

## Response to Reviewer #2

We sincerely thank Reviewer #2 for the constructive and insightful feedback. We appreciate the recognition of the conceptual advance provided by the GCCN-ISP mechanism, and we address each suggestion below (black text: reviewer's comments, blue text: our response, orange text: manuscript revisions):

### 1. Generalizability beyond the AFLUX campaign

Comment: The reliance on a single case study (AFLUX campaign) limits the generalizability of GCCN-ISP. Expanding validation to diverse geographic regions (e.g., mid-latitude mixed-phase clouds) and seasons would strengthen the conclusions. Direct measurements of CCN size/composition (e.g., via aerosol mass spectrometers), if available, can be used to corroborate inferred CCN properties.

Response: We agree that broader validation across seasons and regions is essential. The AFLUX campaign was selected for its detailed in situ measurements and well-characterized Arctic mixed-phase clouds, which provide a valuable starting point. We added other campaigns with similar measurements of cloud properties and possible future applications in the revised manuscript:

Manuscript revision (Line 376):

"Beyond the AFLUX campaign, several other aircraft-based field studies have investigated cloud–aerosol interactions and the microphysical properties of mixed-phase clouds across a range of geographic regions. For example, the ACTIVATE (Aerosol Cloud Meteorology Interactions over the Western Atlantic Experiment) campaign, conducted by NASA over the U.S. East Coast and western North Atlantic, has provided high-resolution measurements of CCN size distribution (0.1 - 2.6  $\mu\text{m}$ ), aerosol composition, and cloud microphysics using dual-aircraft observations (Sorooshian et al., 2025). Similarly, the NAAMES (North Atlantic Aerosols and Marine Ecosystems Study) project included multiple aircraft missions over the North Atlantic to characterize CCN variability (0.02 - 0.5  $\mu\text{m}$ ) and its link to seasonal biogenic emissions (Behrenfeld et al., 2019). Additionally, direct measurements of CCN size and composition (e.g., using aerosol mass spectrometers), when available, offer important means to corroborate inferred CCN properties. Together, these observational efforts highlight the broader relevance of large-particle activation mechanisms and offer valuable datasets for testing the applicability of the GCCN-ISP process under diverse regional and seasonal conditions."

### 2. Simplified aerosol composition ( $\kappa = 1.4$ for NaCl)

Comment: The assumption of pure NaCl CCN ( $\kappa = 1.4$ ) oversimplifies real-world aerosol diversity. Incorporating mixed CCN types (e.g., organics, sulfates) and non-spherical shapes would enhance realism.

Response: Thank you for raising this important point. Indeed, the hygroscopicity parameter  $\kappa$  can vary depending on aerosol composition, and assuming a constant value of  $\kappa = 1.4$  (pure NaCl) does not reflect the full diversity of atmospheric particles. However, this simplification is made for two reasons.

First, as discussed in the manuscript, large CCN particles (with dry diameters  $>1 \mu\text{m}$ ) in the Arctic are most likely dominated by sea salt, especially during winter and early spring. Therefore, we used NaCl as a physically reasonable assumption to construct the look-up table for saturation vapor pressure reduction.

Second, to examine the sensitivity of the process to composition, we also present a comparison using sulfate ( $\kappa \sim 0.8$ ) in Figure 2b. This allows us to show that while the magnitude of the vapor pressure reduction differs slightly, the overall structure and trends in the equilibrium behavior remain consistent between NaCl and sulfate.

We agree that if the internal composition of CCN within individual droplets could be measured, then a more accurate  $\kappa$ -based correction could be applied. This would improve the quantitative accuracy of the vapor equilibrium calculations. We recommend future work to incorporate a realistic distribution of CCN hygroscopicities and shapes to better represent ambient conditions.

### 3. Dynamic processes not considered (e.g., turbulence, entrainment)

Comment: The model neglects dynamic processes like turbulence and entrainment, which could modulate GCCN-ISP efficiency.

Response: We acknowledge that our current work does not explicitly include dynamic processes such as turbulence, entrainment, or localized updrafts, all of which are known to affect cloud microphysical evolution.

In particular, dynamic processes (e.g. vertical motions) influence the ambient water vapor pressure through adiabatic cooling, thereby increasing supersaturation and delaying the depletion of supercooled droplets. This effect, which modulates the efficiency of the Wegener–Bergeron–Findeisen (WBF) process, has been extensively documented in previous studies (e.g., Abade et al., 2024; Gutierrez et al., 2023; Pinsky and Korolev, 2021). Turbulent mixing and entrainment further shape the spatial heterogeneity of supersaturation fields, impacting both the activation and the competition between cloud droplets and ice particles.

In contrast, the focus of our study is to isolate the solute effect—an internal microphysical mechanism that reduces the equilibrium saturation vapor pressure over supercooled droplets. This mechanism enables liquid droplets to persist even in conditions where the environment would otherwise favor ice growth.

To clearly quantify the standalone impact of the solute effect, we intentionally omit dynamic forcing such as vertical motion or turbulence. Nevertheless, we agree that future modeling work, particularly with cloud parcel models or large eddy simulations (LES), should incorporate dynamic processes to evaluate the full environmental modulation of the GCCN-ISP mechanism.

