Responses to Reviewer #2

General comments:

There are several parts in the paper where the reader must work through an algebra heavy presentation to extract the underlying physical concept. Adding an interpretation paragraph after major derivations stating why something is the case or what the math means would make the paper more accessible without sacrificing rigor. Some suggestions where physical concepts could be brought forward are included below.

> We appreciate these thoughtful general comments. In the revised manuscript, we have made a systematic effort to improve the physical accessibility of the presentation in addition to addressing the mathematical details. Accordingly, we have added brief interpretive text after several major derivations to clarify the underlying physical meaning of the results, including why the internally mixed case takes a geometric-mean form, why the internal–external contrast can be understood through the AM–GM relationship, and how the different sensitivity regimes arise in terms of the distribution and "waste" of nucleation-triggering events across particles. We have also expanded the explanation of several proof ideas and mathematical steps in the derivations underlying Theorem 1.

To better guide the reader, it should be stated more explicitly in the introduction that the study presents a mathematical framework developed under controlled and idealized assumptions, rather than a direct physical representation of the real atmosphere. The level of idealization, underlying assumptions, and inherent limitations should be clearly articulated and consolidated in the introduction instead of being dispersed throughout the manuscript. In addition, the authors should specify the types of models or data analyses in which the derived analytical expressions could be meaningfully applied.

> We agree that the scope and level of idealization should be stated more explicitly up front. Accordingly, we have revised the introduction to frame the study more clearly as a controlled mathematical and modeling framework developed under idealized assumptions. We now state more explicitly that the analysis is designed to isolate the effect of mixing-state representation while holding other population-level properties fixed, and we clarify the main assumptions and limitations that underlie the theory. We further indicate more clearly where the framework is most useful: as a tool for sensitivity analysis, interpretation of model representation error, and evaluation of how unresolved particle-level mixing state may bias immersion-freezing predictions in models that are constrained mainly by bulk composition rather than full particle-resolved information.

The central finding, that internal mixtures yield higher frozen fractions than external mixtures rests on the critical assumption that the total ice nucleating surface is identical in both cases. The extent to which this assumption can be considered generally applicable should be discussed in greater detail. In the real atmosphere, changes in mixing state are typically accompanied by processes such as coating and coagulation, which alter both the available ice-nucleating surface area and the particle size distribution. These coupled effects may limit the direct transferability of the theoretical result to atmospheric conditions and deserve further consideration

> We agree that the central internal-versus-external comparison rests on the assumption that the total ice-nucleating surface area of each species is held fixed. In the revised manuscript, we clarify that this is a deliberate controlled assumption used to isolate the role of mixing-state representation itself, not a claim that atmospheric transitions between mixing states generally occur without simultaneous changes in size distribution, morphology, or exposed surface area. We now discuss more explicitly that in the real atmosphere, aging, coating, and coagulation commonly modify these properties together, which can limit the direct transferability of the analytical result.

> We also clarify that this is precisely why the present framework should be interpreted as providing bounds and sensitivities under controlled conditions, and why its most direct application is to diagnosing the effect of unresolved mixing-state assumptions in freezing calculations, rather than to representing the full coupled evolution of atmospheric aerosol populations.

Specific comments: We thank the reviewer for taking the time to review our paper and for the constructive comments. The page and line numbers that we quote for indicating where we changed the manuscript refer to the revised marked-up version.

(2.1) Line 10, 81, 450: Clarify in what aspect the simulation goes beyond a numerical realization of the theory which allow to confirm/ verify the theory.

> Thank you for raising this important point. We agree that a model developed based on a given theoretical framework cannot be used to independently verify that theory. Our intention is not to use the simulations as a validation of the theory itself. Instead, the role of the simulations in this study is twofold. First, they serve to confirm that the numerical implementation correctly reproduces the theoretical expectations in idealized cases where analytical predictions are available. Second, and more importantly, they enable us to extend the analysis beyond the scope of the theory. While the theoretical framework provides insight under simplified and often idealized conditions, the particle-resolved model allows us to explore more general aerosol populations, a broader range of mixing states, and scenarios involving additional physical processes that are not analytically tractable. We have clarified this distinction to better emphasize that the simulations are used to demonstrate the applicability and flexibility of the modeling framework, rather than to validate the underlying theory itself.
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> We have made the following changes to the manuscript:
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> - Abstract (Line 10–13): Revised to "Simulations reproduce the analytical trends in limiting cases and extend the analysis to more general aerosol populations, where mixing state continues to exert a substantial control on frozen fraction."
> - Line 119–123: Revised to "This approach provides a precise and flexible representation of different mixing states, allowing us (i) to confirm that the numerical implementation reproduces the analytical predictions in limiting cases where closed-form results are available, and (ii) to extend the analysis to more general aerosol populations, including polydisperse cases, for which an analytical treatment is not feasible."
> - Line 554–556: Revised to "These results are consistent with the main conclusion in Section 3 and further illustrate, in a particle-resolved modeling framework, how mixing state influences the quantity of ice formation."

(2.2) Line 13-14: To illustrate how important it is to consider the mixing state for modelling, it would be helpful to estimate the influence of the mixing state in relation to the variation in the number of particles containing ice-active species on a temperature spectrum of the INP concentration. It could be expected that the variation in particle concentration would outweigh the influence of the mixing state on the INP concentration as a function of temperature.

> Thank you for this insightful suggestion. To further illustrate the potential magnitude of mixing-state effects from the perspective of INP spectra, we have added a new figure (Fig. 6) in section 3. The figure shows the predicted frozen fraction as a function of temperature during constant cooling under controlled conditions, while systematically varying the aerosol mixing state.
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> We have made the following changes to the manuscript:
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> - Section 3: Added Figure 6.

- Line 331–334: Added "The influence of mixing state can also be interpreted in terms of changes in the INP spectrum, i.e., the temperature dependence of freezing efficiency. Figure 6 presents the predicted frozen fraction as a function of temperature for different mixing states under a constant cooling rate (ccr), showing that external and internal mixtures define lower and upper bounds of the INP spectrum for a given bulk surface area composition."

(2.3) Introduction: Summarize the findings of each cited study in one sentence each. It is not helpful for the reader to have 10 different references provided to support one fact without having pointed out what each of them contribute.

We agree that simply listing multiple references to support a general statement can obscure the specific contributions of individual studies.

We have reorganized four sentences in the introduction to better structure the cited literature by grouping studies according to their respective roles (e.g., laboratory evidence, mechanistic insights, and review/synthesis work), and by explicitly summarizing the key findings or contributions of each group. This allows us to retain the breadth of relevant literature while improving clarity on how each cited study informs the overall scientific context.

