We thank the anonymous Reviewer for the interesting and useful comments, which help to improve the quality of our article.

Our replies are reported in the following with blue fonts, Reviewer's comments are reported in black. Reporting the changes made in the manuscript, we indicate with strikethrough fonts the text previously present in the manuscript and now removed, and with bold fonts the additions we suggest to include in the revised manuscript. References to pages and lines in the responses refer to the revised version of the manuscript, unless noted otherwise.

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.

Thanks for this interesting comment. We have looked at the size-resolved particles' chemical composition as measured by the AMS. In Fig. A we show the results for two exemplary experiments: B1 (coating performed in the APC chamber) and C3 (coating performed in the AIDA chamber). The normalized mass concentration of organics (green), sulfate (red), and ammonium (orange) are shown as a function of the vacuum aerodynamic diameter D_{va}. The three curves overlap nicely for particles with vacuum aerodynamic diameter equal to or larger than the main mode in both experiments, meaning that the organic mass fraction f_{org} is not size-dependent. The slightly higher organic mass fraction for smaller particles can be related to some pure SOA particles nucleated during the coating process, or a higher mass fraction condensed on the smaller AS seeds.

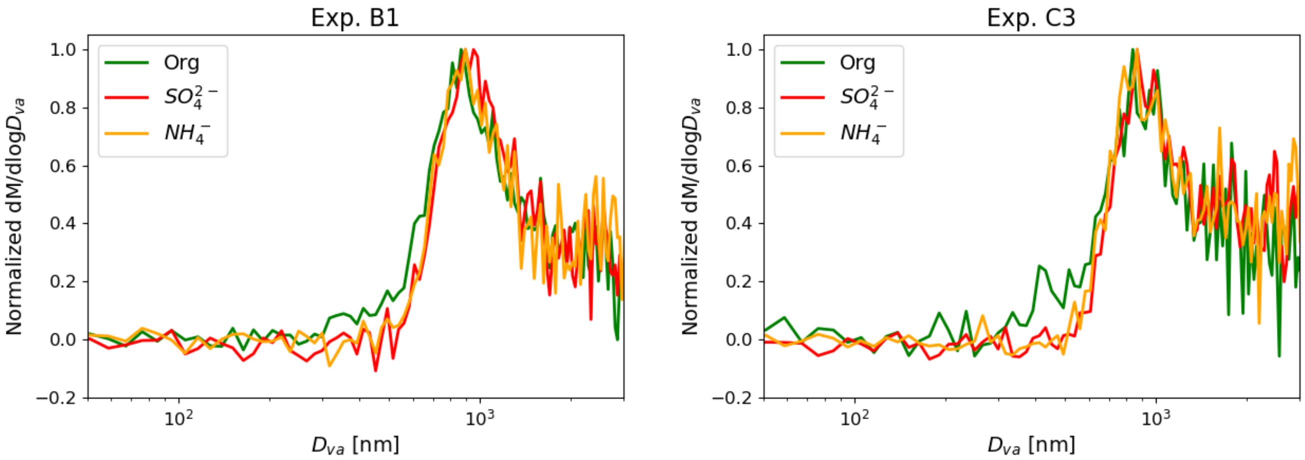

Figure A: Normalized mass distributions measured by the AMS after coating for experiments B1 and C3 as a function of the aerodynamic vacuum diameter D_{va}. Organics are shown in green, sulfate in red, and ammonium in orange.

As suggested by Referee #1, we then evaluated the coating thickness d as a function of the seed particle size D_p for a given organic mass fraction f_{org}. The equations and the text in the manuscript have been adapted accordingly.

