Author Response

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
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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.

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