Author Response

General Comments

To address reviewer comments regarding the effects of surface area and singly charged aggregates on ice nucleation in our study, we adjusted the multimodal curve fitting algorithm to a lower time resolution and increased the number of modes assigned. This procedure is now detailed in the text and tracked changes documents. Due to these changes, improvements in derivative products from the multimodal algorithm have been updated along with a change to the concentration data where it was previously averaged at a 5 minute interval but is now at the same time resolution of the SEMS.

Due to these changes, values throughout the text that depend on SEMS data or its analysis have shifted, including onsets and subsequent statistics. As can be seen in the tracked-changes document, none of these change statistical significance levels or conclusions. These values reflect the an improvement to the previous version and more accurate as requested in peer-review.

Furthermore, we have made grammatical changes throughout the article to improve clarity and flow along with the primary additions requested by viewers.

RC1 of "Ice nucleating properties of α-pinene- and limonene-derived secondary organic aerosol under cirrus conditions" by Rapp et al., 2026

Anonymous Referee #1, 26 May 2026
https://doi.org/10.5194/egusphere-2026-2458-RC1

RC1: Rapp et al. present a study investigating the ice nucleating ability of secondary organic aerosols (SOA) derived from α-pinene and limonene precursors under cirrus-relevant conditions, for both self-nucleated and seed-particle experiments. They investigate the influence of precursor identity, SOA chemical composition, volatility, and pre-cooling on ice nucleation efficiency, and report heterogeneous ice nucleation more clearly for limonene-derived SOA. The use of glass transition temperature (T_g) as a predictor of heterogeneous ice nucleation was not supported by the results.

The manuscript is well written and appropriately organized. The figures effectively display the relevant information, and the references are appropriate and comprehensive. In my opinion, this work makes a meaningful contribution to the understanding of ice nucleation properties of SOA from biogenic sources. However, the limited temperature range explored represents a notable weakness, as discussed below. I recommend publication after minor revisions, detailed in the comments that follow.

We thank the reviewer for their thorough review of our manuscript and their comments/questions. We have compiled the responses to comments and question below which are reflected in a revised manuscript and supplement.

General Comments

Limited temperature range. Only two temperatures were explored (−40 and −45°C), while cirrus clouds form down to ∼−85°C. The authors should discuss how representative their findings are of the full cirrus regime and provide a mechanistic explanation for the absence of heterogeneous freezing at −45°C.

For these experiments, we selected the temperature range of -45 to -35°C. We did not observe ice nucleation at the warmer temperatures, indicating an upper temperature limit (~ -40°C) where these particles are unlikely to be cirrus-relevant (now discussed in Sect. 5.2 Atmospheric implications).

We agree on the limitation of the findings, primarily due to the lower temperature range. For these experiments, -45°C served as the lower functional limit of the current SPIN instrument. Specifically, the SPIN in use is SPIN001 (or MIT SPIN), which served as the first version prior to commercial production (Garimella et al., 2016). This configuration precludes controlled measurements below ~ -46°C due to the temperature range of the warm-wall refrigerant (Rapp, 2026; Wolf et al., 2020). While modifications to SPIN have been performed to bypass this restriction (Welti et al., 2020), we did not implement such modifications for these experiments.

Limits in instrumentation as described above are now included in Sect. 2.6.2. The limitations in interpreting data across the full range of cirrus is included in Sect. 5.2.2.

Phase state is inferred, not measured. Phase state is inferred from T_g calculations rather than measured directly. The authors should state all assumed parameters explicitly and acknowledge associated uncertainties. Direct viscosity measurements (e.g., Virtanen et al. 2010) would benefit the interpretation of results.

We agree that the glass-transition temperature and particle phase state were inferred rather than measured directly. We have revised Section 2.5 to provide the complete assumed parameters and equations for both the volatility-based and molecular-composition-based $T_{g,org}$ estimates. The revised section now explicitly states the assumed temperature (298 K), activity coefficient ($\xi = 1$), ideal-mixing approximation ($C^* = C^0$), Gordon-Taylor constant ($k_{GT} = 1$), and use of normalized Vocus-CIMS signal intensities as proxies for organic mass fractions under an equal-sensitivity assumption. We also provide a description of the Li et al. (2020) chemical composition equation and all coefficients used in that calculation in the supplement (Section S1.3).

We have also distinguished thermogram-fitting precision from parameterization and systematic uncertainties. Specifically, the ±21 K interval associated with the DeRieux parameterization (DeRieux et al., 2018) is treated as a conservative model-uncertainty bound. We further acknowledge uncertainties arising from the thermogram-to-C* calibration, empirical T_g parameterizations, potentially incomplete molecular coverage, and compound-specific sensitivity. Throughout the revised manuscript, we therefore refer to $T_{g,org}$ as parameterized and phase state as inferred or predicted rather than directly measured.

Our response regarding direct viscosity measurements is included in the corresponding comment in the Specific Comments section.

Role of multiphase chemistry remains unresolved. The logical chain connecting seed acidity to SOA phase state and ice nucleation efficiency is central to the study's motivation but is never fully articulated. The authors should either state this chain explicitly.

We thank the reviewer for this comment and have revised the introduction so that this logic is stated explicitly.

Atmospheric implications need development. The section should additionally address: whether observed S_{ice} thresholds occur in the real atmosphere; whether activation fractions are sufficient to compete with homogeneous freezing; the role of updraft velocity in phase state retention; and the source region relevance of limonene versus α-pinene.

These topics have now been discussed in context with our results in Sect. 5.2. Specifically, a section was added dedicated to discussing the relevance of our results to cirrus cloud measurements and more detail has been added to the discussion on various freezing mechanisms (including details pointed out in specific comments below).

