

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

In this study the authors examine the immersion mode freezing efficiency of combustion ashes from different woods and brown coal burning using LACIS. Ashes from brown coal burning are found to exhibit higher nucleating abilities than those from the ashes generated from wood burning. The results presented here also seem to indicate that sample preparation can have impacts on ice nucleation efficiencies; an important point which will need to be considered in future studies.

My major comments below surround increasing the specificity and clarity of statements made. In particular, there are some vague statements made in attempting to account for observations in this work, and how it compares to others such as Umo (2015). Sentence and paragraph structure can also at times make it difficult to make out what is being said without multiple rereads of certain passages. While I recognize that investigating the nature of nucleating sites in a mixture as complex as ash is challenging, I suggest the authors could discuss the difficulties surrounding this endeavour and limit sweeping statements.

I am of the opinion that, following careful consideration of the points below and improvement of the manuscript in the areas listed, this could be accepted in ACP.

Main comments:

- The authors need to evaluate the use of 300 nm particles in this paper. It can be anticipated that physical and chemical composition varies with particle size, and in turn, perhaps the ice nucleating efficiencies. Without further experiments on larger particles it is difficult to see how the results of this study can be generalised to larger particles. I suggest that at the points in the manuscript where the authors are referring to size selected particles, they explicitly state this for clarity (e.g. section 3.1 L1-8).
- P2L12: “As a result, coal ashes contribute a major proportion.....”. This strong statement needs a reference.
- P2L13-15: On the one hand, “the importance of ash particles as potential INPs must be put into perspective by comparing with concentrations” yet in the next sentence an emission rate is given, not a concentration. While both sentences on their own are fine, having these sentences one after the other could be misleading.
- P4L6: Why is this important? Reference to publications demonstrating that surfaces typically used for ice nucleation assays interfere with the nucleation process would seem to be appropriate here, bearing in mind that many studies have been performed where droplets are supported by substrates and this does not appear to be an issue.
- P4L7-8: This statement also needs referencing and elaboration: what size droplets are you speaking of? Homogeneous nucleation CAN be probed in picolitre sized droplets on cold stages.
- P4L24: only three woods are investigated in this study; is this representative of all “deciduous vs. coniferous” trees?

- P5L1-2: What power does this sonicator deliver to the sample?
- P5 L6-7: is there any effects based on how long the samples are left to stir for? Also, what about aggregation in solution? Could you be removing particles smaller than 200 nm by filtration that have formed aggregates over the course of 24 hours of stirring?
- P5L6: Define “ice activity”
- P5L8: Can the authors say anything on the efficiency of these filters and pore size distribution? Can particles larger than 200 nm sometimes make it through? Also, are the authors defining “water soluble material” as any particle which will make it through a 200 nm filter? This should be stated if so.
- Figure 4: How is the “homogenous” region here determined? Citation of the appropriate parameterization(s) should be added.
- Fig 4: State in the legend and text exactly what the “clay mineral baseline” is; i.e. what clays were used?
- Fig 4: Harrison et. al. ACPD (2016) show that K-Feldspar can have different activities: specify exactly which K-Feldspar was used.
- P10L7-8: I don't have an issue with this statement as such, but I think it's important to point out that onset freezing temperatures cannot be used for others to compare with. A simple phrase along the lines of “For within these experiments we can compare our onset freezing temperatures, which....”
- P10L9-13. This section is vague, and should be strongly caveated by noting that the authors don't know what the explanation for the differing nucleation activities is, or further experimental evidence showing that SiO<sub>2</sub> is indeed responsible for this behaviour should be added. For both this point, and the point below, a statement explaining why it is difficult to conclusively identify the active site in such a complex sample at the beginning would definitely help here!
- Page 10L 14- Page 11L3: All of this section is very speculative and at points vague, and I suggest it be rewritten to make it more concise or removed. After providing several possible explanations for their observations, the authors note “other properties such as ice active sites on the particle surface might affect the ice nucleation ability of a substance.” What does this mean? Taking the Vali (2015) definition of a site being a “Preferred location for ice nucleation on an INP”, this statement is rather meaningless.
- Page 11 L5: again, which K-Feldspar was used?
- Page 11 L6: Is it reasonable that clay minerals treated with sulfuric acid are termed the “clay mineral baseline”? Surely this should be justified. Also, if justified, what it is should be stated explicitly in figure 4.
- Page 11 L17-page 12L3: There are a lot of concepts in this rather long paragraph: separating into at least two smaller paragraphs would help readability greatly
- P12L1-3: If it's not the physical or chemical particle properties changing and leading to a destruction of active sites, what else could it be? This non-specific nature of this statement makes it vacuous. The word “likely” doesn't help here either.

- Section 3.3: Discussion of possible differences in composition and hence ice nucleating abilities due to size selection of particles should be presented here.
- Page 13L12: Agreed, but explain why you would not expect it. Reference where appropriate
- P13L13-14: Again, vague. Is there nothing more which can be said here other than the samples were not “completely identical”
- P14 L26: All clay minerals? Which K-Feldspar?

Typos/other:

P3L1: Is the cold stage(s) used by Umo (2015) the same as that used in Murray 2010(a)?

P1L3: Largest rather than biggest

P8L26 and fig 4. SMB should be SBM no?