We have made the following changes to the manuscript:

- Line 20–24: Revised to "Atmospheric aerosols are chemically complex mixtures whose composition reflects both source diversity and chemical aging during transport. Single-particle and chemical imaging studies have shown that individual particles often contain multiple components and exhibit substantial particle-to-particle variability (e.g., Murphy and Thomson, 1997; Laskin et al., 2016, 2019), while laboratory, field, and review studies have documented how atmospheric processing transforms aerosol composition over time (e.g., Rudich, 2003; Hodshire et al., 2019)."
- Line 30–35: Revised to "Aerosol mixing state influences several climate-relevant properties by altering how chemical constituents are distributed across particles. For example, previous studies have shown that mixing state affects cloud condensation nuclei (CCN) activity through its influence on particle hygroscopicity and size-resolved composition (Deng et al., 2013; Ren et al., 2018; Wang et al., 2018; Rejano et al., 2024), and modifies aerosol optical properties through changes in the spatial association of absorbing and scattering material within particles (e.g., Lesins et al., 2002; Liu and Mishchenko, 2018; Zheng et al., 2021)."
- Line 47–50: Revised to "The chemical and physical properties of aerosol species strongly influence their effectiveness as INPs, including surface structure (e.g., Fitzner et al., 2015; Kiselev et al., 2017; Yang et al., 2018), surface chemistry (e.g., Diehl and Mitra, 1998; Zuberi et al., 2002; Knopf et al., 2014; Wang et al., 2016), and hydrophilicity or hydrophobicity (e.g., Fitzner et al., 2015; Bi et al., 2016)."
- Line 51–55: Revised to "Laboratory and modeling studies have shown that heterogeneous ice nucleation efficiency varies substantially across aerosol species, in some cases by several orders of magnitude under similar environmental conditions (Diehl and Mitra, 1998; Murray et al., 2012). This variability has been linked to differences in particle surface properties, nucleation mechanisms, and water-activity-related effects (Knopf and Alpert, 2013; Knopf et al., 2018; Wagner et al., 2021)."

(2.4) Line 69: Instead of putting the singular description out of scope, it can be said that because both CNT and INAS have the same exponential form shown below, Theorem 1 holds for both. In the given function, x changes the distribution of ice nucleating surface area.

$$N_{\mathrm{ice}}(x) = \left[1 - \exp\left(-\frac{c}{x}\right)\right] \cdot x \cdot N_p,$$

For CNT: $c = SJt$, for INAS: $c = Sn_s$. Basically, ϕ in your notation can be replaced by the surface site density n_s.

This is a very insightful suggestion, and it helped us clarify an important conceptual link between our framework and the INAS/singular formulation. Yes, $\Phi = n_\mathrm{S}$ provides the key link that makes our theoretical framework compatible with the INAS/singular formulation.

We have made the following changes to the manuscript:

- Line 85–86: Revised to "However, the compatibility of our theoretical framework with singular/INAS-based formulations is discussed in the Discussion section."
- Line 638–645: Added "In the singular/INAS framework, the quantity Φ in our notation can be interpreted as analogous to the surface site density n_S, which characterizes the ice-nucleating activity of a given species. Specifically, under a given temperature profile $T(\tau)$, we have $\Phi(t) = n_\mathrm{S}\big(\min_{\tau\in[0,t]} T(\tau)\big)$. Although CNT and INAS differ in their underlying physical interpretations, this correspondence implies that Theorem 1 and the sensitivity analysis in Section 3 remain applicable within the INAS framework. For a given species under the same $T(\tau)$, ABIFM/CNT and INAS may assign different numerical values to Φ, but the structural dependence on surface activity is preserved. Accordingly, if Φ_A and Φ_B are interpreted as the corresponding n_S values for species A and B, the sensitivity patterns shown in Fig. 3–5 remain valid, although the species labels in Fig. 3 would differ because those examples are based on ABIFM."

(2.5) Consider discussing the soccer ball model by Niedermeier et al. (2011) and their findings in context of internal or external mixed INP and time dependence.

We have added a brief discussion in the revised manuscript to relate our interpretation to the soccer-ball model of Niedermeier et al. (2011). This addition helps clarify the conceptual connection in terms of particle-to-particle heterogeneity and time-dependent freezing behavior, while also making clear how the focus of our study differs from that framework.

We have made the following changes to the manuscript:

- Line 100–102: Added "In addition, earlier studies have represented heterogeneity in freezing behavior through variability in site-specific nucleation properties within or across particles (e.g., Niedermeier et al., 2011), . . . "
- Line 645–653: Added "This interpretation is conceptually related to the 'soccer-ball' model of Niedermeier et al. (2011, 2014), in which particle surfaces are divided into stochastic surface patches with contact angles drawn from a prescribed distribution. In that framework, small n_site yields stronger particle-to-particle variability in nucleating properties, whereas very large n_site makes particles more similar in their site-property distributions. Their results showed that such surface heterogeneity can make a fundamentally stochastic freezing process appear nearly singular at the population level. Our study differs in that we focus not on heterogeneity in site-specific contact angles, but on heterogeneity in the distribution of chemical species across particles for a fixed bulk composition and size distribution. Both approaches nevertheless point to the same broader conclusion: the distribution of ice-nucleation-relevant properties within and across particles is a key control on the frozen fraction."

(2.6) Line 71-72: support this claim with an example and a citation.

We revised the paragraph to make the knowledge gap more specific and to support it with an example and citations. In particular, we now clarify that previous studies have linked mixing state features to heterogeneous ice nucleation, but have generally not isolated the direct effect of mixing state itself because it co-varied with aging, coating, and associated changes in particle properties.

We have made the following changes to the manuscript:

- Line 97–107: Reorganized the whole paragraph as “While previous studies have suggested that aerosol mixing state can influence ice nucleation, the underlying mechanisms remain poorly constrained. Laboratory and field observations have shown that particle coatings, morphology, and surface composition can all affect heterogeneous ice nucleation by altering active sites and water uptake behavior (e.g., Baustian et al., 2012; Lata et al., 2021), but these effects are often difficult to disentangle. In addition, earlier studies have represented heterogeneity in freezing behavior through variability in site-specific nucleation properties within or across particles (e.g., Niedermeier et al., 2011), yet quantitative investigations that isolate the effect of particle-to-particle compositional mixing state remain scarce. In many cases, changes in mixing state occurred alongside chemical aging or coating processes that also modified particle composition and the surface area contributions of different species, making it difficult to separate the direct effect of mixing state from concurrent changes in particle properties. To our knowledge, no particle-resolved modeling study has systematically quantified the effect of compositional mixing state on freezing behavior while controlling for these confounding factors.”

