

We would like to express our sincere gratitude to the editor and the anonymous reviewers for their valuable comments and suggestions, which have contributed to the improvement of our manuscript. Below, we provide a detailed, point-by-point response to each of the reviewers' comments. The reviewer's comments are presented in blue text, our responses are written in black text, and quotations from our manuscript are presented in italic type.

Referee #1

This review concerns the ACP manuscript of Hua Zhang et al. entitled “Contrasting Impacts of Dust Ice-Nucleating Particles on the Evolution and Radiative Effects of Mixed-Phase and Ice Clouds”.

The authors use a customized version of WRF-Chem to investigate the impact of variable INP setups on the cloud evolution and radiation budget over China and associated regions of Asia. The conclusions which the authors draw from their study are generally sound and of certain relevance to the community. Nevertheless, one has to consider that the study is kind of a synthesis report which deploys a range of existing methods and techniques to investigate the subject of dust impacts on the evolution of the tropospheric ice phase and the radiation budget. This was, however, done before by others, which is also acknowledged by the authors in the manuscript.

Given that the study is not a big leap forward for fundamental research (while it is certainly of some interest and impact for the community), there is no new technique that would require careful evaluation. After having read the study in the framework of this review I thus only identified mostly minor issues. The identification of these minor issues was happily possible rather easy as the quality of the English text is already in the current state pretty advanced.

Find below the listing of the comments. Only general minor (or rather semi-major) comments are listed here. I'm strongly in favor for a second round of reviews, as the implementation of some of my comments might have some impact on the study.

Response: We thank the reviewer for the careful and constructive evaluation of our manuscript, as well as for recognizing its clarity and overall quality. We appreciate the reviewer's assessment that the study is relevant to the community and that the identified issues are mostly minor.

We acknowledge the reviewer's concern that the study builds on existing methods and does not introduce a fundamentally new technique. We would like to clarify that the primary contribution of this work lies not in methodological novelty, but in providing a more comprehensive and process-based assessment of dust INP effects across both mixed-phase and ice clouds within a unified modeling framework. Previous studies have typically focused on either mixed-phase or ice clouds separately, or have treated their impacts independently. In contrast, our study explicitly represents dust INPs in both cloud regimes simultaneously and demonstrates that their effects cannot be

understood as a simple sum of the two, due to interactions between cloud phases and microphysical processes.

Specifically, by implementing improved INP parameterizations (including both non-size-resolved and size-resolved schemes) and evaluating them against observations, we provide a more complete representation of atmospheric dust INPs. This enables us to quantify the contrasting yet coupled impacts of dust INPs on cloud microphysics and radiation. Our results show that dust INPs enhance ice formation in both cloud types but through different pathways, leading to distinct radiative effects. These findings highlight the importance of jointly considering mixed-phase and ice cloud responses when assessing dust–cloud–radiation interactions.

In addition, following the reviewer's suggestions, we have strengthened the manuscript by clarifying the statistical methodology, expanding the discussion of other INP types, incorporating additional observational constraints, and improving figure readability. We have also revised the title to better reflect the modeling nature of the study.

We appreciate the reviewer's recommendation for a second round of review and believe that the revisions have improved the clarity, robustness, and scientific contribution of the manuscript.

The reviewer's comments are presented in blue text, our responses are written in black text, and quotations from our manuscript are presented in italic type.

General comments:

Title: The title is misleading. I was only triggered to accept the review because I was hoping for an observational study. I kindly ask the authors to modify the title of the study in such a way that it is clear to everybody that simulations are used. **Suggestion:** Simulations of the impacts of contrasts in dust ice-nucleating particles on the evolution and radiative effects of mixed-phase and ice clouds.

Response: We thank the reviewer for this helpful suggestion. We agree that the original title did not clearly indicate that the study is based on numerical simulations, which could potentially lead to confusion for readers expecting an observational study. Following the reviewer's recommendation, we have revised the title to explicitly highlight the modeling approach used in this work.

The title has been changed to: *"Simulation of Contrasting Impacts of Dust Ice-Nucleating Particles on the Evolution and Radiative Effects of Mixed-Phase and Ice Clouds."*

This revised title clearly communicates that the study is based on simulations while also emphasizing the contrasting impacts of dust ice-nucleating particles investigated in our work.

Fundamentals of the statistical analysis: The authors provide a lot of statistical analyses, including many mean values. But the theory behind the acquisition of these statistics is not outlined in the manuscript. The output of any model, especially with respect to aerosol and cloud microphysics yields usually output values in every grid box. The evaluation of statistics thus requires a custom decision of the authors about the applied thresholds, below which values are neglected and set to zero. Thus, which thresholds did the authors apply to define the ‘background’ or zero values? This of course addresses all parameters the authors analysed statistically.

I kindly ask the authors to elaborate a bit how the selected minimum realistic data values affected the derived statistics. How do mean and median values behave and which conclusions can be drawn from this comparison?

If there was no threshold applied to filter out the background/simulation noise, the authors must explain/describe what the impact of the background/simulation noise is on the mean/median values.

I see need that the authors add a new section to the manuscript where they describe the statistical approach and where they discuss the decisions for the thresholds and how the impact mean/median values. Perhaps that can be done by means of a table and a new section 2.5: Statistical methods.

Response: We thank the reviewer for their thoughtful comments regarding the statistical analysis and the application of thresholds in our study. In response, we have added a new section to the manuscript (Section 2.6: Statistical methods) to describe our statistical approach and the reason behind the thresholds used for various variables.

For most of the variables analyzed in the study, including PM₁₀ concentration, dust number concentration, dust INP concentration, and radiative variables, we did not apply any thresholds. These variables are relevant across all grid cells, and background noise is not a significant concern for their evaluation. In particular, we did not filter these variables because they provide important model output values that contribute to the overall assessment of aerosol-cloud interactions and radiation effects, which are critical for understanding large-scale processes.

However, for cloud-related variables (e.g., ice crystal number concentration, ice crystal effective radius, ice water content, cloud droplet number concentration, cloud droplet effective radius, and liquid water content), we did apply a threshold to filter out non-cloud grid cells. The criterion used for defining “cloud” grids was based on the sum of the ice water mixing ratio and liquid water mixing ratio exceeding 10^{-6} kg kg⁻¹, as suggested by (Hines et al., 2019). This threshold ensures that only grids with significant cloud water content are considered, which is essential for accurately analyzing aerosol-cloud interactions. We have clarified this methodology in the revised manuscript.

We also compared the mean values of cloud-related variables with and without the threshold applied (Table S3 and Table S4). Specifically, Table S3 (shown below)

corresponds to the mean values shown in Fig. 5, while Table S4 (shown below) corresponds to the mean values in Fig. 6. This comparison showed that, while the average values increased when filtering for cloud grids, the core relationships between dust INP concentrations and cloud responses remained almost identical. This indicates that the dust-cloud interactions, which are central to our study, were not significantly affected by the thresholding approach. The filtering simply helped focus the analysis on the grid cells where dust impacts on cloud properties are most relevant, without substantially altering the conclusions about the effects of dust INPs on cloud microphysics. We have updated Fig. 5 and 6 to show the results with threshold filtering applied to cloud grids, which we believe offers a more accurate representation of the data.

Regarding the use of median values, we primarily focused on mean values in our analysis, as they provide a comprehensive overview of model performance and dust-cloud interactions over the entire study region. Median values, while useful in some contexts, do not always represent the overall distribution of values effectively, especially in large-scale simulations where mean values are more representative of regional-scale processes. Therefore, we did not conduct median value analyses in this study.

We hope these changes provide the necessary clarifications and that the new section addressing the statistical methods and thresholds enhances the transparency of our approach.

Table S3. Mean values of cloud-related variables with and without threshold filtering applied to cloud grids, based on the meridional mean vertical cross-sections shown in Fig. 5. The filtering criterion is that the sum of the ice water mixing ratio and liquid water mixing ratio exceeds $10^{-6} \text{ kg kg}^{-1}$. The abbreviations used are as follows: ICNC (ice crystal number concentration; L^{-1}), ICER (ice crystal effective radius; μm), IWC (ice water content; mg m^{-3}), CDNC (cloud droplet number concentration; 10^3 L^{-1}), CDER (cloud droplet effective radius; μm), and LWC (liquid water content; mg m^{-3}). The “NoFilter” row corresponds to results without threshold filtering, while the “Filter” row corresponds to results with threshold filtering applied.