From <u>line 217:</u>

"The organic mass fraction values $\boldsymbol{f_{org}}$ reported in Table 1 refer to the organic content at the end of the coating procedures, i.e., regarding experiments of type C, they refer to the organic content after the last coating step. **The size-resolved measurements of the chemical composition of the particles by the AMS indicate that the organic mass fraction is constant and not a function of the particle size.**

To estimate the thickness of the organic coating, d, we combined the size distribution of the pure crystalline AS particles from the SMPS measurements and the organic mass ~~concentration Morg~~ **fraction $\boldsymbol{f_{org}}$** from the AMS. Thereby, we assume the crystalline AS seeds to be ~~monodisperse~~ spherical particles with diameter D_p ~~equal to the median diameter measured before the coating procedure by the SMPS (reported in Table 1). Furthermore, we assume that~~**,** **and** the organic material ~~is~~ **to be** evenly distributed on ~~the particles~~ **their** surface, leading to a spherical organic shell. The coating thickness d is thus calculated assuming a perfectly concentric core shell morphology as follows:

~~$D_p^{coated} = (\frac{6}{\pi}\frac{M_{org}}{C_n \cdot \rho_{org}} + D_p^3)^{\frac{1}{3}}$~~

$$d = \frac{D_p^{coated} - D_p}{2}$$

~~where D_p^{coated} is the~~ **The** diameter of the ~~SOM-coated AS particles and C_n is the total particle number concentration.~~ **coated particles $\boldsymbol{D_p^{coated}}$ is calculated by considering the size-dependence of the particles' organic mass $\boldsymbol{M_{org}(D_p)}$:**

$$\boldsymbol{D_p^{coated}(D_p) = [D_p^3 + \frac{6}{\pi}\frac{M_{org}(D_p)}{\rho_{org}}]^{\frac{1}{3}}}$$

With:

$$\boldsymbol{M_{org}(D_p) = \frac{f_{org}}{1 - f_{org}} \cdot \rho_{AS} \cdot \frac{\pi}{6} D_p^3}$$

The resulting estimated organic coating thickness ~~for all the experiments~~ **corresponding to the median diameter of the particle population** is reported in Table 1. **In Table 1, we also indicate between brackets the estimated coating thickness for seed particles with diameters of 300 and 500 nm, i.e., the size range which comprises the major particle mode in the number size distribution (see below and Fig. 3 for the number size distribution measurements)**. The thickness estimated for the thickly SOM-coated AS particles (experiments of type B, D and E) ranged from ~~42 nm to 88 nm~~ **24 nm to 54 nm**. Thinner coatings were obtained at the end of the coating experiments performed in the AIDA chamber (experiments of type C) with ~~9 nm~~ **6 nm**, 5 nm, and ~~11 nm~~ **8 nm** coating thicknesses."

2. On P15L490, you start discussing the Sice 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?

- Thank you for raising this interesting aspect that certainly warrants further discussion in the manuscript. There are a couple of previous studies that observed an efficient preactivation of organic aerosol particles after ice-cloud processing, which may be explained by the formation of a porous structure or the retention of an imprint of the sublimated ice on the highly viscous organic material. Wagner et al. (2012) investigated four different organic solutes, raffinose, 4-hydroxy-3-methoxy-DL-mandelic acid (HMMA), levoglucosan, and a multicomponent mixture of raffinose with five dicarboxylic acids and ammonium sulfate, and observed that the ice-cloud processed glassy aerosol particles catalyzed ice formation at ice saturation ratios between 1.05 and 1.30. These experiments were also conducted in the AIDA chamber. Recently, Kilchhofer et al. (2021) confirmed the improved ice nucleation ability of cirrus-cloud processed raffinose particles in dedicated CFDC ice nucleation measurements. However, repeated ice nucleation experiments with secondary organic aerosol particles produced from the ozonolysis of α-pinene performed by Wagner et al. (2017) in the AIDA chamber showed a much lower susceptibility towards preactivation compared to the above mentioned organic solutes. For example, the ice nucleation onset of the ice-cloud processed α-pinene SOA particles at 220 K was at an ice saturation ratio of about 1.45. A similar ice nucleation threshold was found in an experiment where highly porous α-pinene SOA particles were formed by a freeze-drying process at 243 K (as in the current study) and then cooled to cirrus temperatures to probe their ice nucleation ability.
  So far, there is therefore no experimental evidence that the freeze-dried α-pinene SOA particles must necessarily be extremely efficient INPs. Furthermore, Adler et al. (2013) showed that in mixed AS/OM particles with a 1:1 molar ratio, the formation of a porous structure was reduced compared to the pure organic particles – potentially adding to the effect that the freeze-dried AS/SOM particles are not extremely efficient INPs.
  In our discussion of the freeze-drying experiment, we have only briefly mentioned the preactivation experiments by Wagner et al. (2017) on P16L497 (referred to the original version of the manuscript).