Self-nucleated experiment conditions need clarification. It is unclear whether self-nucleated and seeded experiments were conducted under identical conditions. If generation settings were optimised to minimise self-nucleation in seeded runs, the use of the same settings for seedless runs should be justified.

Conditions used in generating self-nucleated BSOA are included in Table 1. The experiment type has now been added as a column to emphasize the last two entries correspond to self-nucleated particles. Furthermore, Sect. 2.2.2 has been revised to clarify both the general operating conditions for all experiments and how self-nucleated experiments were performed.

Specific Comments

L11. Please clarify in what sense the contribution of BSOA to cirrus cloud formation remains unresolved. What specific aspects (formation mechanism, INP efficiency, atmospheric abundance, or radiative effect) are being referred to?

We have specified that the primary uncertainty regarding BSOA and cirrus formation remains the role of physicochemical properties and discrepancies in laboratory vs. atmospheric observations. The L11-14 now reads:

"The contribution of biogenic secondary organic aerosol (BSOA) to cirrus cloud formation remains unresolved, contributing to uncertainty in aerosol-cloud interactions in global climate models. In particular, the role of physicochemical properties in modulating their apparent ice nucleating efficiency remains unclear. This is further compounded by discrepancies between laboratory and atmospheric observations of ice nucleating efficiency. Laboratory studies report highly variable ice nucleating efficiencies for BSOA, suggesting that these particles may act as either homogeneous or moderately effective heterogeneous ice nuclei."

L14. The relevance of limonene as a SOA precursor should be briefly justified. Are there estimates of global or regional limonene emission fluxes that support its inclusion alongside the more widely studied α-pinene?

We thank the reviewer for raising this point. In the introduction a reference to the MEGAN2.1 model (Guenther et al., 2012) was provided but emissions are now specified along with a reference to a study with comprehensive citations for the study of limonene (Saathoff et al., 2009). This portion of the abstract now reads:

"Here, we investigate the deposition ice nucleating properties of BSOA derived from α-pinene- and limonene, two well-studied abundant monoterpenes. Our investigation included both self-nucleated particles and BSOA coatings on ammonium sulfate and partially neutralized sulfate seed particles."

The introduction now includes:

"BVOC emissions are dominated by isoprene and followed by emissions of as α-pinene, t-β-ocimene, β-pinene, and limonene (Guenther et al., 2012). In particular, α-pinene and limonene are well-studied compounds (Saathoff et al., 2009) *with high emission rates (66.1 and 11.4 Tg yr^{-1} respectively), motivating their selection for this study."*

L65. Please briefly define subvisible cirrus clouds for the reader.

We have now included a citation for an optical thickness (τ) of 0.03 (McFarquhar et al., 2000) to delineate between subvisible cirrus and visible cirrus.

L120. Please briefly describe what multiphase reactions are in this context, as this concept is central to the study's motivation but is introduced without definition.

We have included a brief description in context with this work along with a relevant citation defining multiphase reactions on acidified sulfate aerosol.

L124. Please provide the complete logical chain connecting seed acidity to phase state and from phase state to ice nucleation efficiency. As written, this connection is implied but not explicitly stated, making it difficult to follow for readers less familiar with the topic.

We thank the reviewer for this comment and have modified the paragraph to explicitly state this connection.

L137. Phase state is inferred from composition, volatility, and T_g in this work. Could phase state or viscosity have been measured more directly, for example, using the particle bouncing technique described in Virtanen et al. (2010)? Please discuss why such measurements were or were not feasible in this experimental setup.

We agree with the reviewer here as this is a limitation of this study. Ideally, phase state measurements would have been accomplished via filter collection and subsequent poke-flow measurements. Self-nucleation was attributed to flow irregularities upstream of the PAM-OFR which would be exasperated by sampling flows for filters. In retrospect, asynchronous filter collection could have benefitted the study but would also introduce uncertainties in whether the analyzed particles were representative of particles entering SPIN because of the effects of shipping/storage on collected particles. Given the above, we opted to forego filter collection due to the complexity of the existing experiments.

Regardless, given the results that ice nucleating efficiency for the BSOA examined are unsupported by T_g estimates, we have included a brief statement in the conclusion that future BSOA studies should probe viscosity directly rather than relying on inferred properties. We have also included a reference to (Virtanen et al., 2010) and a brief statement on why direct measurements of viscosity are not always feasible on L85.

L196. Is the 10L mixing volume also applicable to self-nucleated SOA experiments where no seed particles are present? Please clarify.

The 10L mixing volume was applicable to all experiments and this has now been clarified in the methods.

L204. The paragraph describes conditions for generating seeded SOA; however, self-nucleated SOA experiments were also conducted (Table 1, Table S2). Please clarify whether the described conditions also apply to self-

nucleated runs, or specify any differences in setup. In particular, if O_3 concentration and UV intensity were optimised to minimise self-nucleation, why were the same settings used for seedless runs?

See response to corresponding major comment above.

L214, Figure 2. Several clarifications are needed: (i) add labels or a legend to panels to make clear which refers to PNS and which to PNS-coated particles; (ii) define and explain the NRMSE values shown in the panels; (iii) explain the different mode shifts observed for different modes in the coated PNS experiment: is this physically interpretable or an artefact of size resolution?

We thank the reviewer for this comment and have added additional details to Figure 2 and the preceding text. Each panel is titled with the respective particle type along with labels in the top-right corner that are explained in the caption. The NRMSE is defined in Sect. 2.3 proceeding the plot along with a description of the underlying physical interpretation of the modes.