(2.7) Line 73: Provide an example how a quantitative investigation could look like. Explain why size or mass dependent experiments that determine the frozen fraction as function of exposed surface area wouldn’t provide the needed data. You cite Broadley et al., 2012 on line 172. Are their data not covering the question on the mixing state effect on the frozen fraction?

A quantitative investigation of the type we refer to would require comparing particle populations that have the same bulk composition, size distribution, and total exposed surface area, but differ in how the relevant chemical species are distributed across individual particles. Such an investigation would also require independent particle-resolved characterization of surface or near-surface composition in addition to freezing measurements. Surface-area-dependent frozen-fraction measurements alone are highly valuable, but they would not uniquely constrain compositional mixing state, because populations with the same total exposed surface area may still differ in the particle-to-particle distribution of ice-nucleating material.

We agree that the original wording was too broad, especially in light of studies such as (Broadley et al., 2012), which provide quantitative descriptions of heterogeneity in freezing behavior. Our intended point was narrower: in this study, “mixing state” refers specifically to the distribution of chemical species across particles at fixed bulk composition and size distribution. We have therefore revised the manuscript to clarify that the gap concerns quantitative investigations that isolate the effect of particle-to-particle compositional mixing state, rather than heterogeneity in site-specific nucleation properties more generally. Surface-area-dependent frozen-fraction measurements are highly valuable, but by themselves they would not uniquely constrain compositional mixing state, because populations with the same total exposed surface area may still differ in how the relevant species are distributed across particles.

We have made the following changes to the manuscript:

- Line 100–103: Revised to “In addition, earlier studies have represented heterogeneity in freezing behavior through variability in site-specific nucleation properties within or across particles (e.g., Niedermeier et al., 2011), yet quantitative investigations that isolate the effect of particle-to-particle compositional mixing state remain scarce.”
- Line 105–107: Revised to “To our knowledge, no particle-resolved modeling study has systematically quantified the effect of compositional mixing state on freezing behavior while controlling for these confounding factors.”

(2.8) Line 76: Necessarily aging or coating causes these effects. Provide an atmospheric example in which mixing state changes, but surface area is conserved.

We agree that in the atmosphere, changes in mixing state caused by aging or coating processes are often accompanied by changes in particle size, surface area, or morphology. Our intention, however, is not to represent a specific atmospheric transformation pathway in which particles evolve from external to internal mixtures while conserving surface area.

Instead, this study addresses a different question: how predicted immersion freezing changes when the same aerosol population—with fixed bulk composition and size distribution—is represented with different mixing states. In this controlled setup, we vary only how ice-active and less ice-active material are distributed across particles, while holding other population-level properties fixed. This allows us to isolate the role of mixing-state representation itself in determining freezing behavior.

This question is particularly relevant for immersion-freezing modeling. In most models other than particle-resolved approaches, the input information typically consists of the bulk amounts of different aerosol species, without explicit knowledge of particle-level mixing state. As a result, the representation of mixing state is usually implicit, often assuming either internal or external mixtures. Such assumptions can introduce biases in the predicted frozen fraction when the assumed mixing state differs from the actual aerosol population, or more generally lead to uncertainties associated with unknown mixing states. A key motivation of this work is therefore to provide a systematic framework to quantify the sensitivity of freezing predictions to mixing-state representation.

To clarify this point, we have made the following changes to the manuscript:

- Line 108–114: Added a whole paragraph: “In this study, we do not seek to represent a specific atmospheric transformation pathway, such as coating-driven aging from an external to an internal mixture. Instead, we ask a controlled sensitivity question: for aerosol populations with the same bulk composition and size distribution, how does predicted immersion freezing depend on how constituent species are distributed across particles, from external to internal mixing? This framing isolates the role of mixing-state representation while holding other population-level properties fixed. This question is particularly relevant for ice nucleation modeling, where model inputs typically describe the bulk amounts of aerosol species but do not explicitly resolve particle-level mixing state. As a result, freezing predictions can depend on how this unresolved mixing state is represented.”

(2.9) Line 104: Describe how the surface coverage can be determined from the particle morphology.

Determining the surface coverage of different species on a particle is indeed a complex problem and depends on assumptions about particle morphology. Various approximations can be adopted depending on the level of detail required. For example, particle shape may be approximated as spherical, in which case the surface area can be estimated from the particle diameter (e.g., πD_p^2). Alternatively, more complex geometries such as ellipsoids can be assumed, where the particle surface area can be computed from the lengths of the major and minor axes using standard formulas for spheroidal surfaces.

In addition, the relationship between species volume fraction and surface coverage can vary depending on the particle morphology and internal structure. In more complex scenarios, such as coated particles, the surface coverage of each species must be determined by explicitly resolving the particle structure rather than assuming proportionality between volume fraction and surface area fraction.

In Section 2, our goal is not to provide a comprehensive treatment of particle morphology. Instead, we illustrate the meaning of the statement “the surface area occupied by each species can be determined with assumptions about the particle morphology” using the simplest morphological assumption, which is also the one adopted in the modeling framework of this study: particles are assumed to be spherical and the surface area fraction of each species is proportional to its volume fraction. Under this assumption, the surface coverage can be estimated directly from the relative volumes of the constituent species.

To clarify the limitations of this assumption, we have made the following changes to the manuscript:

- Line 144–148: Added "For example, under a simple morphological assumption where particles are spherical and the surface area fraction of each species is proportional to its volume fraction, the surface coverage can be estimated from the relative volumes of the constituent species, which can be calculated from their masses and densities."
- Line 658–663: Added "A related simplification arises in representing particle's morphology in the particle-resolved simulations. In the current PartMC implementation, species composition is tracked in terms of mass, while particle morphology and surface exposure of individual species are not explicitly resolved. In the absence of such information, we assume that the surface area fraction of each species is proportional to its volume (or mass) fraction within a particle. This assumption provides a practical way to prescribe surface area partitioning among species but may not hold for particles with complex internal structures or coating morphologies."

(2.10) Line 115, 120: Why is Eq. 9 not just a substitution but considered simpler than Eq. 7?

