Review of "The immersion freezing behavior of ash particles from wood and brown coal burning" by Grawe et al.

This manuscript presents an interesting data set regarding the ice nucleating abilities of ash particles which are currently poorly understood. Given the lack of information on this topic, and the potential that ash particles may have to influence mixed-phase cloud formation, the present results are a valuable contribution to the ice nucleation community. Although the present manuscript possesses many similarities with the Umo et al. (2015) study, I still see a small level of novelty on it. I got attracted by the title of the paper which indicates that the ice nucleating abilities of wood and brown coal burning ash particles were studied, but I am disappointed that a clear explanation of why brown coal ash particles are better INP than the wood ash particles is not provided. Additionally, the conclusions are not clearly supported given the lack of some key experiments. Therefore, I think that this paper could be accepted in ACP only after the following points are clearly addressed. Note that this review was prepared without reading the comments given by referee #3; therefore, I apologize for any overlap between the two reviews.

Major comments:

1. Multiple charge correction was applied to the dry samples only. Which percentages of the particles were multiple charged? Based on what data was this correction conducted as the authors indicate that the UHSAS did not clearly detect the multiple charged particles? How good is the agreement between the SEM and the UHSAS at detecting multiple charged particles? I encourage the authors to report the size distribution for each sample and the resulted size distributions after size selection (300 nm).
2. There is a poor consistency in the experiments conducted with the 5 ash samples as shown in the table below. I am wondering why there is too much data missing. This lack of information reduces the robustness of the drawn conclusions regarding the particle generation methods and the effect of treating the samples with ultrasound. I suggest conducting more experiments to fill out the table below.
3. Is it the needle formation exclusive to fly-ash brown coal particles? Why SEM images of the bottom brown coal (suspension) are not presented? Why the SEM analysis was not applied to the wood ash samples? The authors indicate that the needle formation in the fly-ash brown coal particles may be cause by $CaCO_3$ which was formed by the presence of CaO detected by the atomic adsorption (AA) analysis. However, Figure 1 shows that the levels of CaO in the wood samples are much higher than in the brown coal samples. Therefore, it would be nice to see the SEM images of e.g. Spruce which has the highest concentration of CaO.
4. I am not fully convinced that the particles produced through the wet system are less efficient. The authors conducted a direct comparison of the ice nucleating abilities of the wet and dry generated particles; however, it is necessary to demonstrated that the monodispersity of the 300 nm particles from both system is comparable.

	Material	Fly/Bottom	Dry Generation	Wet Generation	Ultrasound	Without ultrasound	Filtered	SEM	AA
Wood	Spruce	B	X	X	X				X
	Birch	B	X						X
	Beech	B	X						
Coal	Brown coal	B	X	X	X			X	X
	Brown coal	F	X	X	X	X	X	X	X

Specific comments:

- Page 1, line 20: The Hallett-Mossop (1974) study introduces one of the multiple secondary ice formation mechanisms only. I suggest using a better reference such as Heymsfield and Willis (2014): Heymsfield, A. J., and Willis, P. (2014), Cloud conditions favoring secondary ice particle production in tropical maritime convection. *J. Atmos. Sci.*, 71, 4500–4526, doi:10.1175/JAS-D-14-0093.1.
- Page 1, line 21: I suggest replacing this reference with a more appropriate one.
- Page 2, line3: provide pages from P&K or the actual paper(s).
- Page 2, line 5: Cite the studies here.
- Page 2, line 12: "coal ashes contribute a major proportion of anthropogenic aerosol emissions" please provide the reference and number (estimate).
- Page 2, line 15: "yields global annual emissions of 30 Mt" is this comparable to mineral dust? Please provide the mineral dust annual emissions.
- Page 2, line 17: delete "however".
- Page 2, line 27: This needs to be better organized. The authors jump between old and recent studies back and forward.
- Page 2, line 32-34: "it could be shown that water soluble components were responsible for differences in the ice nucleation ability of fly ash samples from different power plants." Provide reference here.
- Page 3, line 5: "Umo et al. (2015) assumed that the different" Did they assume or did they provide evidence of?
- Pages 3-4, lines 32-1: "the number of particles with three or more negative charges was negligible". Please report the fractions or percentages of the multiple charged particles.
- Page 5, line 7: why 200 nm? What did not the authors try a smaller size to ensure that insoluble particles are not present? Please provide the resulting size distributions of the atomized filtered solution.
- Page 5, line 9: Why was not beech bottom ash particles analyzed by AA?
- Page 5: What are the uncertainties associated with the data presented in Figure 1?
- Page 5, lines 24-30: I am wondering how consistent and how reproducible is the data obtained from the dry generation system. Can the authors provide the size distributions obtained with the dry system separated by 15 or 30 minutes? Was it the multiple charged particles checked continuously with the UHSAS?
- Page 5, lines 33-34: Can the authors provide an activation scan with the filtered solution to confirm this?