L244. Would it have been possible to conduct PALMS-NG analysis specifically on ice crystal residuals, targeting only particles that activated as ice, separately from the bulk aerosol population? This would have provided direct chemical characterization of the INP-active fraction and is worth discussing even if not performed.

Performing PALMS-NG analysis on ice crystals residuals would have required a PCVI and consequently the removal of the SPIN OPC. For this study, determining the ice nucleating properties of the BSOA was the primary focus, with compositional information serving as supplementary information. We have included a statement clarifying why IR analysis was not performed in Sect. 2.4.1.

L245. Are there sections of the experimental setup where freshly nucleated SOA particles are at risk of changing phase state or undergoing partial volatilisation before reaching the ice nucleation measurement?

Prior to the PCU, aggressive drying is implemented (see Figure 1) and temperature is essentially constant ~ 20 °C. Based on the parameterized glass transition temperatures and measurements of at most, 5% RH, it would not be possible for the aerosol to become glassy before measurement. Between the PCU and SPIN, the connecting tubing is the shortest possible length, insulated, and exposed to coolant to prevent a potential glassy to liquid transition. With respect to volatilization, particles were continuously being generated and stabilized in the 5L mixing volume. We estimate that with the long duration of the experiments and continuous injection, the system would be close to gas-particle equilibrium and if volatilization was occurring it would be minimal.

L247. It would be more physically accurate to state that the laser pulse ablates/vaporises the particle and subsequently ionises the resulting fragments, rather than “ionises the particle” directly.

Corrected as suggested.

L253. Please clarify the complementary roles of PALMS-NG and AMS in this study. Both provide chemical composition information. What is the added value of using both systems, and what can each measure that the other cannot?

Fragmentation patterns vary between PALMS-NG and AMS (quasi-bulk). As PALMS has been frequently used in upper tropospheric measurements, markers for organosulfates and/or organic fragments were cataloged to assist in identification in past/future aircraft measurements. We have specified that the data product of AMS used in this

study was predominantly the bulk O:C ratio used for hygroscopicity estimates and the neutralization ratio of the seed particles.

L282. Please explicitly describe the mechanistic expectation for what happens to particles during pre-cooling at −30°C. What change in phase state or diffusivity is anticipated, and how does this translate into the observed differences in ice nucleation onset?

We have re-organized Sect. 2.6.1 to more explicitly state what is inferred to occur during pre-cooling at −30°C and expected effects on ice nucleating behavior. We did not observe the expected changes in ice nucleation onsets which have now been restructured/restated in Sect. 5.2.1

L300. Were the particles injected into SPIN polydisperse? In seeded experiments where both coated seed particles and self-nucleated SOA are present, how were the ice-activating particles identified and distinguished from one another?

Particles injected into SPIN were monodisperse with respect to coated ammonium particles as they were size selected upstream (Figure 1). However, given the presence of self-nucleated particles, the measured particle size distribution demonstrated that the sampled particle population was highly multi-modal. We implemented a multimodal curve fitting algorithm to decompose the complex multi-component aerosol distributions to provide insight on what fraction of certain particle modes entered SPIN (Table 3), their relative surface area contributions (Figure S2), and coating thicknesses (Figure S3).

L304. Please clarify the difference between the machine learning classification model used in this study and the one described by Garimella et al. (2016).

Primary differences are provided in detail by (Rapp et al., 2025). A brief explanation describing differences has been added to the text in the same paragraph after the line specified.

L315. The statement attributed to Koop (2004) regarding temperature inhomogeneities causing earlier-than-expected homogeneous freezing cannot be located in that paper as cited. Please clarify exactly where in Koop (2004) this statement is reflected.

That was unintentionally attributed as written and included the wrong reference (should be Koop et al., 2000). The section now reads:

"In contrast to lamina spreading, inhomogeneities in temperature control have been attributed to homogeneous freezing earlier than expected. As SPIN uses refrigerant valves to control cooling, these locations are colder than neighboring regions of the plate, causing localized pockets of S_{ice} exceeding the homogeneous freezing threshold of aqueous solution droplets (Koop et al., 2000)*."*

L316. Are the SPIN calibration measurements described here part of a published study or available elsewhere?

We have included pre-post onsets of ammonium nitrate in Figure S10.

L324. How sensitive are the reported activation fraction values to the choice of the 100nm threshold for separating self-nucleated from coated seed particles? How would the results change if this threshold were shifted to smaller sizes?

As shown in Figure 2, a 100 nm threshold was not always sufficient in separating coated seed particles from self-nucleated. The primary motivation for the 100 nm threshold was prior work with polydisperse aerosol (Rapp et al., 2025) and was defined prior to starting experiments. We have added two additional plots in the supplement, one using a 50 nm threshold (Figure S7) and another using a 75 nm threshold (Figure S7). From these figures α-pinene is most affected by this threshold due to the abundance of self-nucleated particles in this size range without a compensating increase in ice active particles.

L340, Figure 3. It would be informative to add reference lines indicating the deliquescence relative humidity (DRH) of AS and PNS, to provide context for where phase transitions of the seed particles are expected relative to the observed freezing onsets.

Figure 3 has been adjusted such that the DRH of AS has been plotted assuming a 2:1 ratio of NH_4^+ and SO_4^{2-}. Using AMS data, the average NH_4:SO_4 ratio was 1.63. PNS particles were thus defined as comprising of 1.63 moles NH_4^+, 1 mole SO_4^{2-}, and 0.37 moles H^+ to balance charge. Thermodynamic conditions were retrieved using Model II of E-AIM, with humidity varied. The DRH selected as the first humidity where a non-zero aqueous volume was achieved for temperatures spanning -30 to -50°C (Clegg et al., 1998).