We agree that Eq. (9) is not a mathematical simplification of Eq. (7) but rather a change of notation introduced for convenience. The definition of Φ_i allows the time integral of the nucleation rate to be represented as a species-specific quantity that remains fixed under identical cooling conditions. This notation makes the subsequent theoretical analysis clearer, as each species can be characterized by a single parameter Φ_i.

To avoid confusion, we have made the following changes to the manuscript:

- Line 159: Revised to "…so that Eq. (7) can be ~~simplified~~ written in terms of Φ_i as …"
- Line 164–165: Revised to "Additionally, Eq. (9) provides a ~~simplified form~~ convenient representation of Eq. (7) for the theoretical analysis discussed in the following sections."

(2.11) Line 126, 141-144: Explain why it is stressed that the following analysis is only valid for monodisperse particle populations. Brefly discuss what the expected impact of a polydisperse distribution is, for example, based on Eq. 9. Would it go beyond replacing S_p by the average $\bar{S}_\mathrm{p}$?

We thank the reviewer for raising this important point. We have clarified in the revised manuscript why Theorem 1 is restricted to monodisperse particle populations. Specifically, Theorem 1 compares internal and external mixtures under the assumption that particles share the same surface area. When particle sizes vary, the freezing probability becomes particle-specific because it depends on the product $S_\mathrm{p}\Phi$ (see Eq. (9)). As a result, the comparison between internal and external mixtures is no longer governed solely by how species are distributed across particles, but also by the particle size distribution.

To make this limitation clearer, we have made the following changes to the manuscript:

- Added a new appendix: Appendix E—A counterexample demonstrating the inapplicability of Theorem 1 to polydisperse particle populations.
- Line 186: Added "A counterexample for polydisperse particle populations is discussed in Appendix E."

Regarding the reviewer's question about whether the effect of polydispersity could be approximated by replacing S_p with an average $\bar{S}_p$, we note that such an approximation is only valid in the limit where $S_\mathrm{p}\Phi$ is small for all species (i.e., the Regime 2 shown in Fig. 3). In this regime, Eq. (18) holds and the freezing probability is approximately linear in S_p. Under these conditions, replacing S_p by $\bar{S}_p$ provides a reasonable approximation. Outside this regime, however, the relationship between freezing probability and S_p becomes nonlinear, and therefore the frozen fraction cannot be accurately approximated by using an average surface area.

(2.12) Sec. 3.1. Eq. (12)-(15): The paper presents the algebra but never explains that the geometric mean appears for internal mixtures because a mixed particle remains unfrozen only when no surface component nucleates ice. Given independent nucleation events, the corresponding probabilities of not freezing multiply. Raising each term to the power of its surface fraction accounts for proportional surface coverage.

We agree that the physical reasoning behind the geometric mean form for internal mixtures should be clarified. We have made the following changes to the manuscript:

- Line 211–213: Added "This form arises because a droplet containing internally mixed INPs remains unfrozen only when none of its surface components nucleates ice. Assuming independent nucleation events, the probabilities of remaining unfrozen multiply, and each term is raised to the power of its surface fraction w_i to account for proportional surface coverage."

(2.13) Line 166-167: It is difficult to understand what is meant by "droplets containing multi and single species" in the context of internal mixing. Provide a physical explanation to clarify this relationship.

In the original wording, the phrase "droplets containing multi and single species INPs" may indeed be confusing in the context of internal mixing. Our intention was to express that Eq. (15) relates the frozen fraction of droplets containing internally mixed INPs to the frozen fractions that would arise if the particle surface were entirely composed of a single INP species.

To clarify this, we have made the following changes to the manuscript:

- Line 215–216: Revised to "Thus, this leads us to an expression that relates the frozen fraction of droplets containing internally mixed INPs to a combination of the single-species reference cases, . . . "

(2.14) Line 173-174: The internal vs. external mixture difference is fundamentally a geometric mean vs. arithmetic mean problem. For any set of positive, not all equal numbers the arithmetic mean is always greater than the geometric mean and the difference between them increases as the variability increases. This could be highlighted more. It confirms Theorem 1.

We have made the following changes to the manuscript:

- Line 227–230: Added "Mathematically, this difference corresponds to the inequality between the arithmetic and geometric means: for any set of positive values that are not all equal, the arithmetic mean exceeds the geometric mean. As a result, internal mixtures generally yield higher frozen fractions than external mixtures."

(2.15) Line 175: Relate the statement to the difference in geometric mean vs. arithmetic mean.

We have made the following changes to the manuscript:

- Line 230–232: Added "Moreover, the difference between the two increases as the variability among species increases, meaning that the sensitivity of the frozen fraction to the INP mixing state becomes more pronounced when species have diverse freezing efficiencies (i.e., different $f_\mathrm{unf}^{(i)}$)."

(2.16) Line 207: Explain the situations in which these combinations are relevant.

Our intention was not to imply that each highlighted point in Fig. 3 corresponds to a uniquely identified binary particle type that has been directly observed as such in the atmosphere. Rather, these examples were included to illustrate plausible pairings among classes of ice-nucleating particle (INP) materials that are known to coexist in atmospheric aerosol populations. In this sense, the figure is meant to provide physically meaningful reference points within the theoretical parameter space, rather than a catalog of directly observed binary particle types. Considering that particle compositions vary by region and among laboratory experiments, it is beyond the scope of this study to cover all these different scenarios. Once the particle population is known, analysis such as Fig. 3 can provide the sensitivity map.

To avoid potential confusion, we have made the following changes to the manuscript:

- Line 265–266: Revised to "For reference, some ~~atmospherically relevant~~ examples of species combinations A and B are highlighted as horizontal dashed lines and aligned solid dots."

(2.17) Figure 3: consider repeating the scenario conditions in the caption.

We have made the following changes to the manuscript:

- Caption of Figure 3: Added "The Φ values are calculated for a scenario with a constant temperature of $-33°$C and a duration of 10 minutes using Eq. (8)."

(2.18) Line 212: Define "uncertainty" or for consistency use "sensitivity" instead.

We have made the following changes to the manuscript:

- Line 270: "In Fig. 3, there are three regimes where ~~uncertainties~~ sensitivities are negligible, . . . "

(2.19) Sec. 3.2. Eq. (16)-(19): Taylor expansions are given but the physical picture explaining the different regimes could be expanded: When nucleation rates are very low, freezing is a rare event regardless of species and most particles don't freeze. How the species are distributed doesn't matter. When nucleation rates are very high, most particles freeze regardless of mixing state. The mixing state only becomes important when the good species is efficient enough to cause freezing, but the bad species is not, creating a population where external mixing leaves many particles unfrozen.

