Review Umo et al., 2019:

General comment:

Umo et al. present ice nucleation experiments performed within the large cloud simulation chamber AIDA with different coal fly ash (CFA) samples collected from five different power plants, one situated in the UK and four in the USA. Samples were sieved to isolate the size fraction up to 20 µm diameter and characterized by environmental scanning electron microscopy. In addition, their specific surface areas and pore volume were determined by argon adsorption measurements. There were quite significant differences between the ice nucleation activities of the different CFA samples. The UK sample, which is the best investigated one, showed a strong increase in the ice-active fraction for experiments performed just below the homogeneous freezing temperature of pure water. The authors concluded that this could be related to a pore condensation and freezing process (PCF). To further substantiate the role of pores for the ice nucleation ability of CFA, temperature cycling experiments were performed within the AIDA chamber by precooling the injected particles to 228 K at RH slightly below ice saturation before performing an expansion at warmer temperature. A strong pre-activation was found for the particles with the highest specific surface area and porosity. The authors conclude that the PCF mechanism could be prevalent for the ice nucleation at cirrus temperature and also significant for mixed-phase clouds when CFA particles are injected from higher altitudes.

This study presents innovative experiments aiming to elucidate the relevant ice nucleation mechanisms under cirrus and mixed-phase cloud conditions. Experiments were performed with CFA particles, which are a relevant class of ice nucleating particles from anthropogenic sources. The manuscript is well suited for Atmos. Chem. Phys. and can be recommended for publication after the following points have been addressed satisfactorily:

For all five CFA samples, pre-cooling experiments were described and discussed in the manuscript but only for the CFA UK sample, expansions that reached homogeneous freezing temperatures were mentioned. Have such measurements been carried out also for the CFA samples from the US? If yes, they should be described and discussed in the manuscript.

Experiments of processed and unprocessed samples with different starting temperatures are often compared without discussing the effect of the starting temperature. It would have been more meaningful if processed and unprocessed samples were compared in experiments with the same starting temperature. When such data is not available, the discussion needs to be improved to take the influence of the starting temperature better into account.

The individual experiments need to be characterized better. Table 2 gives an overview over all experiments relevant for this study; however, since the text and figures do not refer to this table, it does not help to obtain an overview over the experiments. Moreover, the table just gives the starting condition and lists no results except the observed freezing mode. The experiments are characterized by their starting temperature throughout the manuscript. Unfortunately, this information is not very useful because the RH and temperature at the freezing onset can only be guessed based on the starting temperature of the expansion. It would be helpful if the RH and the temperature of freezing onset together with f_{ice} were added to Table 2. Moreover, the experiment name should be mentioned in the figure captions, so that the exact conditions can be looked up in Table 2.

Throughout the manuscript, the consistency of the use of present and past tense needs to be checked. Some sentences are hard to understand and should be clarified. Some examples are given in the special comments but the whole manuscript should be worked over.

Specific comments:

Page 1, line 25: what is meant here by “partly”? Homogeneous freezing temperatures were only reached with CFA UK. Is this statement based on experiments that are not shown?

Page 4, Sect. 2.5: the adsorption and desorption isotherms should be given as supplementary information.

Page 5, line 26: is the end of the freezing experiment the end of the expansion?

Page 5, line 29: according to Sect. 2.3 the particles should be < 2.5 μm and not just < ~10 μm? Can you comment on this?

Page 7, line 2: do you mean “at” instead of “from”?

Page 7, lines 2 – 5: It is insinuated here that the generation method (dry vs. wet) might influence the ice nucleation activity of CFA particles. However, for a consistent comparison, the available INP area also needs to be taken into account. In Umo et al. (2015), the ice nucleation active site density does not rise above 10^5 cm^{-2}. The surface area of a spherical 1 μm radius particle is only 10^{-7} cm^2. Therefore, if the CFA particles had the same ice nucleation ability as reported in Umo et al. (2015), only a minor fraction of the particles should be active, which is indeed in accordance with the AIDA experiments.

Page 7, line 5: do you mean “at” instead of “from”?

Page 7, lines 12 – 13: “In this work, the average median particle diameter was 0.58 μm for our CFA samples theirs was size-selected to 0.3 μm.” Improve formulation.

Page 7, line 20: While the ice nucleation activity of the CFA particles investigated here is compared to the ones of other studies and other aerosol types, a comparison of the CFA particles investigated in this work among each other is lacking. Figure 7 shows that the ice nucleation activity of CFA_JA and CFA_Wh is one order of magnitude larger than the one of CFA_Cy and CFA_Mi. Are there differences in morphology, elemental composition, or surface functionalization that might explain the differences?