Scenarios	Averaging method	ICNC	ICER	IWC	CDNC	CDER	LWC
NoINPs	NoFilter	6.80E-01	3.00E+01	3.10E+00	1.60E+00	1.60E+01	3.30E+00
	Filter	1.10E+00	3.50E+01	4.40E+00	2.20E+00	1.70E+01	4.50E+00
MixINPs	NoFilter	4.60E-03	1.30E-01	1.10E-01	-7.70E-02	1.40E-02	-7.60E-02
	Filter	2.90E-03	2.00E-01	1.50E-01	-1.00E-01	2.10E-02	-1.00E-01
IceINPs	NoFilter	6.30E+00	-1.30E+00	1.20E+00	-2.60E-01	3.90E-01	-2.40E-01
	Filter	9.40E+00	-2.00E+00	1.70E+00	-3.50E-01	4.00E-01	-3.40E-01
MixIceINPs	NoFilter	6.30E+00	-1.30E+00	1.30E+00	-2.70E-01	3.90E-01	-2.70E-01
	Filter	9.50E+00	-2.00E+00	1.80E+00	-3.70E-01	4.00E-01	-3.80E-01
(MixINPs – NoINPs) / NoINPs	NoFilter	0.68%	0.43%	3.55%	-4.81%	0.09%	-2.30%
	Filter	0.26%	0.57%	3.41%	-4.55%	0.12%	-2.22%
(IceINPs – NoINPs) / NoINPs	NoFilter	926.47%	-4.33%	38.71%	-16.25%	2.44%	-7.27%
	Filter	854.55%	-5.71%	38.64%	-15.91%	2.35%	-7.56%
(MixIceINPs – NoINPs) / NoINPs	NoFilter	926.47%	-4.33%	41.94%	-16.88%	2.44%	-8.18%
	Filter	863.64%	-5.71%	40.91%	-16.82%	2.35%	-8.44%

Table S4. Mean values of cloud-related variables with and without threshold filtering applied to cloud grids, based on the horizontal distributions shown in Fig. 6. The filtering criterion is that the sum of the ice water mixing ratio and liquid water mixing ratio exceeds $10^{-6} \text{ kg kg}^{-1}$. The abbreviations used are as follows: ICND (ice crystal number density; vertically integrated ice crystal number concentration; 10^6 m^{-2}), ICER (ice crystal effective radius; vertically averaged; μm), IWP (ice water path; vertically integrated ice water content; g m^{-2}), CDND (cloud droplet number density; vertically integrated cloud droplet number concentration; 10^9 m^{-2}), CDER (cloud droplet effective radius; vertically averaged; μm), and LWP (liquid water path; vertically integrated liquid water content; g m^{-2}). The “NoFilter” row corresponds to results without threshold filtering, while the “Filter” row corresponds to results with threshold filtering applied.

Scenarios	Averaging method	ICND	ICER	IWP	CDND	CDER	LWP
NoINPs	NoFilter	1.50E+01	3.20E+01	5.50E+01	2.20E+01	1.60E+01	4.40E+01
	Filter	1.40E+01	3.40E+01	5.60E+01	2.20E+01	1.60E+01	4.40E+01
MixINPs	NoFilter	7.00E-02	1.30E-01	1.80E+00	-1.40E+00	2.50E-02	-1.30E+00
	Filter	6.60E-02	3.70E-01	1.80E+00	-1.40E+00	3.10E-02	-1.30E+00
IceINPs	NoFilter	1.30E+02	4.30E-01	2.30E+01	-4.20E+00	4.70E-01	-3.70E+00
	Filter	1.30E+02	4.00E-01	2.30E+01	-4.20E+00	5.00E-01	-3.80E+00
MixIceINPs	NoFilter	1.30E+02	4.80E-01	2.30E+01	-4.50E+00	4.70E-01	-4.10E+00
	Filter	1.30E+02	4.50E-01	2.40E+01	-4.50E+00	5.10E-01	-4.10E+00
(MixINPs – NoINPs) / NoINPs	NoFilter	0.47%	0.41%	3.27%	-6.36%	0.16%	-2.95%
	Filter	0.47%	1.09%	3.21%	-6.36%	0.19%	-2.95%
(IceINPs – NoINPs) / NoINPs	NoFilter	866.67%	1.34%	41.82%	-19.09%	2.94%	-8.41%
	Filter	928.57%	1.18%	41.07%	-19.09%	3.13%	-8.64%
(MixIceINPs – NoINPs) / NoINPs	NoFilter	866.67%	1.50%	41.82%	-20.45%	2.94%	-9.32%
	Filter	928.57%	1.32%	42.86%	-20.45%	3.19%	-9.32%

Limitation to only dust INP: Meanwhile it is widely accepted that ice formation at temperatures above -15°C is mostly determined by biological particles (Cornwell et al., 2023). This was not discussed at all by the authors. There should be some information about this important INP type provided in the introduction and in the conclusions section. Another important INP type, especially at Cirrus level is smoke (Ansmann et al., 2025), which might also deserve some discussion in the introduction and conclusions.

Response: We thank the reviewer for this insightful and constructive comment. We agree that biological particles at relatively warm temperatures above -15°C and smoke particles in cirrus regimes represent important INP types that deserve discussion. Our original manuscript focused exclusively on dust INPs in order to isolate their microphysical and radiative impacts. However, we acknowledge that this focus should be better contextualized within the broader INP framework.

We have revised both the *Introduction* and the *Conclusions* sections to explicitly (1) acknowledge the role of biological particles in ice nucleation at warmer temperatures in mixed-phase clouds, and the potential contribution of smoke particles in cirrus clouds, and (2) discuss the importance of these particles in ice nucleation processes during the spring in East Asia. We further clarify that our study design intentionally isolates dust to reduce complexity and enable mechanistic interpretation.

The revision to the *Introduction* section in the revised manuscript (Lines 62–78) is shown below:

“Other aerosol types may also contribute to heterogeneous ice formation under certain conditions. For example, biological particles such as bacteria, fungal spores, and pollen fragments can dominate ice nucleation at relatively warm temperatures above about -15°C (Cornwell et al., 2023). Biomass-burning smoke may also act as INPs under cirrus conditions, although their ice-nucleating efficiency depends strongly on particle composition and atmospheric processing (Mertes et al., 2007; Phillips et al., 2013; Vergara-Temprado et al., 2018; Nichman et al., 2019).”

The revision to the *Conclusions* section in the revised manuscript (Lines 987–995) is shown below:

“It should be noted that this study focuses exclusively on mineral dust as the source of INPs. The simulations were conducted for spring over East Asia, a period characterized by frequent dust outbreaks originating from the Taklimakan and Gobi deserts. Under these conditions, mineral dust is expected to dominate the regional INP population and therefore represents the primary driver of heterogeneous ice nucleation in the simulated environment. Nevertheless, other aerosol types such as biological particles, biomass-burning smoke may also contribute to ice nucleation under different environmental conditions or during other seasons. Their potential roles should therefore be investigated in future studies.”

Lack of referencing to observational studies of INP/ICNC relationships under ambient conditions: There are meanwhile plenty of observational studies discussing relationships between INP concentration and cloud properties. It would be nice if the authors would add some to their study (introduction). Here some examples (e.g., Zhang et al., 2018; Ansmann et al., 2019; He et al., 2025, Villanueva et al., 2025). There are also nice detailed spectral-bin simulations of the impact of perturbations in INP on mixed-phase cloud properties (Lee et al., 2024).