L353. Would pre-cooling of pure AS or PNS seed particles (without any SOA coating) have changed their ice nucleation ability? This control experiment would help isolate the effect of pre-cooling on the seed from the effect on the coating.

We are unaware of any effects that pre-cooling would induce on either AS or PNS particles without an organic layer, assuming low humidity conditions. Given this, pre-cooling experiments on pure AS or PNS seeds were not performed. For AS or PNS particles, if sufficient humidity were present the low temperatures of the PCU could elevate the RH above the DRH liquefying the particles. Given the inlet PCU RH readings were 0±5%, it could be argued deliquescence could be induced by the lower temperatures in the PCU increasing the relative humidity; however, in these instances an organic layer was always present.

L355. Please explain mechanistically how AS particles can trigger heterogeneous freezing at S_{ice} values above their deliquescence RH, given that deliquesced AS solution droplets are expected to follow the Koop homogeneous freezing line. How do these results compare with Welti et al. (2020)?

Heterogeneous freezing of ammonium sulfate has been observed within SPIN (Rapp et al., 2025) and documented by others (Abbatt et al., 2006; Shilling et al., 2006). Diffusion modeling of the SPIN chamber for citric acid supports the theory that if the characteristic ice nucleation timescale is shorter than the time for water to liquefy the organic particle, any ice formed is to be inferred as heterogeneous in formation. By extension, this would be expected for well-studied inorganic salts such as AS where the transition from a liquid to crystalline state is well-defined by deliquescence. The detection of ice formation at S_{ice} below the lower uncertainty limit in homogeneous freezing indicates heterogeneous nucleation on crystalline ammonium sulfate.

This is in direct contrast to the conclusions by (Welti et al., 2020) using SPIN where the steep onset in ice formation was argued to be homogeneous. The authors there also indicated that time-dependent effects could be

responsible for the discrepancy. We refer the reviewer to Figure 2 of (Rapp et al., 2025) where a more gradual onset prior to homogeneous freezing was observed for -45°C and -40°C, alongside a freezing onset at -35°C where homogeneous freezing is not possible based on theory (Koop et al., 2000).

L360. Please provide a mechanistic explanation for why heterogeneous freezing was observed at −40°C but not at −45°C. This temperature dependence is counterintuitive given that lower temperatures generally favour glassy phase states and should enhance heterogeneous nucleation efficiency.

We agree with the reviewer that this result is counterintuitive. We have included a discussion on possible mechanisms responsible for this observation in Sect. 5.2.1.

L373. Please clarify how the onset of water uptake was observed experimentally.

Detailed description is provided elsewhere (Rapp et al., 2025) but we have briefly mentioned this is from the expanded description of the ML classification of the SPIN OPC data. Furthermore, the parameters used in preliminary classification are provided in Table S1.

L479, Figure 4. It would strengthen the comparison to literature if data could be presented at a common activation fraction threshold. The authors should consider contacting the original authors of referenced studies to obtain data at matching AF values.

While we agree with the reviewer that such a comparison would be benefitted from a common activation fraction threshold, variations in instrument-specific thresholds or operating conditions greatly complicate such comparisons. In pre-experimental design (2023), the selection of 0.5% as an activation threshold was purposefully selected to match that of the most recent work on the topic (Kasparoglu et al., 2022). A comparison study among common ice nucleating measurements for the same population of organic particles with predetermined activation thresholds and ice classification schemes would be invaluable in interpreting experimental results, however that is beyond the scope of this work.

L515, Figure 6. The lack of overlap with literature data at lower temperatures is a significant limitation. Please discuss whether it was technically possible to scan at temperatures below −45°C with the SPIN instrument used, and whether additional temperature points between −40°C and −45°C could have been explored. The sparse temperature coverage limits the ability to compare and contextualise prior results.

Please see response to major comment above describing the experimental limitations.

L645. Please note that Piedehierro et al. (2021) specifically suggests that the short residence time in SPIN (~10s) is insufficient for particle equilibration with the surrounding RH under some conditions, which can lead to inhibition of homogeneous freezing.

We thank the reviewer for this point and have expanded this point more clearly. We have also discussed this point in the discussion focusing on freezing mechanisms.

L654. This statement incorrectly characterises both Piedehierro et al. (2021) and Ignatius et al. (2016). Neither study pre-cooled particles in the sense used in this manuscript. Ignatius et al. (2016) generated SOA inside the CLOUD chamber at CERN. Please correct and revise this statement accordingly.

We thank the reviewer for identifying this and note we unintentionally mischaracterized with respect to pre-cooling. We have revised accordingly, specifying that our results for α-pinene-derived BSOA more closely resemble those by Piedehierro et al., 2020 due to the use of the PAM-OFR with the same oxidants. Regarding Ignatius et al. (2016), the particles generated in the CLOUD chamber at CERN were produced at lower temperatures prior to sampling in SPIN, achieving a viscous state (Järvinen et al., 2016) which is similar to the motivation behind the PCU. Nevertheless, we have clarified to prevent any unambiguity that each of the studies used pre-cooling.

L660. Please discuss how the different particle sizes used across studies affect the comparison of ice nucleation onset conditions, and whether it is possible to scale or normalise results to account for size effects.

Please see comment above regarding sizes along with similar responses to RC2. Normalization such as INAS was considered for this study; however, a major assumption required for INAS is that each surface site has an independent probability of initiating ice nucleation. Given there are both coated particles and self-nucleated present, with each contributing a different amount of surface area, this assumption is invalid. Even with the capabilities of multimodal to determine mode specific contributions to surface area (Figure S2), we are unable to isolate mode specific n_s as the chemistry varies for self-nucleated vs. coated sulfate.