We thank the reviewer for this insightful comment and for clearly summarizing the physical interpretation of the different regimes. We agree that the physical picture behind Eqs. (16)–(19) should be explained more explicitly. Following this suggestion, we have expanded the discussion in Sec. 3.2 to clarify the physical mechanisms underlying the different regimes.

We have made the following changes to the manuscript:

- Line 283–289: Added "In this regime, the result also has a simple physical interpretation. Multiplying Eq. (18) by the total number of particles N_{p} shows that the expected number of ice particles is approximately equal to the expected number of ice-nucleation triggering events. When nucleation probabilities are small, each triggering event typically leads to the freezing of a single particle. Since the total surface area of each species is the same for internal and external mixtures, the expected number of triggering events is also the same regardless of the mixing state. As a result, the number of frozen particles is nearly identical for the two mixing states, leading to the small sensitivity observed in Regime 2."

- Line 301–316: Added "From a physical perspective, the differences among these regimes can be understood in terms of how ice-nucleation triggering events are distributed among particles. For a given cooling condition and fixed total surface area of each species, the expected total number of triggering events in the particle population is approximately independent of the mixing state. The mixing state instead determines how these events are distributed among particles. Because only the first triggering event on a particle causes droplet freezing, additional events occurring on the same particle are effectively wasted. Section 3.1 explains the difference between external and internal mixtures precisely from this perspective. In Regime 2, nucleation rates of both species are very small, so the expected number of triggering events per particle is much less than one. Multiple events on the same particle are therefore rare, little waste occurs, and the mixing state has negligible influence on the frozen fraction. In Regime 3, nucleation rates of both species are very large, and each particle surface experiences many triggering events. Even if the nucleation efficiencies of the two species differ by several orders of magnitude, the bad species still produces multiple triggering events on particles composed solely of that species. Consequently, the frozen fraction approaches unity ($\approx 100\%$) for both internal and external mixtures, because the additional events generated by the good species are effectively wasted once a particle has already frozen. Only in the Regime 4 does the mixing state become important. In this case, particles containing the good species generate more than one triggering event on average, while particles composed of the bad species generate fewer than one. Internal mixing distributes these excess events across particles and reduces waste, whereas external mixtures concentrate them on particles that already freeze easily, leading to a larger difference in frozen fraction."

(2.20) Line 232: Is the significant sensitivity what is marked as red 20% line in Figs. 3, 4, 5?

Yes, in Figs. 3–5 the "significant sensitivity" referred to in the text corresponds to the region enclosed by the 20% sensitivity contour, which is shown as the red line in the figures.

To clarify this point, we have made the following changes to the manuscript:

- Line 296–297: Added "(exceeding approximately 20%, as indicated by the red contour in Figs. 3–5)"

(2.21) Line 250: Provide an estimation of how relevant the error in the frozen fraction from the mixing state is for calculating the INP concentration compared to the uncertainty regarding the quantity and nucleation rate of the INP species.

We appreciate the reviewer's suggestion. We agree that the practical relevance of mixing-state-induced differences should be viewed in the broader context of other uncertainties affecting INP predictions. At the same time, we think that a generally applicable quantitative comparison is difficult, because the relative importance of these uncertainties depends strongly on the specific INP species considered, their nucleation parameterizations, and the temperature or cooling regime.

Rather than attempting a universal estimate based on additional assumptions, we now use Fig. 6 to provide a more cautious interpretation. The figure shows that, for representative mixed-species cases, changing only the mixing-state representation while holding bulk composition and particle size fixed can produce substantial differences in the predicted frozen fraction, and therefore in the inferred ice number concentration. This demonstrates that mixing state can be a non-negligible source of uncertainty in immersion-freezing predictions, particularly in the regime where freezing is sensitive to contrasts in species-specific ice-nucleating activity.

We therefore do not attempt to rank mixing-state effects against all other uncertainty sources in a general sense. Instead, our point is that Fig. 6 illustrates that the influence of mixing state can be large enough to warrant consideration alongside other known uncertainties in INP-related calculations.

(2.22) Line 314: According to the previous section there is no insoluble components. Mention in the text why the effect of solutes has not been tested, especially as the ABIFM can specifically consider them.

> We agree that the ABIFM framework, including its implementation in PartMC, can represent immersion freezing in particles containing soluble species. However, the physical mechanisms by which soluble species affect immersion freezing are fundamentally different from those considered in Section 3 for mixtures of insoluble ice-nucleating species only.
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> In particular, when soluble species are present, their influence does not arise through the same surface-area-based mixing-state mechanism analyzed in our theory. They affect immersion freezing through coupled effects on solution thermodynamics and droplet activation, which alter the nucleation behavior in ways not represented by the assumptions of Section 3. Addressing the mixing state between soluble and insoluble species therefore requires a different theoretical treatment and is beyond the scope of the present study.
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> To clarify this scope, we have made the following changes to the manuscript:
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> - Line 335–343: Added "It is important to note that the theoretical analysis presented in this section applies to mixtures of insoluble ice-nucleating species only. When soluble species are present in mixed particles, their influence on immersion freezing arises through different mechanisms. After dissolution, soluble species do not contribute nucleating surface area but instead modify the nucleation efficiency of insoluble species through changes in water activity (Knopf and Alpert, 2013). In addition, soluble species can affect the activation of particles into cloud droplets through their hygroscopic properties (Petters and Kreidenweis, 2007), which is a prerequisite for immersion freezing. These processes introduce additional couplings between particle composition, nucleation efficiency, and droplet activation that are not represented in the present theoretical framework. Investigating the role of mixing state in systems containing both soluble and insoluble species therefore requires an extension of the theory developed here and is left for future work."

(2.23) Line 326: The notation changes from before where P was used for probability and N_p for particles and π as a number (line 217). Consider using the same notation for all parts of the paper.

> In the revised manuscript, we have simplified and unified the notation accordingly. Specifically, we have removed the definition of the full particle set P, as it is not used elsewhere in the manuscript. To maintain consistency and avoid confusion with $\pi = 3.1415\ldots$, we use the uppercase symbol Π as the set of particles that satisfy the immersion freezing conditions. The subset of particles in the i-th bin is then denoted as Π_i. We continue to use the notation $|\cdot|$ to denote the cardinality of a set. Although $|\Pi|$ is conceptually similar to the previously used N_p, this notation provides greater flexibility in expressing bin-resolved quantities and avoids more cumbersome expressions such as $N_\mathrm{p}^{(i)}$. This notation is also consistent with that used in Curtis et al. (2016).
>
> We have made the following changes to the manuscript:
>
> - Line 420–421: Revised to "We define Π as the set of particles that contain an insoluble core and sufficient water to satisfy the conditions necessary for immersion freezing."
> - Section 4.3: All instances of π and π_i have been replaced with Π and Π_i, respectively.