- Page 8, lines 8-10: Which fraction of particles passed through? Was this confirmed with the size distributions from the UHSAS?
- Page 8, lines 15-16: Why the data from the water soluble material remaining in the filtered ash-water suspension is not shown?
- Page 10, lines 3-4: "There is a trend of beech bottom ash being the most effective". I am wondering why out of the three wood samples beech was the least studied (although it was the most effective at nucleating ice)?.
- Page 10, lines 15-18: "The fact that the wood ashes contain significantly more K than the brown coal ashes, which in this case is soluble, is a possible reason for the lower ice activity in comparison to the brown coal ashes. According to this, insoluble K could be the decisive element determining the freezing behavior of the brown coal Ashes" What about CaO?
- Page 10, lines 20-22: "the brown coal ash particles might be more efficient at nucleating ice because of surface defects such as lattice dislocations caused by impurities or crystallographic dislocations." This is not clearly supported by the presented data. I am wondering why the authors did not provide SEM images for the bottom wood ashes to compared the surface defects with those of the bottom coal ash particles.
- Page 11, Lines 1-3: "It has been shown that certain types of amorphous particles are able to nucleate ice (Murray et al., 2010b; Wilson et al., 2012), but it remains to be examined whether the amorphous components in fly ash are ice active as well". This only happens at very low temperatures relevant to cirrus clouds.
- Page 11, lines 4-6: "Tab. 1 and Fig. 4 additionally show the 5 parameters and fit curves to measurements with K-feldspar and mineral dust particles (K-feldspar, Arizona Test Dust, NXillite, Fluka kaolinite) coated with sulphuric acid (clay mineral baseline, Augustin-Bauditz et al., 2014). I may have missed but I could not find the Arizona Test Dust, NXillite, and fluka kaolinite data.
- Page 11, lines 26-30: "By counting ~ 900 particles on SEM images, it was determined that ~ 78 % of all particles are crystals. This value may be smaller in the flow tube as the fragile crystals could break upon impact on the filter leading to a multiplication. As only 22 % of the droplets contained a spherical fly ash particle during the experiments with the suspension sample (+US), the original data was corrected by a factor of 1/0.22 = 4.54 which is also shown in Fig. 4 for a direct comparability to the ice nucleation ability of dry particles from the same sample." How confident are the authors about this calculation? What is the uncertainty associate to it? 900±?? 78%±??, 22%±??, 4.54±??
- Page 11, lines32-35: "which is a clear lowering of the ice nucleation activity by a factor of 4 compared to dry particle generation, i.e., suspending the particles in water reduced their ice activity in the temperature range below -31 °C." I am not sure how valid is to directly compared the ice nucleating abilities of the ash particles generated from the wet and dry systems given that those obtained from the wet generation are not corrected for multiple charged particles.
- Page 12, lines 4-13: This is an interesting observation. I am wondering why the authors did not further expand this with other samples (e.g., bottom ash brown coal).
- Page 12, lines 4-5: "the fice values of the fly ash suspension which was not put in the ultrasonic bath are clearly lower than those 5 of the fly ash suspension with ultrasonic treatment." Here and along the results section, how many scans were conducted for each sample for each specific set of conditions? How reproducible are they?

- Page 14, lines 1-14: This part is too speculative with many unsupported assumptions. This should be deleted.
- Figures. Be consistent with the labels (a and b, or 1 and 2)
- Figure 1. I am not sure how useful is panel 2.
- Figure 3: It would be nice to have a similar image of the wet generation system of a bottom ash sample. Additionally, Fig. 1 indicates that Wood ashes contain much more CaO than the coal ashes. Therefore, the needle crystals should be more pronounced in the wood ashes if the reasoning presented here is correct.