  We propose to extend this discussion as follows (lines 549ff):
  "Third, the porous organic material formed during the freeze-drying process could be a better ice nucleus on its own via the pore condensation and freezing mechanism **or by retaining an imprint of the sublimated ice on the highly viscous organic material.** ~~However, Wagner et al. (2017) found in their study with pure secondary organic aerosol particles that the porous, freeze-dried particles do not have an improved ice nucleation ability compared to the unprocessed particles.~~ **Previous studies observed an efficient preactivation of glassy organic aerosol particles after ice-cloud processing (Wagner et al., 2012; Kilchhofer et al., 2021). Wagner et al. (2012) investigated in the AIDA chamber four different organic solutes, raffinose, 4-hydroxy-3-methoxy-DL-mandelic acid (HMMA), levoglucosan, and a multicomponent mixture of raffinose with five dicarboxylic acids and ammonium sulfate, and observed that the ice-cloud processed glassy aerosol particles catalyzed ice formation at ice saturation ratios between 1.05 and 1.30. Kilchhofer et al. (2021) confirmed the**

improved ice nucleation ability of cirrus-cloud processed raffinose particles in dedicated CFDC ice nucleation measurements. This raises the question of why the ice-cloud processed AS/SOM particles (observed ice onset at S_{ice} = 1.41) do not reveal an even better ice nucleation ability. However, repeated ice nucleation experiments with secondary organic aerosol particles produced from the ozonolysis of α-pinene performed by Wagner et al. (2017) showed a much lower susceptibility towards preactivation compared to the above mentioned organic solutes. For example, the ice nucleation onset of the ice-cloud processed α-pinene SOA particles at 220 K was at an ice saturation ratio of about 1.45. A similar ice nucleation threshold was found in an experiment where highly porous α-pinene SOA particles were formed by a freeze-drying process at 243 K (as in the current study) and then cooled to cirrus temperatures to probe their ice nucleation ability. Furthermore, Adler et al. (2013) showed that in mixed AS/OM particles with a 1:1 molar ratio, the formation of a porous structure was reduced compared to the pure organic particles, adding to the effect that the freeze-dried AS/SOM particles are not extremely efficient INPs."

- Regarding the phase of the AS fraction in the particles, the infrared spectrum of the freeze-dried a-pinene SOA particles in the regime of the ν_3 (SO_4^{2-}) mode was similar to spectra A and C in Fig. 7c, indicating that the sulfate was effloresced.

  This is now mentioned in the revised manuscript, we added lines 526-528:
  "After the chamber cooling, the infrared spectrum of the freeze-dried AS/SOM particles in the regime of the ν_3 (SO_4^{2-}) mode was similar to spectra A and C in Fig. 7c, indicating that the sulfate was effloresced."

Minor/Technical Comments
Title: I believe that "Ammonium, Sulfate, Secondary, and Organics" should all be lowercase.
We agree and corrected the title.

P2L38: To me, the phrase "allows to" here reads very awkward. Perhaps change to "allows us to"?
Done.

Equation 1: What was the prescribed density of organics in this work?
The density of the organic material is estimated to $1.25\ g/cm^3$. The density value is introduced at P7L209, referred to the original manuscript version (line 216 in the revised version).

P8L249: Here the phrase "allow to" shows up again and reads slightly awkward. I would suggest deleting it and changing "evacuate" to "evacuates".
Done.

P9L274: Perhaps change "thanks to" to "because of".
Thanks for the suggestion. Unfortunately, the sentence was wrong, now it reads (lines 287-288): "Thus, ~~a~~ **because of the non-linear dependency of the equilibrium vapor pressure as a function of temperature, a** supersaturation profile establishes ~~thanks to the faster diffusion of water vapor than heat~~ **between the walls**."

P9L285: Perhaps change “allows to minimize” to “minimizes”
Done.

P10L305: Change “that” to “which”.
Done.

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”.
Yes, agreed and modified.