(2.24) Line 327: Explain how the condition of sufficient water is defined and if water uptake on insoluble particles is simulated.

> We have made the following changes to the manuscript:

- Line 422–424: Revised to "In the numerical implementation, this condition is defined as particles whose water mass fraction exceeds 2%, which is used as a practical criterion to identify particles activated to droplets. In the simulations presented in this study, all particles are initialized with sufficient water to satisfy this condition, so the specific threshold does not influence the results."

(2.25) Line 388: Explain the χ index in the text and point to Appendix G.

We have made the following changes to the manuscript:

- Line 484–487: Revised to "Additionally, this section includes results for monodisperse INPs with intermediate mixing states, demonstrating how the frozen fraction varies with the mixing state index χ, which quantifies the degree of internal versus external mixing in the particle population (Riemer and West, 2013); see Appendix G for details."

(2.26) Line 389: Explain why this aspect cannot be analysed using the framework.

We have clarified this point in the revised manuscript:

- Line 488–490: Revised to "This aspect cannot be analytically quantified using the current theoretical framework, which only establishes the bounds corresponding to internal and external mixtures but does not describe how the frozen fraction varies continuously with the mixing state index χ."

(2.27) Line 402: Clarify why it is important to note that the population is not monodisperse.

We agree that it is important to clearly distinguish the assumptions of the theoretical framework from the simulation setup. We therefore explicitly highlight this difference so that readers recognize that the application of the theory to the simulation results is conditional. This clarification also guides the reader to Section 5.3, where the implications of polydispersity for interpreting the theoretical results are discussed in more detail.

We have made the following changes to the manuscript:

- Line 505–507: Added: "It is important to note that the simulations are performed for polydisperse particles, whereas the theoretical framework in Section 3 is derived for monodisperse INP populations with identical particle surface areas. The implications and conditional applicability of the theory in this context are discussed in Section 5.3"

(2.28) Figure 9: At what time of the simulation are the frozen fraction compared?

In Figure 9, each point represents the frozen fraction at one simulation time step, so the comparison includes results from all time steps of the simulation. We have clarified this in both the main text and the figure caption:

- Caption of Fig. 10 (originally Fig. 9): Revised to "Comparison of frozen fraction between analytical calculations and PartMC simulations for the eight cases listed in Table 2 (Cases 1–8). The PartMC results are obtained from an ensemble mean of 20 repeated simulations. Each panel corresponds to one case, and each point represents the frozen fraction at one simulation time step (all time steps are included). The red dashed line indicates the 1:1 relationship between the simulated and theoretical values."

- Line 558–559: Added “Each point represents the frozen fraction at one simulation time step, so results from all time steps are included.”

(2.29) Line 457: Quantify the minimum number of computational particles required to suppress sampling noise for low frozen fractions.

We have added an order-of-magnitude estimate of the minimum number of computational particles required to suppress sampling noise for very low frozen fractions. To keep the main text concise, the derivation is presented in Appendix I, and the main text now briefly summarizes the resulting estimate and its implications for Cases 1 and 2.

Specifically, we have made the following changes to the manuscript:

- Added Appendix I.
- Line 559–567: Revised to “Generally, the model results closely align with the theoretical values, with an RMSE of less than 1% and a correlation coefficient exceeding 0.9999, except for Case 1. The discrepancy in Case 1 arises from the finite number of computational particles in the PartMC simulation. Because the frozen fraction in this case is extremely low, on the order of 10^{-3}%, accurately resolving it with only 10 000 computational particles becomes challenging. An order-of-magnitude estimate of the computational particle number required to suppress sampling noise is provided in Appendix I. That estimate suggests that resolving frozen fractions at the level of Case 1 with a relative uncertainty of about 10% would require on the order of 10^7–10^8 computational particles. The results in this section therefore demonstrate the implementation of the immersion freezing algorithm for multi-species INPs in PartMC, verified against the analytical solution.”

(2.30) Line 481-482: The same argument presented on line 176-181 applies to (b) vs. (c). Explain why nevertheless Theorem 1 is not applicable when comparing the particle population of (c) to (a) instead of bin wise comparison. A plot demonstrating the failure of Theorem 1 would be helpful.

The key reason why Theorem 1 cannot be applied when comparing panel (c) with panel (a) in Fig. 11 is that Theorem 1 is derived for monodisperse particle populations, where particles share the same surface area. We have added a dedicated Appendix (Appendix E) to present a counterexample illustrating this point.

In Fig. 11, panels (a) and (b) satisfy the conditions required by Theorem 1 within each bin: the surface area ratio between species A and B is identical within every size bin, and therefore the internal mixture (a) produces a higher frozen fraction than the corresponding external mixture (b). However, panel (c) represents a different type of external mixture in which the relatively efficient species are preferentially associated with smaller particles while the less efficient species are associated with larger particles.

This configuration is conceptually equivalent to the counterexample presented in Appendix E. When the particle size distribution is sufficiently broad, such a redistribution of species can reduce the variability of particle freezing probabilities across the population, and the frozen fraction of the particle population in panel (c) can exceed that in panel (a), even though both populations have the same total surface area of each species.

(2.31) Appendix D, Eq. D8-D9: Explain that $\mathcal{K}$ is constant because it depends only on the total surface area of each species, not on how that area is distributed among particles.

We have made the following changes to the manuscript:

- Line 915–917: Added "It is a constant because it depends only on the total surface area of each species $S^{(i)}$, which is fixed in Optimization 1 and independent of how this surface area is distributed among particles (i.e., independent of the mixing state). We therefore define this constant as $\mathcal{K}$: ..."

(2.32) Appendix D, Eq. D15–D20: The Lagrange multiplier method is correct but a bit overkill. It could be replaced by the inequality of the arithmetic and geometric mean: For a fixed product, the sum is minimized when all terms are equal.

$$\frac{1}{N_p}\sum_{j=1}^{N_p} P_{\mathrm{unf},j} \geq \left(\prod_{j=1}^{N_p} P_{\mathrm{unf},j}\right)^{1/N_p} = \mathcal{K}^{1/N_p}$$

with equality iff $P_{\mathrm{unf},1} = \cdots = P_{\mathrm{unf},N_p}$. Thus, the minimum possible unfrozen fraction (maximum frozen fraction) requires equal freezing probabilities of all particles.