Response: We thank the reviewer for this insightful and constructive comment. In response, we have revised the Introduction to incorporate references to several relevant observational studies that discuss the relationship between INP concentrations and cloud properties. Specifically, we added references to studies highlighting the association between dust loading and ice particle production in mixed-phase clouds (Zhang et al., 2018), lidar-derived INP concentrations in Saharan dust layers explaining ice crystal number concentrations (Ansmann et al., 2019), and long-term observations demonstrating the influence of INP variations on mixed-phase cloud microphysics and precipitation formation (He et al., 2025). We also included satellite analyses suggesting the role of dust-driven droplet freezing in determining cloud-top phase (Villanueva et al., 2025). Additionally, we discussed high-resolution spectral-bin simulations (Lee et al., 2024) that provide process-level insight into how perturbations in INP concentrations can influence mixed-phase cloud microphysics.

We have included observational studies and spectral-bin simulations in the *Introduction* section, as suggested by the reviewer (Lines 88–110). The revised text is shown below:

“Observational studies have provided important constraints on the relationship between INPs and cloud microphysics. For example, collocated A-Train satellite observations reveal that enhanced ice particle production in mid-level stratiform mixed-phase clouds is associated with elevated dust loading, indicating that dust-induced ice formation can substantially modify cloud phase and ice crystal number concentrations (Zhang et al., 2018). Aerosol–cloud closure studies further demonstrate that lidar-derived INP concentrations in Saharan dust layers can explain observed ice crystal number concentrations within an order of magnitude, highlighting the importance of heterogeneous nucleation on mineral dust in altocumulus and cirrus (Ansmann et al., 2019). More recently, long-term observations from two sites with contrasting aerosol conditions, characterized by desert dust and continental aerosols versus marine aerosol, also indicate that variations in INP concentrations can significantly influence mixed-phase cloud microphysics and precipitation formation (He et al., 2025). At larger spatial scales, satellite analyses further suggest that dust-driven droplet freezing plays a key role in determining cloud-top phase across the Northern Hemisphere (Villanueva et al., 2025).”

“Process-level insight into these interactions has also been obtained from high-resolution spectral-bin simulations. For example, Lee et al. (2024) demonstrated that higher INP concentrations lead to smaller ice particles and more irregular crystal

shapes, highlighting the influence of INP levels on ice particle morphology, number, and cloud microphysics. However, these studies typically focus on idealized cloud systems and therefore do not directly quantify the large-scale impacts of INPs on cloud evolution and radiation.”

Impact for future studies: I suggest to the authors to extend their conclusions a bit by means of an outlook. Their study has the potential to evaluate how projected changes in landuse and surface type will probably effect the regional climate in China and surrounding parts of Asia. It would be an easy task to just modify the dust immersion scheme in order to investigate how clouds and radiation would change with changing emissions (and a certainly warmer climate). Such a future study would be a really nice follow-up to the presented one.

Response: We thank the reviewer for this insightful and constructive suggestion. We agree that including a forward-looking perspective on factors influencing dust emissions and their subsequent influence on regional climate would be a valuable addition to the conclusions. In response, we have expanded the Conclusions section to include a brief outlook on how future studies could investigate the effects of climate change and land use changes (such as afforestation) on key factors such as vegetation cover, soil moisture and wind speed, which in turn affect dust emissions. These changes in dust emissions could further impact cloud properties and radiative effects through dust-cloud interactions, providing an important direction for extending the current study.

The revision has been made in accordance with the reviewer’s suggestion in the revised manuscript (Lines 1000–1005). The revised text is shown below:

“Looking ahead, future studies could investigate how climate change and land use changes (such as afforestation) might alter vegetation cover, soil moisture, and wind speed, which in turn affect dust emissions. These changes in dust emissions would likely influence cloud properties and radiative effects through dust-cloud interactions, offering an important area for further research in the context of regional climate impacts in East Asia and surrounding regions.”

We hope these revisions effectively address the reviewer’s suggestion by providing a forward-looking perspective on the potential implications of future changes in dust emissions due to climate change and carbon-neutral policies.

All figures: I kindly request that the authors aim on increasing the size of fonts and legends in all of their figures to the maximum. The figures are very hard to read. I was hardly able to read any number in the printed version of the manuscript. Especially Figs. 5 and 6.

Response: We thank the reviewer for pointing out this issue. Following the reviewer’s suggestion, we have increased the font sizes of all labels, tick marks, legends, and

annotations in every figure to improve readability. Particular attention was given to Fig. 5 and 6, where the labels and legends have been significantly enlarged to ensure that all values and annotations are clearly readable in the printed version of the manuscript.

Reference

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Referee #2

In this work, the authors introduce two new parameterizations for ice-nucleating particles (INPs) in WRF-Chem to investigate the impacts of dust INPs on mixed-phase and ice clouds and their radiative effects. Given the large uncertainties surrounding the role of dust emissions in ice formation and cloud evolution, this study is relevant to the community. The methods and results are described in detail, the manuscript is generally well written, and the inclusion of model validation is commendable.

Response: We thank the reviewer for the positive and encouraging assessment of our work. We appreciate the recognition of the relevance of this study, as well as the clarity of the presentation and the inclusion of model validation.

However, I find the paper largely descriptive, with few overarching conclusions that advance the state of the field, and I question whether it is a good fit for the journal. Several mechanisms are proposed to explain the results, but these remain mostly speculative. In addition, there may be fundamental flaws in the implementation of the ice nucleation parameterizations that warrant major revisions. These issues must be addressed before the paper can be considered for publication.

Response: We thank the reviewer for the constructive and critical assessment. We acknowledge the concerns regarding the descriptive nature of the manuscript, the lack of direct evidence supporting the proposed mechanisms, and the potential issues in the implementation of the ice nucleation parameterizations.

In response, we have substantially revised the manuscript to address these points. Specifically, we have added a dedicated process-level analysis (Section 3.2.6) to explicitly diagnose key microphysical mechanisms, including the Wegener–Bergeron–Findeisen (WBF) process and ice crystal sedimentation, using model output. These diagnostics provide direct evidence supporting the interpretations and make the proposed mechanisms central to the conclusions rather than speculative.

We have also clarified the implementation of the ice nucleation parameterizations, particularly the treatment of subgrid vertical velocity, and added a new sensitivity experiment (Section 3.2.5) using an alternative TKE-based parameterization. The results demonstrate that our conclusions are robust and not driven by the representation of vertical velocity.

In addition, the discussion has been strengthened throughout the manuscript, and Fig. 10 has been repositioned and better integrated with the supporting analysis. Together, these revisions address the reviewer’s concerns and improve the overall scientific rigor and clarity of the study.

The reviewer’s comments are presented in blue text, our responses are written in black text, and quotations from our manuscript are presented in italic type.

General comments:

Overall, the manuscript is clearly written, detailed, and the simulation experiments are well designed. At the same time, the discussion often falls back on straightforward descriptions of figures without deeper analysis. When mechanistic explanations are proposed (for example involving sedimentation or the WBF process) they are not supported by evidence. It should be possible to output the relevant microphysical rates from the model to demonstrate that the proposed mechanisms are actually operating. As written, these explanations are often lost in the text and do not appear central to the paper's conclusions. I encourage the authors to include a dedicated discussion section where these mechanisms can be addressed in more depth, and where Figure 10 and the hypotheses raised in the conclusions can be more fully developed.

Response: We thank the reviewer for this constructive and important suggestion. We agree that the discussion of physical mechanisms should be more explicitly supported by model diagnostics and more clearly integrated into the main conclusions.

Following this suggestion, we have added a dedicated analysis section (3.2.6 Microphysical process analysis) in the revised manuscript to directly examine the key microphysical processes underlying our interpretations, including the WBF process and sedimentation.

For the WBF process, we note that the Morrison microphysics scheme does not provide a direct diagnostic variable for the WBF rate. Instead, it represents the underlying processes through individual microphysical tendencies, including hydrometer collection, condensation, evaporation, freezing, melting, deposition, sublimation, sedimentation, and ice nucleation (Morrison et al., 2005). To diagnose the WBF process, we output the rates of cloud droplet evaporation and vapor deposition, and restrict the analysis to grid cells that satisfy WBF-favorable conditions (Pruppacher et al., 1998): supersaturation with respect to ice, subsaturation with respect to liquid water, temperatures between -37 and 0°C , and coexistence of cloud droplets and ice crystals. Under these conditions, the transfer of water from liquid to ice phase can be interpreted as a proxy for the WBF process.