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.
The usual preparation procedure of the AIDA chamber described at P5L148 (in the original manuscript) includes the humidification step, that leads to the formation of a thin ice layer on the chamber walls. However, when ice is present in the chamber, it acts as a source of water vapor and does not allow to reduce the relative humidity, as needed for experiments of type D to induce the crystallization of the AS component. We think it is important to mention this experimental detail, but will extend our statement to better explain why we deviated here from the standard preparation procedure:

Lines 444-445 now read:
“Note that in this specific experiment the chamber walls were not ~~coated with an ice layer~~ **ice-coated, because the ice layer would have acted as a source of water vapor and prevented the reduction of the relative humidity.**”

P14L434: Could cite the work of Freedman et al., 2020 here.
Thanks for the suggestion. The citation has been added.

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”.
Thank you for this comment. The ice nucleation onsets of the thickly SOM-coated AS particles are close to the homogeneous freezing threshold (Koop et al., 2000), but such particles are certainly not comparable to aqueous solution droplets for which the water-activity based parameterization for homogeneous freezing was developed. We will remove statements like that thickly coated particles are “not active as INPs” from the manuscript text and be more careful in our description.

Lines 511-513 become:
This might provide a pathway to ~~transform~~ **increase the ice nucleation ability of** internally mixed AS/SOM particles with high organic mass fraction ~~into heterogeneously ice active~~

~~particles~~, because a porous organic coating might less efficiently cover the ice ~~active~~ **nucleating** active sites of the crystalline AS component.

Lines 590-592 have been changed to:
"We performed two experiments to investigate whether there are pathways by which **the ice nucleation ability of** mixed AS/SOM particles with high organic weight fractions (25−50 wt%), as often found in the upper troposphere (Froyd et al., 2009), could ~~still be active as INPs~~ **be increased**."

Corrigendum

Additionally, we modified the manuscript in the following parts due to the presence of errors.

Lines 287-288:
Thus, ~~a~~ **because of the non-linear dependency of the equilibrium vapor pressure as a function of temperature, a** supersaturation profile establishes ~~thanks to the faster diffusion of water vapor than heat~~ **between the walls**.

Line 353:
"Effects" changed to "affects"

Line 429:
"Measurments" changed to "measurements"

We removed the sentence at lines 481-483, referred to the original version of the manuscript:
~~"After the freeze-drying process, the residual particles appeared at slightly larger sizes in the optical particle counters, indicating a change in the morphology and/or in the refractive index of the processed particles."~~
In fact, the change detected by the optical particle counter is probably related to the deliquesced ammonium sulfate component.

Figure 5b: in the legend the reference to "Ladino, 2016" has been changed to "Ladino, 2014".

References

Adler, G., Koop, T., Haspel, C., Taraniuk, I., Moise, T., Koren, I., Heiblum, R. H. and Rudich, Y.: Formation of highly porous aerosol particles by atmospheric freeze-drying in ice clouds, Proc. Natl. Acad. Sci., 110(51), 20414–20419, doi:10.1073/pnas.1317209110, 2013.

Kilchhofer, K., Mahrt, F. and Kanji, Z. A.: The Role of Cloud Processing for the Ice Nucleating Ability of Organic Aerosol and Coal Fly Ash Particles, J. Geophys. Res. Atmos., doi:10.1029/2020jd033338, 2021.

Wagner, R., Möhler, O., Saathoff, H., Schnaiter, M., Skrotzki, J., Leisner, T., Wilson, T. W., Malkin, T. L. and Murray, B. J.: Ice cloud processing of ultra-viscous/glassy aerosol particles leads to enhanced ice nucleation ability, Atmos. Chem. Phys., 12(18), 8589–8610, doi:10.5194/acp-12-8589-2012, 2012.

Wagner, R., Höhler, K., Huang, W., Kiselev, A., Möhler, O., Mohr, C., Pajunoja, A., Saathoff, H., Schiebel, T., Shen, X. and Virtanen, A.: Heterogeneous ice nucleation of {α}-pinene SOA particles before and after ice cloud processing, J. Geophys. Res., 122(9), 4924–4943, doi:10.1002/2016JD026401, 2017.