Anonymous Review of Ice nucleation ability of Ammonium Sulfate aerosol particles internally mixed with Secondary Organics

Anonymous Reviewer

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1 Summary

In this work, Bertozzi et al. investigated the heterogeneous ice nucleation ability of ammonium sulfate (AS) coated with α-pinene SOA (dark ozonolysis) from -65 to -50 °C. These mixed particles were generated to have a core-shell morphology, or to change their morphology through droplet activation and drying or homogeneous ice nucleation and subsequent sublimation. Ice nucleation by these particles was measured by the AIDA chamber, and also the continuous-flow diffusion chamber INKA. Both of instruments are well known and well characterized, and their operation and limitations are well represented in this paper.

This manuscript consisted of four different experiments. Ice nucleation on bare AS, AS with thick and thin SOA coatings (AS+SOA), AS+SOA particles that underwent droplet activated and subsequent evaporation below the AS deliquescence point, and AS+SOA particles that underwent homogeneous ice nucleation and subsequent sublimation. These experiments were well planned and well thought out. The last two experiments were novel, and take advantage of the unique capabilities of combining the AIDA chamber with INKA.

In this work, the authors found that their ice nucleation onsets for ammonium sulfate and thickly coated AS+SOA particles agreed with previous literature values, with bare AS having an ice nucleation onset of $S_{ice} = 1.3$, and the SOA coatings inhibiting ice nucleation until S_{ice} was $\geq$1.45. They also found that organic mass fractions of 5-8% were enough to cause this shift in S_{ice} onset. Finally, the authors found that the droplet activated/evaporation and ice formation/sublimation changed the S_{ice} onsets for even thickly coated AS+SOA. For these particles, the onsets were in between bare AS and thickly coated AS+SOA in a core-shell configuration, or an S_{ice} of 1.35.

The paper was a refreshing read. It was well written and well organized. Overall, I have only two major comments, and a few minor/technical comments.

2 Major Comments

1. On P7L212, you use the median diameter to derive your coating thickness. How good of an assumption is this? For example, how does the mass fraction of organics change with size for a single experiment, and how does the coating thickness change a function of size for a given mass fraction? You should be able to derive these quantities from the SMPS and AMS.

2. On P15L490, you start discussing the S_{ice} onset for the freeze-dried particles. I was surprised to see that the particles did not nucleate ice extremely efficiently as suggested in the paper by Adler et al, 2013. This suggests that the AS+SOA particles were not glassy, or had sufficient self-diffusion rates to not retain a perfect imprint of the sublimated ice. This needs to be discussed further in the paper. You should be able to tell from the IR if the sulfate was locked in glassy matrix or effloresced, correct?

3 Minor/Technical Comments

- Title: I believe that "Ammonium, Sulfate, Secondary, and Organics" should all be lowercase.
- P2L38: To me, the phase "allows to" here reads very awkward. Perhaps change to "allows us to?"
- Equation 1: What was the prescribed density of organics in this work?
- P8L249: Here the phrase "allow to" shows up again and reads slightly awkward. I would suggest deleting it and changing "evacuate" to "evacuates."
- P9L274: Perhaps change "thanks to" to "because of."
- P9L285: Perhaps change "allows to minimize" to "minimizes"
- P10L305: Change "that" to "which."
- P11L340: Perhaps change "reddish" to "red." I see that the symbols are slightly different in color, but, in this reviewer's opinion, it's okay to simple say "red."
- P11L341: Perhaps change "greenish" to "green."
- P13L415: The sentence "Note that in this specific experiment the chamber walls were not coated with ice" could be deleted. It feels extraneous to this reviewer.
- P14L434: Could cite the work of Freedman et al., 2020 here.

- P15L468: It seems as if the authors are suggested that the coated particles were not heterogeneously ice active, when Figure 5 seems to suggest they are. Perhaps the authors mean that they have higher onsets than homogeneous nucleation?
- P16L518: Same comment as the one previous–the authors are implying that thickly coated particles are not "active as INPs"