Figure S19 (shown below) show that, in the MixINPs scenario, both cloud droplet evaporation and vapor deposition rates are significantly enhanced compared to the NoINPs scenario, with comparable magnitudes, indicating an accelerated transfer of water from liquid to ice phase. This provides direct evidence that the WBF process is strengthened by dust-induced ice nucleation.

For sedimentation, we analyzed the diagnosed ice crystal sedimentation flux. Figure S20 (shown below) indicates that, in the IceINPs scenario, regions with enhanced sedimentation flux correspond to areas with increased ice crystal number concentration compared to the NoINPs scenario (Fig. 5c). Meanwhile, the increase in sedimentation flux below these regions is relatively weaker, suggesting that a portion of sedimenting ice crystals is retained within the underlying mixed-phase cloud layers.

This accumulation of ice crystals in mixed-phase clouds contributes to enhanced ice number concentrations and further reinforces the WBF process, as supported by the diagnosed cloud droplet evaporation and vapor deposition rates in Fig. S19b and S19e.

These additional diagnostics confirm that the mechanisms proposed in the original manuscript are physically consistent and actively operating in the simulations. We have incorporated this analysis into a new section in the revised manuscript and expanded the discussion of Fig. 10 accordingly, making these mechanisms more central to the overall conclusions.

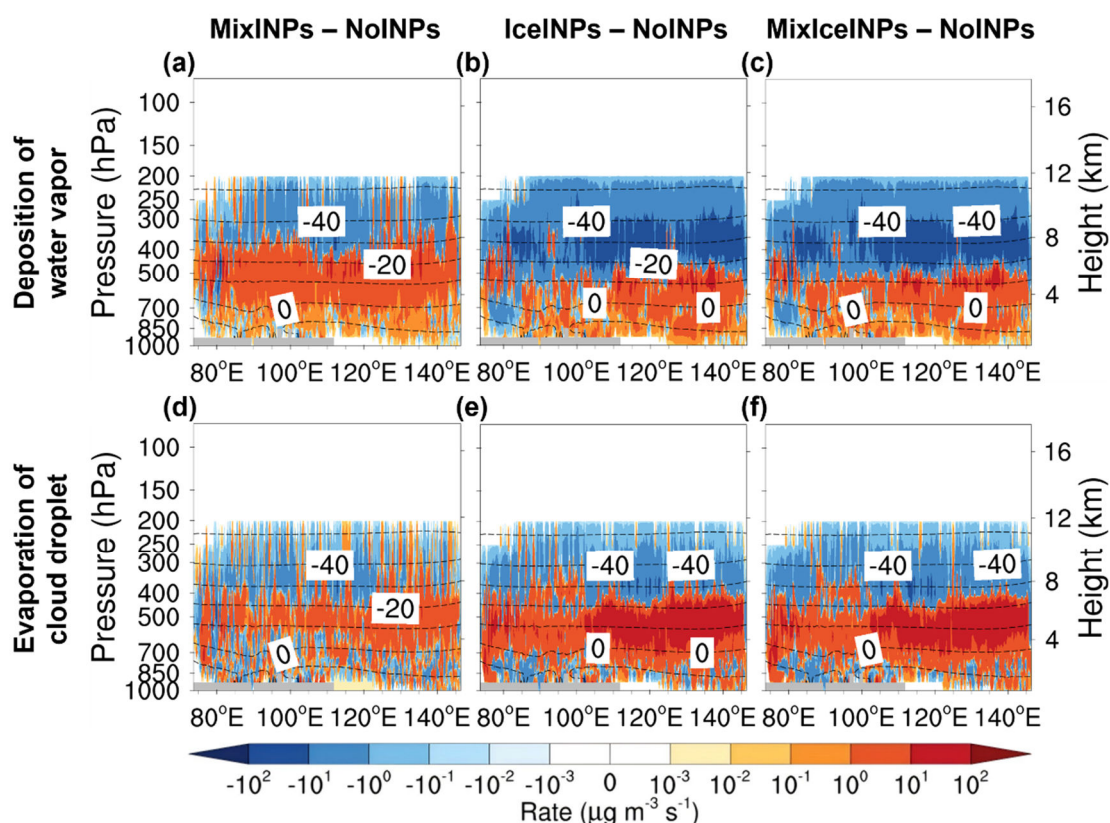


Figure S19. Simulated meridional mean vertical cross-sections of (a–c) deposition rate of water vapor, (d–f) evaporation rate of cloud droplet, for the MixINPs – NoINPs (first column), IceINPs – NoINPs (second column), and MixIceINPs – NoINPs (third column) scenarios. Dashed lines indicate isotherms, and grey shading denotes missing values due to terrain. The simulated results are averages of hourly values from March to May 2018, calculated for grid cells where WBF process occurs, defined by the conditions: (1) the air is supersaturated with respect to ice but subsaturated with respect to liquid water, (2) temperature ranges between -37 and 0°C , and (3) cloud droplets and ice crystals coexist.

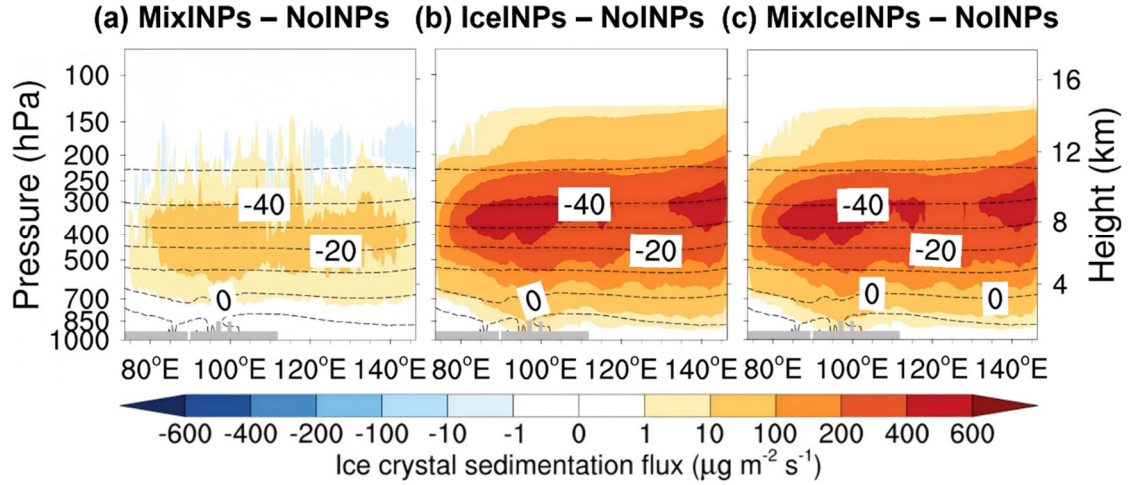


Figure S20. Simulated meridional mean vertical cross-sections of ice crystal sedimentation flux differences: **(a)** MixINPs – NoINPs, **(b)** IceINPs – NoINPs, and **(c)** MixIceINPs – NoINPs. Dashed lines indicate isotherms, and grey shading denotes missing values due to terrain. The simulated results are averages of hourly values from March to May 2018.

More fundamentally, there may be a major flaw in the implementation of the ice formation parameterizations for ice clouds. It is never clear what subgrid-scale vertical velocity is used. At the model resolution employed (27 km), the dynamics of individual parcels are not resolved; using grid-scale vertical velocities would significantly underestimate peak supersaturation and ice formation rates, particularly for homogeneous ice nucleation. This issue would strongly affect, and likely alter, the conclusions of the study. The authors should provide more detailed information on the implementation of the ice nucleation parameterizations and clearly state the assumptions made regarding subgrid-scale dynamics.

Response: We thank the reviewer for raising this important concern regarding the treatment of subgrid-scale vertical velocity in the ice nucleation parameterizations. We agree that at the model resolution used in this study (27 km), grid-scale vertical velocities cannot adequately represent the small-scale variability that governs supersaturation and ice formation, particularly for homogeneous nucleation.