Page 7, lines 31 – 33: “This occurred at a lower RH_{ice} = ~105 % than the experiment with unprocessed CFA_UK particles which RH_{ice} = ~130 % (corresponding to water saturation).” Improve formulation.

Page 8, lines 7 – 8: Was this the third expansion of the same sample or an expansion with a new sample? Please clarify.

Page 8: lines 33 – 35: “At T_{start} = 253 K, the f_{ice} for CFA_Cy particles after the pre-activation process was ~0.86 % slightly lower than what was observed for the unprocessed CFA_Cy particles.” Improve formulation.

Page 9, lines 2 and 3: “T_{start} = 255 K (Fig. 7)”: Where is this starting temperature shown in Fig. 7?

Page 9, line 10: “256 K start temperature (Fig. 7)”: Where is this starting temperature shown in Fig. 7?

Page 9, lines 10 – 12: “Again, for the processed CFA_Ja particles, no appreciable enhancement of its ice formation abilities was observed as the f_{ice} at T_{start} = 249 K was 2 % at RH_{ice} = ~125 %.” Do you mean “enhancement compared to the unprocessed CFA_Ja?” Yet, Fig. 7 shows a decrease in f_{ice} rather than “no appreciable enhancement”. Please clarify.

Page 9, lines 15 – 16: “We cannot completely rule out that the actual formation mechanism in both scenarios after the temperature cycling is not via a condensational freezing pathway.” Formulate clearer, avoid double negative.

Page 9, lines 16 – 17: “This was not seen for the unprocessed CFA_Ja particles; after reaching water saturation, there was a time lag before ice particles were detected.” Is this a valid comparison? According to Fig. 7, the unprocessed CFA_Ja sample had a higher starting temperature. The onset of f_{ice} was therefore still at a warmer temperature for processed compared with unprocessed particles. This difference should be taken into account when discussing the effect of processing.

Page 9, line 26: T_{start} = 256 K is not shown in Fig. 7.

Page 9, line 28 – 29: “occurred in a shorter temperature step”: please formulate better.

Page 9, line 30: “pre-activation by PCF may not be very important compared to other particles that are less ice-active”. Improve formulation.

Page 9, line 33: “and the relative humidity which summary is given in Figs 6 & 7.” Improve formulation.

Page 10, lines 29 – 30: An additional factor that should be discussed here is the competition between pre-activation and immersion freezing. The ice nucleation mode is given in Table 2 but this table is neither connected with the text nor with the figures. Figure 7 needs to be improved to clearly state when water saturation is reached.

Page 10, lines 35 – 36: “Depending on the transport of CFA particles in the atmosphere, they can pass through different altitudes and temperature regimes which can naturally provide a temperature-cycling and freezing process for these particles to be pre-activated.” Improve formulation.

Page 11, line 8: “There is a need by the modelling community to study the impact that…”. Improve formulation.

Page 11, line 26: This is the first and only time that the chemical compositions of CFA particles is mentioned in this manuscript. Indeed, the chemical composition might be relevant to explain the differences in immersion freezing of the different CFA samples. If chemical composition is mentioned in the conclusions, it should also be discussed in the section “Results and Discussion”.

Page 18, caption of Table 1: How was the median diameter determined?

Page 18, Table 2: Consider to add f_{ice} and the freezing onset temperature to Table 2.

Figure 3: the tags on the y-axes should be increased for better visibility. The measurements shown in this figure should be related to the experiments listed in Table 2.

Figure 6, figure caption: It should be made clear whether the start temperatures of the experiments are shown in this figure.

Figure 7: It would be helpful to indicate the temperature where water saturation is reached for all experiments. Consider to add experiments with CFA_UK for better comparison.

Technical comment:

Page 4, line 10: “range” might be more adequate than “limit” since a range is given in brackets.

Page 4, line 28: “microscope” instead of “microscopy”.

Page 5, line 31: “β” does not appear correctly in the pdf.

Page 5, line 33: “γ” does not appear correctly in the pdf.

Page 8, line38: “their” instead of “its”.

Page 9, line 29: remove “an”.

Page 10, line 14: “than those of other CFA particles” instead of “than the other CFA particles”.

Page 12, line 3: “need” instead of “needs”.

Page 14, line 26: John is the first name of J. G. Morris. Please revise reference.