L664. Please explain the mechanism proposed for pre-activation. What physical or chemical process might particles have undergone during pre-cooling that would result in them being more effective INPs?

We have clarified the proposed mechanism for preactivation in regards to pre-cooling in context with prior literature (Ladino et al., 2014).

L670. Please clarify whether the evaporation section of SPIN was active or intentionally disabled during experiments where water uptake behaviour was being evaluated. If the evaporation section was off, please provide additional information about how δ_{SPIN} during RH ramps was interpreted.

The evaporation segment of SPIN was always active and have clarified this in the methods after discussing the water uptake classification by the OPC. We also want to clarify that a primary metric distinguishing between water uptake and droplet breakthrough is whether the SPIN lamina exceeded water saturation (Table S1). The observations of water uptake are well below water saturation in the chamber; however, a shift in δ_{SPIN} is clearly observed for these aerosol, indicating changes in optical properties.

L675. An alternative mechanistic interpretation should be acknowledged: homogeneous freezing of the outer liquid shell in a core-shell morphology, where the glassy core facilitates water accumulation at the surface but does not itself act as the heterogeneous nucleating substrate.

We thank the reviewer for making this comment and this alternative mechanism has been added to Sect. 5.2.1. This also prompted the inclusion of an additional interpretation where ice nucleation may occur on crystalline AS engulfed in an aqueous organic layer.

Technical Comments

L164. The PNS abbreviation is already defined in the introduction.

Removed definition of PNS abbreviation and refined distinction between ABS and PNS per RC2.

L181. "detailed" → "detail".

Corrected as stated.

L185. Define THC on first use.

Included the definition of this abbreviation as "total hydrocarbons"

L585. "size" → "six".

Corrected as stated.

L216/Figure 2. Define D_{pg} and σ_g explicitly in the figure caption.

Both are now clearly defined prior to the introduction of Figure 2.

L240. Clarify whether BSOA and SOA are used interchangeably and apply terminology consistently throughout the manuscript.

For references to particles generated from the biogenic precursors in this work, we have adopted the terminology of BSOA. If referencing literature that includes anthropogenic precursors, we have opted to use SOA as it would mischaracterize the cited studies as using only BSOA. Other studies have used the terminology of secondary organic material or matter. In these cases, we have used the common notation of SOA as a general definition of aerosol containing oxidized organic compounds existing in the particle phase.

L269. Define $T_{g,org}$ explicitly on first use, including the meaning of the "org" subscript.

This has now been properly defined when T_g is first introduced in context with these measurements. Furthermore, the manuscript has replaced all instances of T_g to reflect that the quantity being referred to throughout the text was the dry bulk-organic glass transition temperature $T_{g,org}$ unless citing literature where this distinction was not made.

RC2 Review of "Ice nucleating properties of α-pinene- and limonene-derived secondary organic aerosol under cirrus conditions" by Rapp et al., 2026

Anonymous Referee #2, 17 Jun 2026
https://doi.org/10.5194/egusphere-2026-2458-RC2

RC2: Rapp et al. present laboratory measurements of the ice nucleation (IN) ability at cirrus conditions of aerosol particles composed of self-nucleated α-pinene (AP) and limonene (LIM) secondary organic aerosol (SOA), and of ammonium sulphate (AS) or partially neutralized sulphate (PNS) particles coated with AP or

LIM. The experiments probe the role of the precursors' physicochemical properties, their estimated glass transition temperature (Tg), multiphase chemistry, and precooling.

The manuscript is well organized and clear to follow. The research hypotheses are well described and addressed. IN of SOA remains a genuinely difficult topic where instrument accuracy and reproducibility are compounded by complex, hard-to-constrain aerosol systems, and this study provides novel measurements that improve our understanding of the IN ability of SOA.

I recommend publication in ACP after the authors address the comments below.

We thank the reviewer for their thorough review of our manuscript and thoughtful comments. We have addressed the comments detailed and revised the text accordingly.

General comments and questions:

1. In Section 2.6.3 you classify a freezing onset as heterogeneous when it falls below the lower bound of the homogeneous freezing region. However, at several points you note that even when the onset lies in the heterogenous regime, its uncertainty extends well into the homogeneous conditions (e.g. AP-PNS at -40°C). This makes it ambiguous whether such particles should be regarded as heterogeneous nuclei, poor heterogeneous nuclei, or effectively homogeneous. Is there additional evidence that could help resolve the freezing mode for points near the homogeneous threshold? In particular, the shape of the activation fraction spectrum may be informative: heterogeneous freezing tends to produce a gradual increase in AF with S_{ice}, whereas homogeneous freezing tends to produce a sharp, threshold-like rise. The AF spectra in Figures S3 and S4 could be examined on this basis.

   We thank the reviewer for raising this important point as these categories should be defined explicitly. We unfortunately did not find shape of the activation fraction curves to be helpful in distinguishing between heterogeneous and homogeneous onsets in these experiments. A primary factor complicating this for α-pinene-derived BSOA was the gradual growth of a water uptake mode prior to subsequent ice formation beyond the homogeneous freezing threshold of 100 nm aqueous solution droplets. Furthermore, gradual growth was observed for limonene-derived BSOA onsets that were clearly beyond the homogeneous freezing regime. As such, we would prefer to keep the qualitative definition as follows:

   "Additionally, we classify a freezing onset as heterogeneous by the onset being below the lower uncertainty range in water activity (Δa_w = -2.5%) for homogeneous freezing of 100 nm aqueous droplets (Koop, 2004; Koop et al., 2000). Onsets which have error in S_{ice} with significant overlap are considered at best poor heterogeneous nuclei."