In our implementation, this issue is addressed through a subgrid-scale vertical velocity parameterization following Morrison et al. (2005), based on Ghan et al. (1997). The vertical velocity used in the microphysical calculations is not the resolved grid-scale velocity, but a characteristic subgrid turbulent vertical velocity (w'), representing unresolved fluctuations associated with turbulent mixing. The subgrid vertical velocity is parameterized as:

$$w' = \sqrt{2\pi} \frac{K}{\Delta z}$$

where K (in $\text{m}^2 \text{s}^{-1}$) is the vertical eddy diffusivity and Δz is the model layer thickness.

In this framework, w' is diagnosed from turbulence properties and linked to the intensity of vertical mixing, typically through the vertical eddy diffusivity and the model layer thickness. This parameterized vertical velocity determines the local cooling rate and thus controls supersaturation and ice nucleation rates. Such approaches have been widely adopted in cloud microphysics schemes in both regional and global atmospheric models (Storelvmo et al., 2006; Morrison and Gettelman, 2008).

This treatment allows the model to account for unresolved vertical motions that are essential for cloud microphysical processes, thereby avoiding the substantial underestimation of supersaturation and ice formation that would occur if only grid-scale vertical velocities were used.

To clarify this aspect, we have revised the manuscript (Lines 317–333) to explicitly describe the role of subgrid vertical velocity, its formulation, and the associated assumptions.

Minor comments:

Line 69: Remove “simple.”

Response: We have removed the word “simple” as suggested.

Lines 92–94: Please clarify this sentence. Parameterizations derived for cirrus should be applicable wherever cirrus clouds occur.

Response: We apologize for any confusion caused by the original wording. We appreciate the reviewer’s suggestion for clarification. To address the concern, we have revised the sentence to better explain that INP parameterizations derived for cirrus clouds, which do not account for mixed-phase cloud conditions, cannot fully represent the impact of dust INPs on clouds and radiation.

The revision has been made in accordance with the reviewer’s suggestion in the revised manuscript (Lines 138–140). The revised text is shown below:

“Because most schemes are based solely on cirrus cloud conditions and neglect mixed-phase cloud conditions, they do not fully capture the impact of dust INPs on clouds and radiation.”

Line 159: Consider “the latter mechanism remains...”

Response: We appreciate the reviewer’s suggestion. We have revised the sentence in the revised manuscript (Line 205) to incorporate “the latter mechanism remains” for greater clarity and flow.

Section 2.2.1: Whether immersion freezing is restricted to the presence of liquid water has a significant impact on simulated cloud properties (see, e.g., Tan and Barahona, 2022).

Response: We appreciate the reviewer's comment. We confirm that, in our model, the immersion freezing scheme is explicitly restricted to the presence of liquid water droplets. Immersion freezing only contributes when liquid water droplets are present, as the activation of INPs occurs within these droplets. We have revised the manuscript to better reflect this clarification.

The revised text in the revised manuscript (Lines 223–224) is shown below:

“Immersion freezing is restricted to the presence of liquid water droplets in our model, as INP activation occurs only within these droplets.”

Lines 215–217: Any droplet reaching -40°C would freeze; it is unclear why heterogeneous ice nucleation would be more likely.

Response: We appreciate the reviewer's comment. For cloud droplets, freezing is expected to occur at temperatures below -40°C . However, for aerosol solution, the occurrence of homogeneous or heterogeneous nucleation is governed by supersaturation, and freezing does not necessarily occur below -40°C (Koop et al., 2000; Kanji et al., 2017). Heterogeneous nucleation requires lower supersaturation and is therefore easier to occur. This is because INPs provide a surface for ice formation, lowering the activation barrier for freezing. In contrast, homogeneous nucleation requires the formation of a critical ice cluster purely from water molecules, which is more difficult to occur. We have revised the sentence to better clarify this point.

The revised text in the revised manuscript (Lines 261–267) is shown below:

“In the default Morrison microphysics scheme, all cloud droplets and raindrops are assumed to homogeneously freeze into ice crystals at temperatures below -40°C . However, for aerosol solution, the occurrence of homogeneous or heterogeneous nucleation is governed by supersaturation, and freezing does not necessarily occur below -40°C . Heterogeneous nucleation requires lower supersaturation and is therefore easier to occur, as INPs provide surfaces for ice formation, thereby lowering the activation barrier for freezing (Koop et al., 2000; Kanji et al., 2017).”

Line 224: “Before” is ambiguous here.

Response: We apologize for the ambiguity in the original sentence. We have replaced “before” with “earlier than” to clearly indicate the temporal sequence of events, meaning that heterogeneous nucleation can occur prior to homogeneous nucleation. This revision clarifies the intended meaning and eliminates potential confusion.

The revised text in the revised manuscript (Lines 272–275) is shown below:

“In ice clouds, homogeneous nucleation of supercooled droplets can produce numerous small ice crystals, whereas heterogeneous nucleation requires lower relative humidity, may occur earlier than homogeneous nucleation, produces fewer but larger ice crystals, and suppresses homogeneous nucleation.”

Line 316: Please specify whether, in the NoINPs experiment, INPs are represented by the default parameterization or removed entirely.

Response: We appreciate the reviewer’s comment. In the NoINPs experiment, INPs are removed entirely, meaning that heterogeneous nucleation is completely turned off. We have clarified this in the manuscript.

The revised text in the revised manuscript (Lines 384–386) is shown below:

“NoINPs: Heterogeneous nucleation was turned off in both mixed-phase and ice clouds (INPs were removed entirely), while homogeneous nucleation of cloud droplets was active.”

Line 348: Consider “homogeneous and heterogeneous ice nucleation in ice clouds followed...”

Response: We appreciate the reviewer’s suggestion. We have revised the sentence to clarify that both homogeneous and heterogeneous nucleation processes in ice clouds are based on the same reference.

The revised text in the revised manuscript (Lines 417–418) is shown below:

“Homogeneous and heterogeneous ice nucleation in ice clouds followed Barahona and Nenes (2009).”

Line 393: This is a large number of sites. Please describe how samples are collected.

Response: We appreciate the reviewer’s comment. As mentioned in the Section 2.5 (Measurements) of the manuscript, surface PM₁₀ concentrations were obtained from the National Environmental Monitoring Center of China (<https://quotsoft.net/air/>, last access: 28 Oct. 2025). We have also clarified in this section that the data correspond to the sites shown in Fig. 1. Additionally, the figure caption has been revised to indicate the source of the observation data.

The revision to the Section 2.5 (Measurements) in the revised manuscript (Lines 420–422) is shown below:

“Surface PM₁₀ concentrations were obtained from the National Environmental Monitoring Center of China (<https://quotsoft.net/air/>, last access: 28 Oct. 2025) and

correspond to the observation sites shown in Fig. 1.”

The revision to the caption of Fig. 1 in the revised manuscript (Lines 503–508) is shown below:

Figure 1. Comparison between simulated and observed surface monthly mean PM_{10} mass concentrations in (a) March, (b) April, and (c) May 2018. Circles denote monthly mean observations from 1,035 sites, and color shading indicates the corresponding simulated monthly means. The statistics shown above each panel include the normalized mean bias (NMB), normalized mean error (NME), and correlation coefficient (R). The simulated results are from the MixIceINPs scenario and represent averages of hourly values. The observation data were obtained from the National Environmental Monitoring Center of China.

Figure 2: How is supersaturation diagnosed? What assumptions are made regarding subgrid-scale variability?

Response: We greatly appreciate the reviewer’s thoughtful comments. As mentioned in Section 2.5 (Measurements) of the manuscript: *For comparison with observations, modeled INP concentrations were diagnosed offline using parameterization schemes and simulated aerosol properties.* In the evaluation, we assume that the supersaturation used in the simulation is the same as that in the observations, and we do not consider subgrid-scale variability of supersaturation. This is because the observed INP concentrations are measured under specific temperature and ice supersaturation conditions, and including subgrid-scale variability in the simulation would not be meaningful for this comparison.

