Responses to Reviewer #1

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.

(1.1) Overall, I find results from this study interesting. However, I am concerned about a key assumption—that the volume ratio of the species equals their surface area ratio. This assumption is unlikely to be held under realistic atmospheric conditions (e.g., I am thinking about aging or coating). My main concern is the representativeness of this assumption for real atmospheric INPs and how it affects the applicability of findings in this study to other research. I think it is worth including a discussion of this assumption, along with its potential limitations.

> We thank the reviewer for raising this important point. We agree that the assumption that the volume ratio equals the surface area ratio is unlikely to hold for many realistic atmospheric particles, particularly when particle morphology is influenced by processes such as aging or coating. We have added a discussion (Line 658–663) in the revised manuscript to clarify the role and limitations of this assumption.
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> First, we would like to emphasize that the theoretical framework presented in Section 3 does not rely on the assumption that the volume ratio of species equals their surface area ratio. The theory is formulated entirely in terms of surface area fractions of the constituent species, which directly determine heterogeneous ice nucleation activity. In this framework, the distribution of surface area among species is treated as the fundamental variable controlling freezing probability, independent of how that surface area arises from particle morphology or mass composition. Consistent with this perspective, the mixing-state index used in this study is also defined based on species surface area (see Appendix G).
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> The volume–surface equivalence assumption only appears in the particle-resolved simulations (Sections 4–5) as a practical modeling simplification. In PartMC, the composition of mixed particles is represented in terms of species mass, and particle morphology is not explicitly resolved. In the absence of additional morphological information, we adopt the assumption that species surface area fractions are proportional to their volume (or mass) fractions as a default representation. This allows us to control the relative surface area of species within mixed particles by specifying their mass fractions, enabling a systematic exploration of mixing-state effects in the model. Importantly, the simulations in this study are intended as a proof-of-concept demonstration of the theoretical framework rather than as a realistic simulation of specific atmospheric processes such as coating or aging. For realistic atmospheric applications, the relationship between species mass and surface availability will depend on particle morphology and internal structure, and more advanced representations would be required. Incorporating such morphology-dependent surface area parameterizations in particle-resolved models such as PartMC would be an important direction for future model development but is beyond the scope of the present study.
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> We have made the following changes to the manuscript:
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> - Line 658–663: Added “A related simplification arises in representing internally mixed particles 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.”
> - Line 666–667: Added “...and from simplified representations of species surface exposure.”

(1.2) Line 20: I suggest adding "extreme" before "external mixture" and "internal mixture" when first mentioning them in the paper, because there are many situations of "external" and "internal" mixture. "Each particle consists of a single chemical species" and "all particles share the same composition" are extreme conditions.

We have made the following changes to the manuscript:

- Line 26–29: Revised to "In an extreme external mixture, each particle consists of a single chemical species, whereas in an extreme internal mixture, all particles share the same composition (Winkler, 1973). For brevity, these two limiting cases are hereafter referred to as external and internal mixtures."

(1.3) Line 140: From the measurement point of view, people usually measure INP efficiency (INP spectrum) without knowing the chemical composition or mixing state of aerosols. Theorem 1 is not surprising from this point of view because internal mixture basically increases the number concentration of efficient INPs compared with external mixture. Therefore, I think it is equivalent to say the mixing state can change the INP spectrum. I wonder whether the authors can have one figure to show the change of INP spectrum by changing the mixing state.

Thank you for this insightful suggestion. We agree that interpreting the results from the perspective of INP spectra provides a useful connection between our theoretical framework and the way ice nucleation efficiency is typically represented in measurements.

We have made the following changes to the manuscript:

- Added Figure 6 in Section 3.2.
- 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."

(1.4) Line 186: Provide parameterization equation of Jhet and values of parameters of Fe2O3 and other species (e.g., those used in Figure 3) in the appendix, instead of just referring to Knopf and Alpert (2013).

We have made the following changes to the manuscript:

- Added Table B1.
- Line 840–844: Added "The heterogeneous ice nucleation rate coefficient used in this study is calculated with the water activity-based immersion freezing model (ABIFM) of Knopf and Alpert (2013):

  $$\log_{10} J_{\text{het}} = m \cdot (a_w - a_w^{\text{ice}}) + c, \tag{1}$$

  where J_{het} is in $\text{cm}^{-2}\ \text{s}^{-1}$, a_w is the water activity of the droplet, a_w^{ice} is the water activity in equilibrium with ice, and m and c are species-specific fitting parameters. Table B1 lists the values of m and c for the aerosol species used in this study."
- Figure 3 Caption: Added "(listed in Table B1)."

(1.5) Figure 3. Mention the simulation condition in the caption, e.g., a constant temperature of −33 °C and a duration of 10 minutes.

We have made the following changes to the manuscript:

- Figure 3 caption: 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).”

(1.6) Figure 7 caption. Specify “2, 5, and 9 minutes” in the caption, instead of saying “at specific times”.

We have made the following changes to the manuscript:

- Figure 8 (originally Figure 7) caption: Revised to “Snapshots of number concentration density distribution of unfrozen (green) and frozen droplets (blue) at at 2, 5, and 9 min corresponding to the blue dashed lines in the lower panel.”

(1.7) Line 388, “the χ index” is not defined before. Consider changing it to “the mixing state”.

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.”

References

Knopf, D. A. and Alpert, P. A.: A water activity based model of heterogeneous ice nucleation kinetics for freezing of water and aqueous solution droplets, Faraday discussions, 165, 513–534, https://doi.org/10.1039/C3FD00035D, 2013.

Riemer, N. and West, M.: Quantifying aerosol mixing state with entropy and diversity measures, Atmospheric Chemistry and Physics, 13, 11 423–11 439, https://doi.org/10.5194/acp-13-11423-2013, 2013.

Winkler, P.: The growth of atmospheric aerosol particles as a function of the relative humidity—II. An improved concept of mixed nuclei, Journal of Aerosol Science, 4, 373–387, https://doi.org/10.1016/0021-8502(73)90027-X, 1973.