleation: Interplay Between Surface Morphology and Hydrophobicity, , doi:10.1021/JACS.5B08748, 2015.

Harrison, A. D., Whale, T. F., Carpenter, M. A., Holden, M. A., Neve, L., O'Sullivan, D., Vergara Temprado, J. and Murray, B. J.: Not all feldspars are equal: a survey of ice nucleating properties across the feldspar group of minerals, Atmos. Chem. Phys., 16(17), 10927–10940, doi:10.5194/acp-16-10927-2016, 2016.

Lupi, L., Hudait, A. and Molinero, V.: Heterogeneous Nucleation of Ice on Carbon Surfaces, J. Am. Chem. Soc., 136(8), 3156–3164, doi:10.1021/ja411507a, 2014.

Schnell, R. C., Van Valin, C. C. and Pueschel, R. F.: Atmospheric ice nuclei: No detectable effects from a coal-fired powerplant plume, Geophys. Res. Lett., 3(11), 657–660, doi:10.1029/GL003i011p00657, 1976.