The observational INP concentrations are derived from environmental aerosol samples collected at the surface, where the ice nucleation activity is measured under controlled temperatures and ice supersaturation conditions. These observations reflect the potential of aerosols to activate as INPs, but they do not provide direct measurements of actual INP concentrations in the atmosphere. To compare the simulation results with the observational data, we apply the same activation conditions (i.e., temperature and ice supersaturation) as those used in the observations. The simulated INP concentrations are then calculated based on the INP parameterization schemes and the simulated aerosol number concentrations.

In summary, the evaluation compares the offline-diagnosed simulated INP concentrations, calculated under the same supersaturation conditions as those in the observations, with the observed INP data. As the evaluation is based on this comparison approach, subgrid-scale variability is not included in this process.

We have revised Section 2.5 (Measurements) to clarify our evaluation approach for simulated dust INP concentrations. The revision in the manuscript (Lines 454–463) is shown below:

“For comparison with observations, modeled INP concentrations were diagnosed

offline using parameterization schemes and simulated aerosol properties. The supersaturation used in the simulation is assumed to be identical to that used in the observations, and no subgrid-scale variability is considered. The observational INP concentrations are derived from aerosol samples collected at surface sites under specific temperatures and ice supersaturation conditions, reflecting the potential for aerosol activation as INP. To compare the simulation results with the observed data, the same activation conditions (temperature and ice supersaturation) as in the observations are applied in the simulation, and the modeled INP concentrations are calculated based on the simulated aerosol number concentrations and the relevant parameterization schemes.”

Lines 524–526: Low ice crystal concentrations from homogeneous ice nucleation may indicate overly weak vertical velocities.

Response: We thank the reviewer for this insightful comment. We agree that overly weak vertical velocities could potentially lead to underestimated supersaturation and thus reduced homogeneous ice nucleation.

To further examine this issue, we conducted an additional sensitivity experiment using an alternative subgrid vertical velocity parameterization based on turbulent kinetic energy (TKE), following Morrison and Pinto (2005). The subgrid vertical velocity is parameterized as:

$$w' = \sqrt{\frac{2}{3} TKE}$$

In this approach, the subgrid vertical velocity is diagnosed from TKE provided by the planetary boundary layer (PBL) scheme, which generally yields stronger vertical velocity fluctuations compared to eddy-diffusivity-based formulations, as supported by the simulated subgrid vertical velocity in Fig. S12 (shown below).

Previous studies have shown that TKE-based subgrid vertical velocity tends to produce higher updraft velocities and may even lead to overestimation of ice crystal number concentrations (Zhou et al., 2016). Therefore, this experiment provides a useful upper-bound test for the role of subgrid vertical velocity in controlling homogeneous nucleation.

Figures S13 and S14 (shown below) show that, despite the use of stronger subgrid vertical velocities in the TKE-based scheme, the overall behavior remains unchanged: the contribution of homogeneous nucleation remains much smaller than that of heterogeneous nucleation, and the low concentration of homogeneously formed ice crystals persists. This indicates that the limited occurrence of homogeneous nucleation in our simulations is not primarily caused by weak vertical velocities, but rather by the relatively low frequency of sufficiently high ice supersaturation conditions. Furthermore, the relatively low frequency of high ice supersaturation is

consistent with the climate characteristics of the dust regions of East Asia in spring, where dry conditions prevail (Gerelmaa et al., 2015).

Therefore, we conclude that our original interpretation remains valid. To clarify this point, we have added a new sensitivity experiment and corresponding discussion (Section 3.2.5: Sensitivity to subgrid vertical velocity parameterization) in the revised manuscript (Lines 877–909).

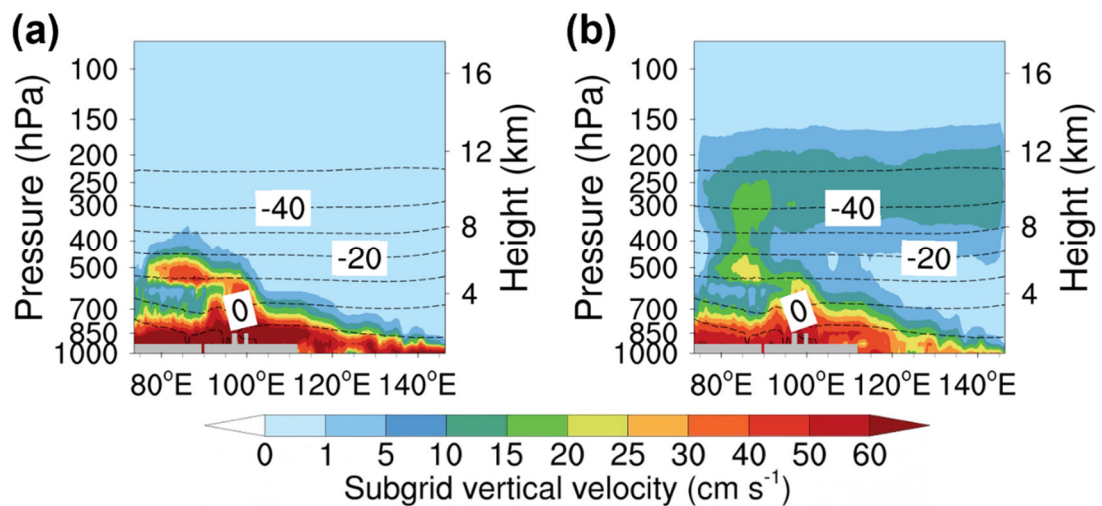


Figure S12. Simulated meridional mean vertical cross-sections of subgrid vertical velocity from (a) the eddy-diffusivity-based parameterization and (b) the turbulent-kinetic-energy-based parameterization. Dashed lines indicate isotherms, and grey shading denotes missing values due to terrain. The simulated results are averages of hourly values from May 2018. The results for May are very similar to those for the entire spring season (March–May), as shown in Fig. 3 and Fig. S13, Fig. 5 and Fig. S17, and Fig. 6 and Fig. S17, indicating that May’s results effectively represent the spring season.