2. The methods used to estimate the glass transition temperature (Tg) need a fuller description even with references to prior work. Specifically, I think it is important to show the relations between Tg and C* and Tg and molecular composition. How do errors in C* and bulk composition propagate into the Tg estimates? The uncertainties associated with the thermogram-derived Tg (Table 2) likely don't reflect the variability of the parameterization and therefore underestimate the true error.

   Are there multiple Tg parameterizations in the literature? Do they agree? What motivations brought

you to use the two representations presented in the paper and not for example one based on the functional groups?

We have revised Sect 2.5 to address these points and included a detailed description in Section S1.3 of the supplement. With regards to parameterization selection, we utilized two methods: a thermogram-based volatility $T_{g,org}$ estimate (Li et al., 2020; Niu et al., 2025; Zhang et al., 2019) and a molecular-composition based $T_{g,org}$ allowing the inclusion of CHOS compounds (Li et al., 2020) which is an updated form of (DeRieux et al., 2018). We are unaware of a $T_{g,org}$ parameterization that could incorporate chemical data from Vocus-CIMS, AMS, or PALMS-NG to estimate functional group densities. A molecular dynamics simulation utilizes a functional group method (Siachouli et al., 2025); however, we cannot quantitively identify the number specific functional groups.

The estimated Tg for the pure and SOA-PNS systems are very different (Table 2), but the difference is considerably more pronounced in the bulk composition-derived parameterized Tg than for the thermogram-derived one. The bulk composition is very constant across the different systems, what then explains the difference?

We agree that the similar bulk O:C and H:C ratios do not, by themselves, explain the substantially differences between the two parameterized $T_{g,org}$. Bulk elemental ratios represent only the average oxidation state and do not uniquely constrain molecular composition, functional groups, or volatility distributions. Consequently, aerosols with similar bulk O:C and H:C ratios may still exhibit different thermal-desorption behavior and apparent volatility distributions, leading to different thermogram-derived C* and $T_{g,org}$. The larger difference obtained from the molecular-formula-based parameterization may additionally be influenced by compound-dependent Vocus-CIMS sensitivity. In our derivation, normalized ion signals were used as proxies for component abundances under the assumption of equal sensitivity among detected compounds. If the PNS and self-nucleated systems contain different molecular classes, differences in ionization efficiency or molecular coverage could alter the signal-weighted molecular-formula distribution and therefore the calculated $T_{g,org}$ difference. We have added these as potential explanations and now treat the molecular-formula-derived values as sensitivity estimates.

For AP-PNS experiments, you mention that ice nucleation onset was preceded by an onset of water uptake. How can you unambiguously assign the morphology to a core-shell type and not to a homogeneously mixed particle? This leads to two additional questions: how can you assess that the PNS seeds had crystallized after generation? If not, shouldn't you consider a glass transition temperature for the mixture?

We cannot unambiguously assign the morphology to a core-shell type particle or confirm the phase state of the PNS seeds after passing through the PAM-OFR. However, AMS measurements of NH_4^+:SO_4^{2-} indicate some degree of neutralization had occurred, with more ammoniated character expected with time. Using E-AIM, a particle with a NH_4^+:SO_4^{2-} of 1.63 entering the PAM-OFR after exiting multiple dryers would not be thermodynamically stable as a droplet unless it experienced humidities exceeding 68%. This supports that some crystalline character is almost guaranteed, however we do not have continuous AMS measurements for each experiment.

Regarding the second point, the molecular $T_{g,org}$ parameterizations used here do not support the inclusion of inorganic sulfate. Vocus-CIMS measurements allow for the inclusion of CHOS compounds in the described parameterization that would be present if any reaction formed. To our knowledge, there is no validated parameterization of $T_{g,org}$ that explicitly allows for mixtures of inorganic and organic sulfate.

Across different parts in the manuscript I'd soften the assertion that Tg (generally) fails to predict ice nucleation. I suggest instead saying that the chosen parameterizations applied to your data do not show it (whether this generalizes to all systems and all Tg parameterizations it is not shown in the manuscript).

We appreciate this comment from the reviewer and softened the assertion that T_g more generally fails to predict ice nucleation. We are indeed constrained by the selected T_g parameterizations and precursors studied.

3. Referring to the seeds as ammonium bisulphate (ABS) in some parts of the manuscript is confusing, since by your own account only the first experiment with fresh ABS can be considered "true" ABS. For example, the coatings described in the abstract (L15) were not on ABS. Because you have AMS data, I would suggest labelling all the non-AS seeds as PNS consistently throughout and stating the neutralization degree explicitly for each experiment, including the AS, fresh ABS, and PNS cases.

We have removed all mentions of ABS unless it is in direct context with discussions on aerosol generation or the observation of purely homogeneous freezing as expected.

We note that we do not have AMS measurements for each experiment as these were conducted after the ice nucleation experiments as stated in L160 – 162 (now L225 – 227). Similarly, the VIA-Vocus-CIMS measurements were conducted during that timeframe. We have explicitly stated the timelapse between ice nucleation measurements and these physicochemical measurements similarly to the comment addressing PALMS-NG. This is also clarified when discussing the aging of the ABS to PNS in the materials part of the methods.

4. Changes in aerosol concentration and temperature can shift the gas-particle partitioning of the organic components. Can you rule out changes in chemical composition between the inlets of the different instruments, or during the precooling phase of the IN measurements? Did you try measuring the aerosol composition before and after the PCU?

We cannot rule out changes in chemical composition between inlets of the different instruments or during the pre-cooler as we did not do chemical measurements at different sections of the setup. However, multiple experimental design constraints were in place to minimize such changes. Tubing length between inlets were cut to their minimum lengths and temperature maintained nearly constant, except for the PCU. If chemical reactions were to occur in the PCU we would expect slow reaction rates given the temperature.