#### Manuscript revision (Line 129):

“As mentioned earlier, in this equation, there is no new water supplied by turbulent mixing, meaning that updrafts are not considered. In atmospheric clouds, the evolution of cloud droplet size is governed by two key factors: (1) the ambient water vapor pressure, which determines the vapor availability for condensation or deposition, and (2) the equilibrium saturation vapor pressure over the droplet surface, which is influenced by droplet size, curvature, and composition (via the solute effect). Updrafts affect cloud microphysics by cooling air parcels and enhancing supersaturation, thereby suppressing the WBF process and delaying ice growth (G. Abade et al., 2024; M. Gutierrez et al., 2023; M. Pinsky and A. Korolev., 2021). In contrast, the solute effect reduces the surface saturation vapor pressure, directly influencing whether supercooled droplets can persist. While both mechanisms can prolong the lifetime of mixed-phase clouds, this study intentionally excludes vertical motion to isolate the microphysical contribution of the solute effect to droplet–ice vapor competition.”

### 4. Broader climatic implications not explored

Comment: While the mechanism’s microphysical basis is plausible, its broader climatic impacts (e.g., on cloud radiative forcing or Arctic amplification) remain unquantified.

Response: We agree that linking the GCCN-ISP mechanism to cloud radiative effects is an important next step. Our current study focuses on the microphysical conditions that can extend droplet lifetimes, which could potentially impact cloud longevity and optical thickness. Although this work does not explicitly quantify radiative forcing, we now note in the revised

manuscript that the suppression of the WBF process by GCCN may increase cloud optical depth and extend lifetime, thereby influencing cloud feedbacks and Arctic amplification. Future research will aim to incorporate this mechanism into cloud and climate models.

Manuscript revision (Line 400):

“Finally, we emphasize the broader climatic relevance of the GCCN-ISP mechanism. The persistence of supercooled liquid water affects the radiative properties and lifetime of mixed-phase clouds. Suppressing the WBF process via solute effects could extend cloud longevity and enhance optical depth, potentially altering cloud radiative forcing and Arctic amplification. Although the present study does not explicitly quantify such impacts, we hope to explore these implications in future work by coupling the GCCN-ISP framework with climate models.”

## 5. Competition/synergy with other ice pathways (e.g., SIP, coalescence)

Comment: The study does not explore how GCCN-ISP competes or synergizes with other ice nucleation pathways (e.g., secondary ice production) or collision-coalescence.

Response: Thank you for this insightful comment. We agree that interactions between GCCN-ISP and other ice-related processes such as secondary ice production (SIP) and collision-coalescence need discussion.

SIP mechanisms—particularly rime splintering—are known to be most active in the  $-3^{\circ}\text{C}$  to  $-8^{\circ}\text{C}$  temperature range (Korolev and Leisner, 2020). However, the cloud layer observed during the AFLUX campaign maintained top temperatures around  $-14.5^{\circ}\text{C}$ , making SIP highly unlikely in this case. Therefore, we did not include SIP in our modeling framework.

As for collision-coalescence, this process predominantly governs droplet growth in warm clouds above  $0^{\circ}\text{C}$  and is generally ineffective in mixed-phase cloud regimes where ice processes dominate and droplet mobility is limited. Given that the cloud in our study was well below freezing and contained both droplets and ice, we consider the omission of warm-cloud collision-coalescence appropriate for the thermodynamic focus of this work.

Manuscript revision (Line 389):

“Secondary ice production (SIP) mechanisms, such as the Hallett–Mossop process, are generally restricted to a narrow temperature range of  $-3^{\circ}\text{C}$  to  $-8^{\circ}\text{C}$  (Korolev and Leisner, 2020). The cloud temperature analyzed in this study is around  $-14.5^{\circ}\text{C}$ , making SIP unlikely. With this consideration, in the current modeling framework, the SIP process is excluded. Extending this work with more comprehensive models that incorporate ice particle shape, ventilation effects, aggregation, and turbulence will be important for assessing the robustness of this mechanism under realistic conditions.”

## 6. Need for laboratory validation

Comment: Controlled laboratory experiments (e.g., cloud chamber studies) are critical to isolate GCCN-ISP under varying temperature, humidity, and CCN conditions.

Response: We strongly agree. Laboratory studies with controlled thermodynamic conditions, variable CCN size/composition, and co-located droplet/ice measurements would provide definitive tests of our theoretical framework. We have added a discussion on how cloud chamber platforms such as AIDA (Aerosol Interaction and Dynamics in the Atmosphere (Lamb et al., 2023)) and the Pi Chamber (Wang et al., 2024) can enable such studies. We particularly encourage future laboratory efforts to target Arctic-relevant conditions ( $T < -10^{\circ}\text{C}$ ) where solute effects may dominate and SIP is suppressed.

Manuscript revision (Line 394):

“Cloud chamber facilities such as AIDA (Aerosol Interaction and Dynamics in the Atmosphere (Lamb et al., 2023)) and the Pi Chamber (Wang et al., 2024) offer controlled environments to systematically vary CCN size, composition, and ambient conditions, allowing detailed observation of droplet–ice competition. While such platforms have been extensively used for

studying ice nucleation and SIP, targeted experiments focusing on solute-induced suppression of glaciation remain scarce. We encourage future laboratory activities that replicate conditions relevant to the Arctic to explore solute-driven persistence in supercooled droplets.”

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