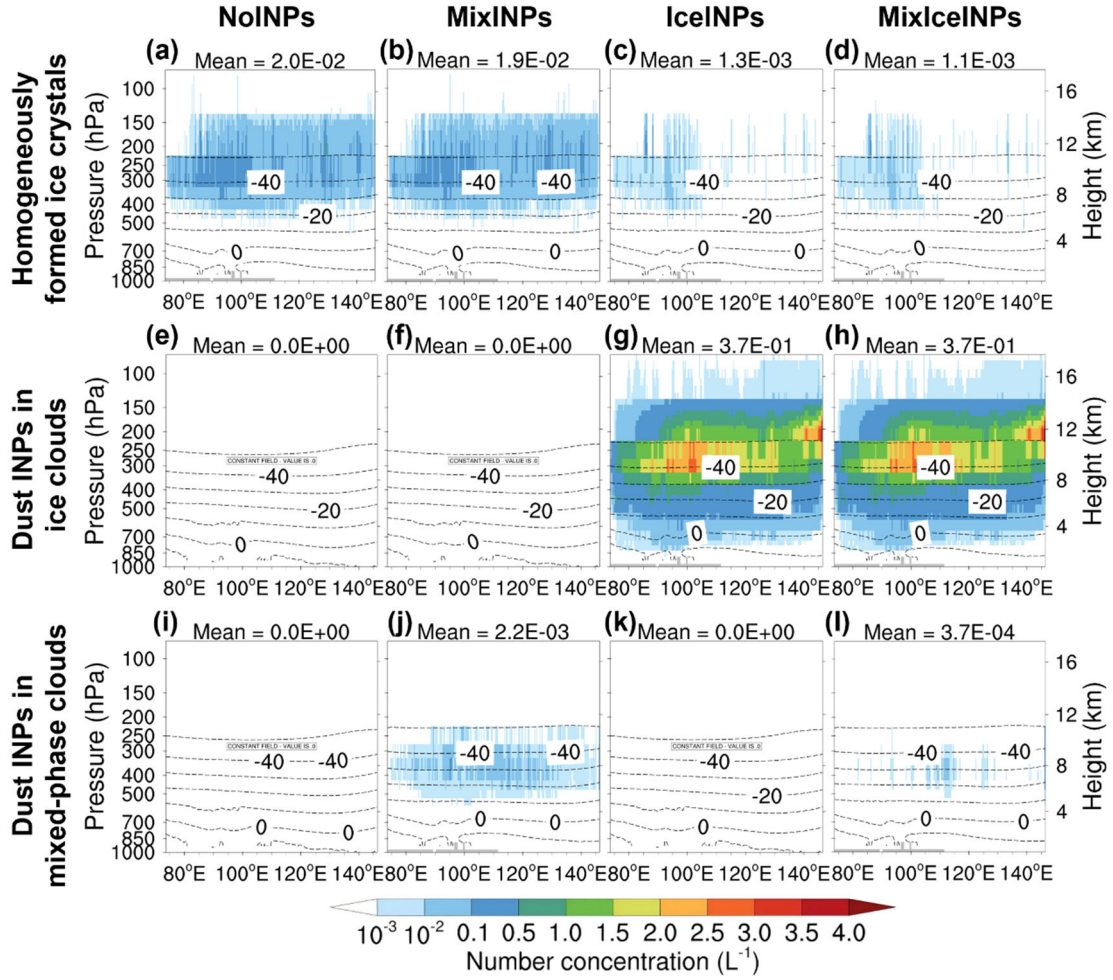


Figure S13. Simulated meridional mean vertical cross-sections of (a–d) homogeneously formed ice crystals, (e–h) dust INPs in ice clouds, and (i–l) dust INPs in mixed-phase clouds, for the NoINPs (first column), MixINPs (second column), IceINPs (third column), and MixIceINPs (fourth column) scenarios. Dashed lines indicate isotherms, and grey shading denotes missing values due to terrain. The mean value over the plotted region is shown above each panel. The simulated results are averages of hourly values from May 2018.

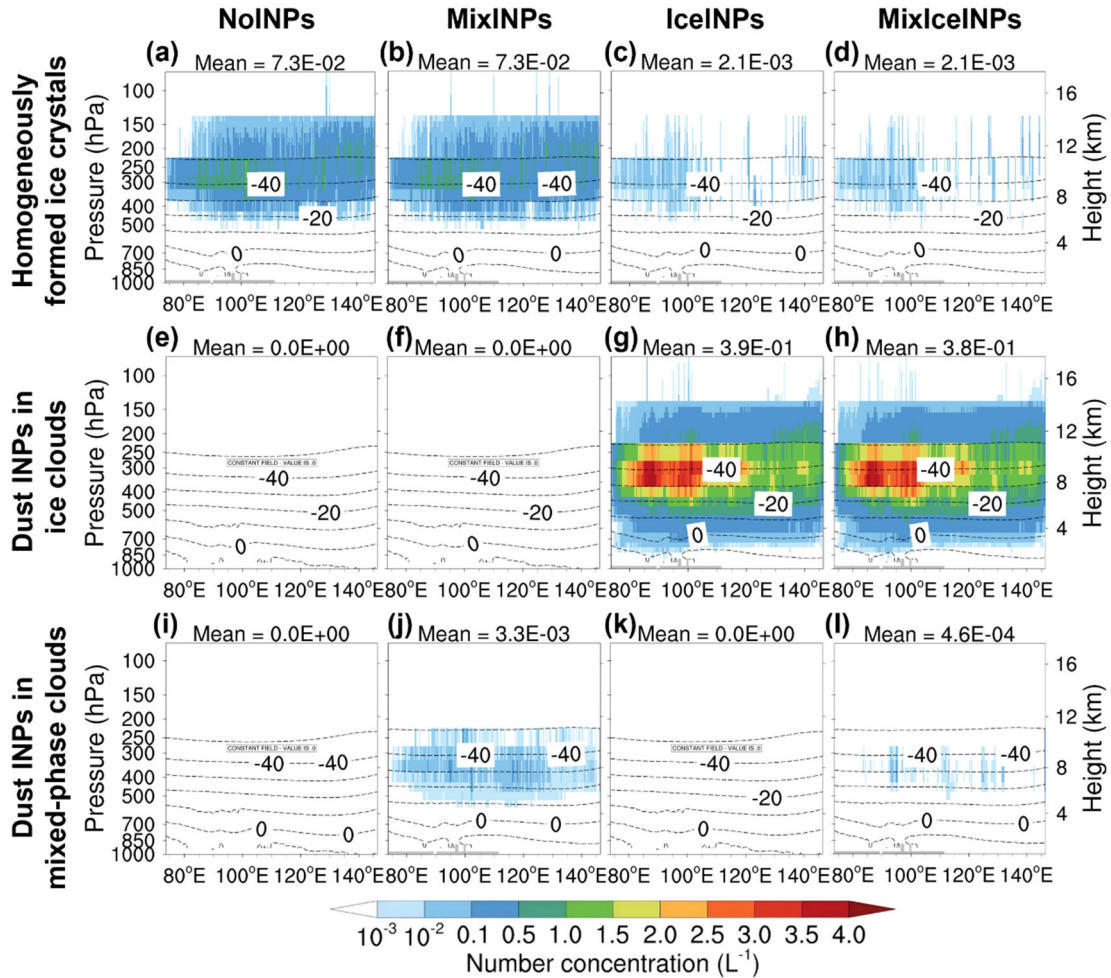


Figure S14. Same as Fig. S13, except that the subgrid vertical velocity is diagnosed using the turbulent-kinetic-energy-based parameterization, with stronger values.

Line 538: Is dust transported with cloud droplets in the convective parameterization?

Response: We thank the reviewer for this insightful question. In the WRF-Chem model (Grell et al., 2005), dust is treated as an aerosol tracer and is transported by convective mass fluxes when chemistry convective transport is activated. When convective wet scavenging is enabled, a fraction of dust is partitioned into convective cloud water, effectively coupling it to cloud droplets. This cloud-borne fraction can subsequently be transferred to precipitation and removed via wet scavenging. In our simulations, both processes are active, so dust is indirectly transported together with convective cloud droplets through the combined effect of chemistry convective transport and convective wet scavenging, even though it is not physically attached to individual cloud droplets.

Lines 600–602: Again, please ensure this result is not driven by weak subgrid-scale vertical velocities.

Response: We thank the reviewer for this helpful comment. We agree that it is important to ensure that the simulated increase in ice crystal number concentration is not an artifact of underestimated subgrid vertical velocities.

To address this concern, we performed a sensitivity experiment using an alternative subgrid vertical velocity parameterization based on turbulent kinetic energy (TKE), following Morrison and Pinto (2005). As discussed in our response above, this approach generally produces stronger vertical velocity fluctuations and has been shown in previous studies (Zhou et al., 2016) to potentially overestimate ice crystal number concentrations due to enhanced supersaturation.

The results of this sensitivity experiment, shown in Fig. S15 and Fig. S16 (shown below), indicate that the key features of our findings remain unchanged. In particular, the sharp increase in ice crystal number concentration in the IceINPs scenario persists, and the dominance of heterogeneous nucleation over homogeneous nucleation is still evident. This demonstrates that the simulated enhancement of ice crystal number concentration is not driven by weak subgrid vertical velocities.

Instead, the results are primarily controlled by the much higher concentrations of dust INPs relative to homogeneously nucleated ice crystals, which strongly favor heterogeneous nucleation. Therefore, the mechanism described in the manuscript remains robust.

We have added a sensitivity analysis and corresponding discussion (Section 3.2.5: Sensitivity to subgrid vertical velocity parameterization) in the revised manuscript (Lines 877–909) to clarify this point.