5. Can PALMS be used to say something about the distribution of the coating across the seed population?

PALMS-NG is unable to quantify the coating on a single particle; however, across the measured seed population we can quantify the number of spectra without any common organic ion markers. We have added the following sentence:

The number of particles without an organic coating were quantified by evaluating the intensities of common organic ion markers (see Section S2.1.1 of the SI). The fraction of uncoated particles observed ranged from 0.03% to at most 1.09%.

6. Because you compare single-component and mixed particles with different size distributions, why not adopt an INAS-density framework for the heterogeneously freezing systems, which would normalize for surface area?

Normalization such as INAS was considered for this study; however, major assumptions such as independent probabilities of initiating ice nucleation per activation site are invalid for our dataset. With the multimodal algorithm we can decompose the size distributions to retrieve modal areas (Figure S2); however, we are unable to isolate mode specific n_s due to the mixed efficiencies of our particles as shown by the self-nucleated experiments.

Please also clarify the D > 100 nm cut used in the AF denominator: how sensitive is the AF of the LIM experiments to this threshold? Given the low 0.5% AF threshold, could the small but efficient self-nucleated limonene fraction alone exceed it and account for the onset attributed to the coated seeds in the mixed experiments?

We replicated the analysis for 50 and 75 nm cut offs, with respective figures included in the supplement. Based on these figures, restricting to smaller sizes significantly effects the activation fractions of the self-nucleated limonene particles but the coated population is largely unchanged for limonene. This is supported by the observation that self-nucleated limonene rarely exceeded 100 nm when seed particles were present (Table 3). Self-nucleated particles however can grow larger and participate in ice nucleation (Figure 2c).

7. Given the high variability across IN counters, more detail on the measurement strategy is needed. Please report (i) how often filtered, particle-free air was sampled during each measurement; (ii) the background counts for each measurement; (iii) the IN activity measured on the descending S_{ice} branch (if the descending ramp returns to ice saturation, L303 should read “ice saturation” rather than “water saturation”); (iv) how often the chamber walls were re-iced or the ice layer refreshed; and (v) example ramps and ice onsets for the ammonium nitrate calibration experiments.

These points have been addressed in the text and/or supplement.

(i) Filtered, particle-free air was sampled before and after each S_{ice} ramp for 5 minutes. The background counts were interpolated across the entire dataset. The correction factor of 1.4 was also applied to the background ice.

(ii) Background counts used in correcting ice counts have been totaled for each experiment and listed in Table S5 along with relative ratios.

(iii) We specified that the IN activity on the ascending and descending branches were averaged.

(iv) Chamber walls were not re-iced or refreshed as the longest time at which SPIN operated continuously was ~ 10 hours. Background frost levels so increase with time, however they are accounted for in the background subtraction.

(v) We have also included example measurement ramps of the primary experiments (Fig. S9) and ice onsets for pre/post ammonium nitrate calibration (Fig. S10)

8. Section 4 (Experimental uncertainties) reads as a qualitative description of methodological limitations, much of which would be better integrated as concise uncertainty statements within the relevant sections.

We kindly disagree with the reviewer as we would like to summarize methodological limitations in a clear section. We prefer to leave the section as is. Furthermore, we have also included a short paragraph explaining the uncertainty in phase state as that was a key point raised during review.

Specific comments

L113: Please add Bertozzi et al. (2021) on organic coatings of seed particles (relevant also to Figure 5).

Bertozzi B, Wagner R, Song J, Höhler K, Pfeifer J, Saathoff H, et al. Ice nucleation ability of ammonium sulfate aerosol particles internally mixed with secondary organics. Atmospheric Chem Phys. 2021 Jul 16;21(13):10779–98. doi:10.5194/acp-21-10779-2021

We thank the reviewer for including this reference. The data from Figure 5 of (Bertozzi et al., 2021) has also been added to Figure 5 of this manuscript.

L136: This appears to be the only sentence describing how coating thickness was evaluated; it needs more description in the methods/aerosol-generation section. Did you compare the coating thickness for the different modes? Did you compare to the coating estimated from the AMS data assuming a core-shell morphology?

We thank the reviewer for this comment and have added a complete description of coating thickness evaluation in Section 2.3. We have since included coating thickness evaluation for all modes using the multimodal analysis where it is summarized per experiment for singlets in Table 3 and in Figure S3.

Previously, coating thickness was only evaluated for the singlet mode. This was performed by analyzing the diameter in the SEMS data corresponding to the maximum concentration ($dN/dlogD_p$). To exclude any influence from self-nucleated modes or multi-charged singlets, we only looked at particles larger than 200 nm. The difference in maximum diameter between the SEMS scans of the seed particles and those with active BSOA formation was used to calculate coating thickness.

In the revised manuscript, coating thickness is evaluated by subtracting the initial particle diameters for each mode (excluding self-nucleated) as detailed in Section S2. An example of the modal assignment used for an experiment is included in the SI (Figure S11).

While coating estimates using AMS have been used assuming a spherical core-shell morphology (Bertozzi et al., 2021), we argue that SEMS measurements coupled with multimodal analysis provide a more accurate, real-time analysis of the particle distribution entering SPIN.

L220: From the size-distribution plots it looks like there is an additional neutralizer in front of the SEMS. If this is correct, please mention it in the description.

This is correct and has been specified in the text accordingly (L225-226)

L243: Please define "asynchronously" specifying the time elapsed between the PALMS and IN measurements.

PALMS-NG was being prepared to be shipped to Europe at the beginning of these experiments. PALMS-NG measurements were performed during the testing/refinement of stable particle generation. Generating conditions were then replicated with SPIN measurements; however, PALMS-NG was no longer available for simultaneous sampling.