Thank you for this helpful suggestion. We agree that the result can be derived more directly using the arithmetic–geometric mean (AM–GM) inequality, which provides a simpler and more transparent argument than the Lagrange multiplier formulation.

We have made the following changes to the manuscript:

- Line 930–938: Revised to "Now, we solve optimization 2. Based on the AM–GM relationship, there is

$$\frac{1}{N_\mathrm{p}}\sum_{j=1}^{N_\mathrm{p}} P_{\mathrm{unf},j} \geq \left(\prod_{j=1}^{N_\mathrm{p}} P_{\mathrm{unf},j}\right)^{\frac{1}{N_\mathrm{p}}} = \mathcal{K}^{\frac{1}{N_\mathrm{p}}}, \tag{1}$$

  with equality if and only if

$$P_{\mathrm{unf},1} = P_{\mathrm{unf},2} = \cdots = P_{\mathrm{unf},N_\mathrm{p}} = \mathcal{K}^{\frac{1}{N_\mathrm{p}}}. \tag{2}$$

  Therefore, $1 - \frac{1}{N_\mathrm{p}}\sum_{j=1}^{N_\mathrm{p}} P_{\mathrm{unf},j}$ attains its maximum only when Eq. (2) hoods. Thus, we have

$$P^*_{\mathrm{unf},j} = \mathcal{K}^{\frac{1}{N_\mathrm{p}}} = \exp\left(-\frac{1}{N_\mathrm{p}}\sum_{i=1}^{N_\mathrm{p}} S^{(i)}\Phi_i\right), \text{ for } \forall j \in [1, N_\mathrm{p}]. \tag{3}$$

  Therefore, when each particle has the same unfrozen (or freezing) probability, Eq. (D13) in optimization 2 reaches its maximum, where

$$f^*_{\mathrm{frz}} = 1 - \frac{1}{N_\mathrm{p}}\sum_{j=1}^{N_\mathrm{p}} P^*_{\mathrm{unf},j} = 1 - \mathcal{K}^{\frac{1}{N_\mathrm{p}}} = 1 - \exp\left(-\frac{1}{N_\mathrm{p}}\sum_{i=1}^{N_\mathrm{s}} S^{(i)}\Phi_i\right). \tag{4}$$

  "

(2.33) Appendix D, Eq. D28-D36: Add some interpretation or an example explaining that the index-heavy algebra shows that exchanging equal amounts of inefficient with efficient species between particles preserves each species' total surface area and each particle's total surface area. Making one particle more efficient and the other less efficient increases the sum of their probabilities not to freeze. The new configuration has a higher unfrozen fraction (a lower frozen fraction) than the original. The external mixture therefore must be the least efficient mixing state.

We have added brief interpretation in several places following Eqs. (D28–D36) to clarify the physical meaning of the algebraic steps. Specifically, we now explain that the construction corresponds to exchanging equal surface areas of relatively inefficient and efficient species between two particles, which preserves both the total surface area of each species and the total surface area of each particle. This exchange increases the heterogeneity in particle freezing probabilities, leading to a larger sum of unfrozen probabilities (i.e., a lower frozen fraction). This interpretation clarifies why the external mixture represents the least efficient mixing state.

We have made the following changes to the manuscript:

- Line 959–961: Added "…at least one particle must contain more than one species, such that the surface area ratio of each species within that particle is less than 1. We refer to it as 'mixed particle' thereafter."
- Line 964–967: Added "In other words, there must be at least two mixed particles. If there were only one mixed particle, one of our assumptions would be violated—we could not form a strict external mixture. Since there are at least two such particles, we can select two of them that both contain species i_0, and denote their indices by j_A and j_B, respectively (The above argument shows that such particles j_A and j_B must exist)."
- Line 974–976: Added "This statement holds because i_0 is defined as the species with the smallest Φ among all species present in mixed particles. Since the other species contained in particle j_A also belong to this set, the average Φ of those species, $\bar{\Phi}_{j_A}^{(i \neq i_0)}$ must be greater than Φ_{i_0}."
- Line 981–992: Added "This new mixing state can be understood as a surface-area exchange between particles j_A and j_B relative to the original mixing state. In essence, making one particle less efficient and the other more efficient increases the sum of their probabilities of remaining unfrozen. As a result, the new configuration has a higher unfrozen fraction (and thus a lower frozen fraction) than the original one. The external mixture therefore represents the least efficient mixing state. Specifically, a surface area corresponding to a fraction δw of the particle surface area is exchanged between the two particles. We transfer an area $\pi D_\mathrm{p}^2 \delta w$ of species i_0 from particle j_B to particle j_A, while transferring from particle j_A to particle j_B the same total surface area of the remaining species, distributed according to their original proportions. This exchange leaves the freezing probabilities of all other particles unchanged, while modifying those of particles j_A and j_B. In particular, particle j_A, which may initially have a relatively small freezing probability, now contains a larger fraction of the relatively inefficient species i_0. In contrast, particle j_B, whose freezing probability may initially be larger, now contains a smaller fraction of species i_0. As a result, the difference in freezing probabilities between the two particles increases. We next examine how this increase in heterogeneity affects the overall frozen fraction."

Technical corrections: We thank the reviewer for the detailed technical corrections. All comments have been carefully considered and the corresponding revisions have been implemented throughout the manuscript. Each comment and its associated modification are listed below.

(2.34) Line 76: correct sentence structure.

We have made the following changes to the manuscript:

- Revised to: "To our knowledge, no particle-resolved modeling study has systematically quantified the effect of mixing state on freezing behavior while controlling for these confounding factors."

(2.35) Line 85, and other places: It could be argued that it is not the particle that freezes in immersion freezing but the droplet. Consider specifying "freezing of a droplet" or "ice nucleation on the surface of a particle" where practical.

We have made the following changes to the manuscript:

- Line 125–126: “Following classical nucleation theory, heterogeneous ice nucleation occurs when the first nucleation event on the surface of an ice nucleus triggers the entire ~~particle~~ droplet to freeze.”
- Line 77–79: “The singular approach assumes that each ~~particle~~ droplet freezes at a characteristic temperature, independent of time, while the time-dependent formulation treats freezing as a stochastic process governed by a temperature-dependent nucleation rate.”
- Line 722: “. . . which is also the freezing probability of a droplet containing the particle within the time interval Δt . . . ”

(2.36) Line 119 is a repetition of line 117.