Similarly, AMS and VIA-Vocus-CIMS were not simultaneously measuring during the SPIN measurements. This has been now explicitly mentioned in Sect. 2.4.2 and 2.4.3 following the same style of description (time elapsed).

L271: Citation format error.

Citations were re-indexed throughout the document with the Zotero citation management software and checked for errors.

L357: 13% of particles are doublets and your ice onset is at <1%. On what basis can you exclude that these activated particles originate from the 13% doublets, i.e., that the freezing is driven by the larger seeds?

We have included a surface area comparison of our measurements in the supplement (Figure S2) which demonstrates that the singlet mode dominates surface area in each experiment. In developing this plot, we have also improved the identification of doublet modes and their number concentrations, increasing the singlet/doublet ratios to 90- 93% across experiments. Given the above, we are confident that the singlet mode is predominantly responsible for the activated ice.

L374: How can you establish core–shell morphology rather than a homogeneously mixed particle?

Primarily due to the phase state of the ammoniated sulfate core. RH was maintained below the efflorescence point of AS prior to organic coating. Furthermore, thermodynamic testing using E-AIM as referenced in this response and the response to RC1 do not suggest a liquid core for the PNS particles.

L380: You state that no clear delineation between pre-cooling conditions was observed for the self-nucleated and LIM-PNS experiments. However, the LIM-PNS points at −45°C (Fig. 3) appear to sit very close to the homogeneous freezing region (comparable to the AP-AS experiments you classify as homogeneous) raising the same classification ambiguity discussed above. They also appear to show pre-cooling shifting the onset to higher S_{ice}, opposite to the trend in all other systems and to the summary at L734. Can you provide a suggested explanation?

We have clarified the statement and agree with the reviewer that the PNS limonene point at -45 is more like the a-pinene-AS experiments which were classified as homogeneous. The comment on L734 is attributed to the fact

statistical testing on the dataset using pre-cooling showed no effect. The only instance for limonene in which the expected response of pre-cooling allows ice onsets at lower S_{ice} was for AS particles.

We have included a discussion regarding the counterintuitive nature of pre-cooling shifting ice onsets upwards in Sect. 5.2.

L390: Why did you restrict the PALMS-NG analysis to the singlet particles and exclude the larger ones that could also contribute to the IN signal?

Averaged PALMS-NG spectra shown in Figure S1 used singlets per standard data processing. Given the multimodal PSD analysis, singlets dominate total surface area (Figure S5). If doublets contributed a larger surface area fraction we would reconsider and add these into the averaged spectra but with the given results this is not warranted.

L448 / L689: You report p = 0.28 at L448 and p = 0.59 at L689 for the seed-and-precursor combination, please reconcile.

This was a typo. Given the changes detailed in the multimodal output where a higher resolution time series was used for CPC counts, some test statistics throughout the article have been updated to the most current values, however no p-values have changed above or below pre-defined significance thresholds. Furthermore, no onsets have changed to modify the conclusions of the work.

L465: Do you have AMS data to confirm the degree of neutralization?

We used the AMS to confirm the degree of neutralization of seed particles as summarized in Table 2.

L605: As written, this reads as though you measured Tg directly, please clarify.

We have removed the instance of measured T_g as this incorrectly attributes the methods applied.

L647: If LIM shows no water uptake but its glass transition is ~−70°C, what is the phase state of the particles?

We have corrected the statement that no water uptake occurred. It is more accurate to state that in comparison to a-pinene it was minimal. Regarding the phase state of the particles, we are uncertain and this should be the subject of future studies, particularly direct viscosity measurements as discussed by RC1.

L729: Please reframe specifying which Tg parameterizations fail to predict IN behaviour.

We have clarified that the volatility and molecular composition-based parameterizations failed to predict IN behavior.

L742: Please clarify why the simultaneous presence of self-nucleated particles affects the multiphase-chemistry result specifically.

We have clarified this statement in the conclusion to emphasize that the simultaneous presence of ice active seeds prevents us from solely attributing ice nucleating properties to the coated seed particles.

"Overall, heterogeneous onsets were observed more frequently for PNS seeds than for AS seeds; however, the simultaneous presence of ice active self-nucleated particles prevented us from conclusively attributing the observed ice nucleating properties to the BSOA-coated seed particles. Furthermore, uncertainties in onset S_{ice} and restricted reaction time preclude a definitive conclusion."

Figures

Figure 2: An example of size distributions from a LIM self-nucleated and LIM-coating experiment would be helpful.

This examples have been added and combined with the existing plots.

Figures 4 and 5: Figures 4–6 are cited out of numerical order and are minimally described in the main text. Please cite them in sequence and describe their content/significance in the context.

We have described these figures in more detail within the text and cited in numerical order.

Figure S1: What does "heterogeneous AF" mean? Does it cover AF for S_{ice} from 1.0 to 1.6? Are the AF values already background-subtracted? If so, how do you explain AF > 0 at S_{ice} = 1.0? Does AF > 1 indicate a miscounted ice background?

We have clarified this figure by expanding the caption accordingly. Heterogeneous AF were defined as activation fractions below the lower uncertainty limit in homogeneous freezing. Also, this is represented as % and was not clearly represented in the label. For the last several points if this is regarding Figures S3 and S4 of the original supplement, all data is background subtracted however the SPIN was set to sample at S_{ice} = 1.0 until the PCU concentrations stabilized. In this timeframe, any uncertainty in background subtraction becomes compounded. However as shown in the now included Table S5, the number of backgrounds identified is very small in comparison to the total number of ice observed in each experiment.

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