We have made the following changes to the manuscript:

- Previous sentence in Line 161 has been removed: “~~The value of Φ_i describes the freezing ability of the i^{th}~~”

(2.37) Line 119: Clarify why the cooling rate and duration must be controlled. Do you mean they must be “known”?

What we intended to convey is that the cooling rate and the duration of the cooling process must be known in order to accurately quantify the sensitivity. Even for a fixed combination of species (e.g., Fe_2O_3–illite with a 50:50 split of total surface area), different cooling rates and durations lead to different sensitivities.

From a mathematical perspective within the CNT framework, the value of Φ for each species depends on the time integral of the nucleation rate, which in turn depends on the cooling history. Therefore, the cooling rate and the duration must be specified to compute Φ.

As an illustrative example, in the ABIFM model the heterogeneous nucleation rates of Pahokee Peat and illite intersect near -24 °C. If a cooling trajectory is centered around this temperature, the resulting Φ values of the two species may become similar, leading to a sensitivity close to zero. In this case, although the nucleation rates differ above and below -24 °C, the integrated freezing behavior over the cooling period can be effectively equivalent to that of a single species, making the frozen fraction insensitive to mixing state. Under other cooling histories, however, differences in Φ between the species will generally arise, and the sensitivity increases as the contrast between their Φ values becomes larger.

(2.38) Line 135: To include all restrictions mentioned on line 127, “monodisperse INP” could be replaced with “this scenario”.

We have made the following changes to the manuscript:

- Line 179: “Here, we present a theorem regarding the most and least efficient mixing states for ~~monodisperse INPs~~ this scenario.”

(2.39) Line157, Appendix A2, B: As “mode” and “species” seem to refer to the same thing, consider using only “species”.

We agree that the terms “mode” and “species” were used redundantly in Line 157 and Appendix A2, which may cause confusion. We have therefore revised these sections to consistently use “species” and clarified the corresponding descriptions. Therefore, we have made the following changes to the manuscript:

- Line 201–202: “For the externally-mixed case, the following applies. Since each particle contains only one species, the whole INP population can be conceptualized as a collection of ~~multiple modes~~ distinct species ~~, with each mode representing a different species~~.”
- Line 740–742: “An externally mixed monodisperse INP population can be conceptualized as ~~a collection of multiple modes~~ a set of distinct species, each represented by particles composed of a single species. ~~, with each mode representing particles composed of a single species~~.”

However, in Appendix B, we retain the term “mode” as it serves a distinct purpose. Specifically, Appendix B considers a single-mode polydisperse INP population, where a “mode” is defined as a group of particles whose sizes follow a specified distribution, while sharing the same composition. This definition is necessary to describe polydisperse populations and is consistent with the multi-modal formulation introduced in Appendix C. In this context, “mode” refers to a size-distribution-based grouping rather than chemical species, and is therefore not interchangeable with “species”.

(2.40) Line 183: …in unfrozen droplets, producing more ice.

We have made the following changes to the manuscript:

- Line 240–241: “In contrast, internal mixtures distribute the good nucleator across all INPs, increasing the probability of new nucleation events ~~on unfrozen droplets and maximizing freezing efficiency~~ in unfrozen droplets, producing more ice.”

(2.41) Line 204: How are the Φ identified as “typical”? Better “exemplary”.

We have made the following changes to the manuscript:

- Line 264: “…with ~~typical~~ exemplary Φ values denoted as Φ_A and Φ_B, respectively. ”

(2.42) Line 344: … are as follows.

We have made the following changes to the manuscript:

- Line 440–441: “…the details of the algorithm ~~is~~ are as follow.”

(2.43) Line 422 is a repetition of line 420.

We have made the following changes to the manuscript:

- Line 522–526: “As an example, Fig. 7 presents the results of Case 6 (all INPs consist of Fe_2O_3 and are exposed to a constant cooling rate of 2 $°C\ min^{-1}$), displaying the simulated frozen fraction ~~and comparing it with the analytical results using Eq. (B12)~~ , which is confirmed by the analytical solution using Eq. (B12) in Appendix B. As the temperature drops from −10 °C to −30 ∘C, more and more INPs become activated and the frozen fraction increases (green dots). ~~The simulation results are confirmed with the analytical solution using Eq. (B12) in Appendix B~~.”

(2.44) Line 508: specify “error” as “overprediction”.

We have made the following changes to the manuscript:

- Line 619–620: "All other intermediate mixing states would result in smaller ~~errors~~ overprediction."

(2.45) Line 580: do you mean "described" instead of "decreased"?

Yes, and we have made the following changes to the manuscript:

- Line 717: "The immersion freezing probability of supercooled droplet can be ~~decreased~~ described by the Poisson model . . . "

(2.46) Line 589: Clarify what simulation. At the point in the paper the appendix A is mentioned the simulation is not introduced. Do you mean calculation?

Here, we do not refer to a specific simulation setup, but rather to the general calculation framework used throughout the manuscript. In all frozen fraction calculations presented in this study, the temperature is assumed to remain below 0 °C, such that no melting occurs. As a result, once a droplet freezes, it remains frozen for the remainder of the calculation.

If the temperature were to rise above 0 °C and complete melting were to occur, the freezing process would need to be reinitialized (i.e., the time reset to zero) before further freezing calculations.

To avoid ambiguity, we have replaced the term "simulation" with the more general term "calculation" at the indicated location.

We have made the following changes to the manuscript:

- Line 725–727: "On the other hand, if it freezes at any time point, it will remain frozen for the remainder of the ~~simulation~~ calculation, as the melting process is not considered."

(2.47) Line 596: reformulate. In the limits $\Delta t \to \infty$ and $\Delta h \to \infty$, the unfrozen probability $P_{\mathrm{unf}}(0, t)$ is given by the time integral of λ.

In the derivation, h denotes the number of time intervals used to discretize the time period from 0 to t, and therefore represents an integer rather than a continuous increment. For this reason, we keep the notation $h \to \infty$ rather than $\Delta h \to \infty$. To avoid potential confusion, we have added a clarification in the manuscript: "where h is the number of time intervals dividing the time period from 0 to t."

We have made the following changes to the manuscript:

- Line 733: ". . . where h is the number of time intervals dividing the time period from 0 to t . . . "
- Line 734: ". . . the unfrozen probability $P_{\mathrm{unf}}(0, t)$ ~~will involve~~ is given by the time integration of λ:"

(2.48) Line 589: Pöschl.

We have corrected the LaTeX encoding issue to ensure that the character 'ö' is displayed correctly in the References. See Line 1174.

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