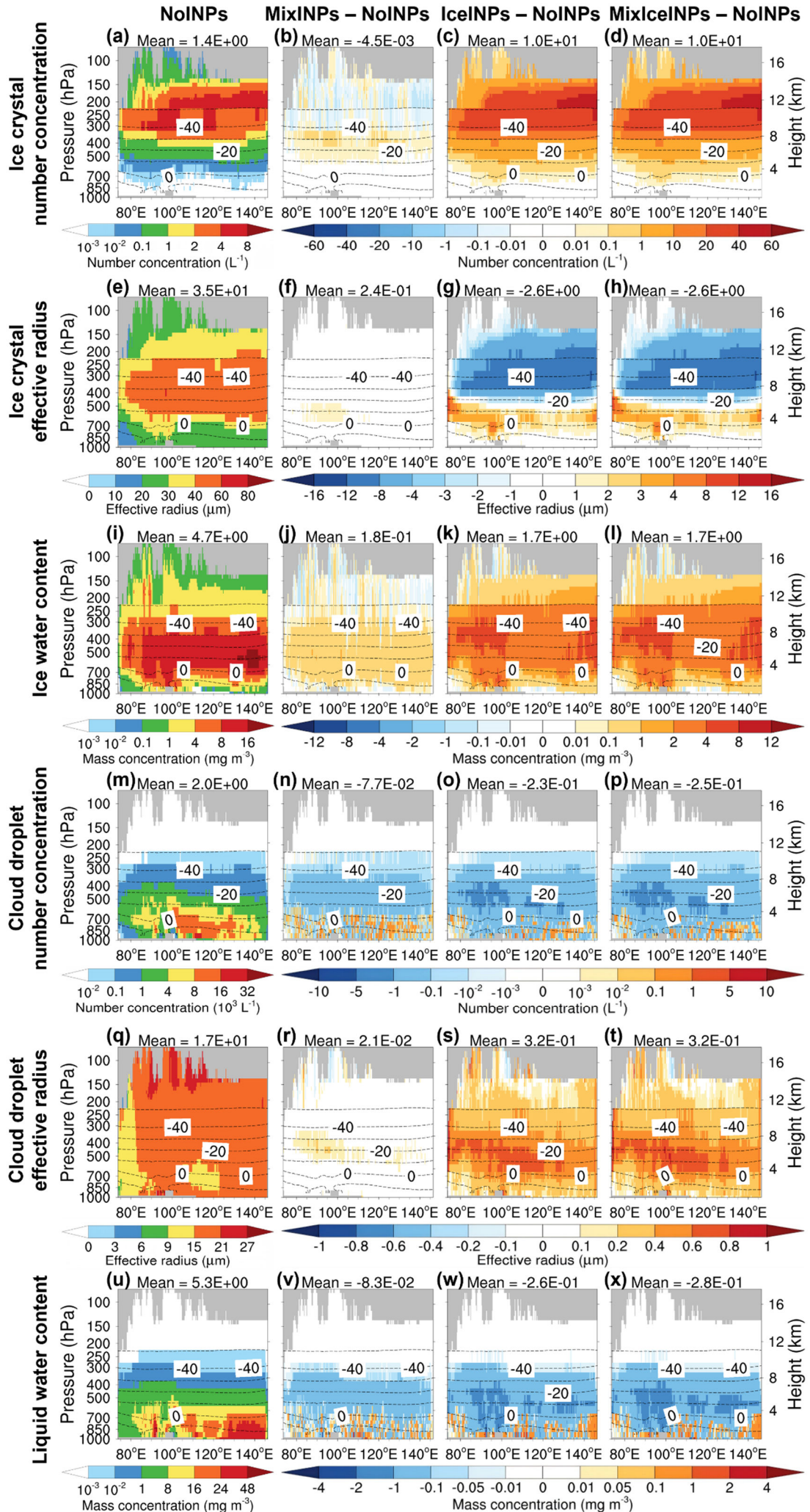


Figure S15. Simulated meridional mean vertical cross-sections of **(a–d)** ice crystal number concentration, **(e–h)** ice crystal effective radius, **(i–l)** ice water content, **(m–p)** cloud droplet number concentration, **(q–t)** cloud droplet effective radius, and **(u–x)** liquid water content, for the NoINPs (first column), MixINPs – NoINPs (second column), IceINPs – NoINPs (third column), and MixIceINPs – NoINPs (fourth column) scenarios. Dashed lines indicate isotherms, and grey shading denotes missing values due to terrain or filtering of non-cloud grid cells (where the sum of ice and liquid water mixing ratios is less than 10^{-6} kg kg⁻¹). The mean value over the plotted region is shown above each panel. The simulated results are averages of hourly values from May 2018.

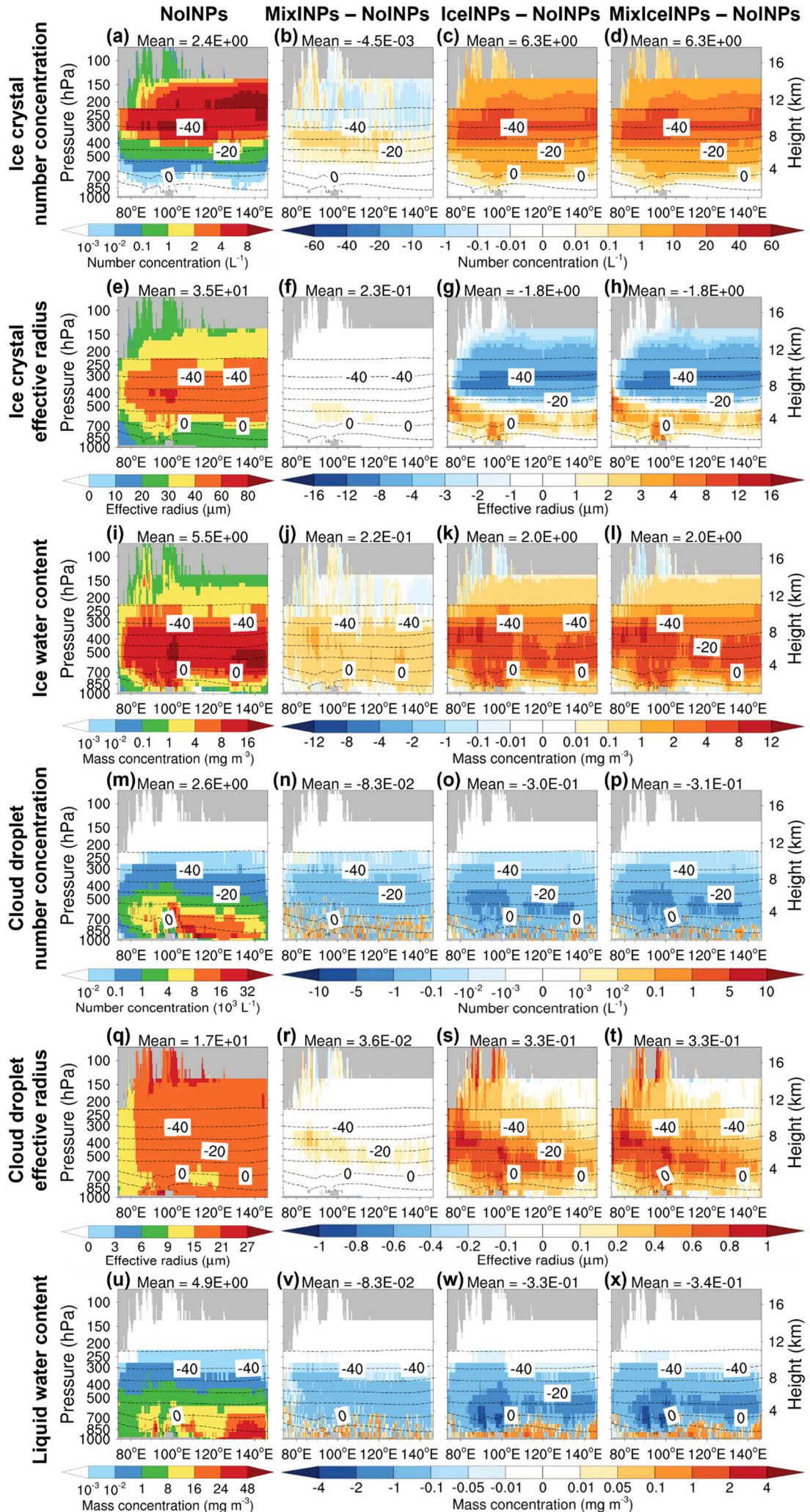


Figure S16. Same as Fig. S15, except that the subgrid vertical velocity is diagnosed using the turbulent-kinetic-energy-based parameterization, with stronger values.

Figure 10: The proposed mechanisms are largely speculative and should be supported by diagnosed WBF rates. The figure also seems misplaced in the conclusions.

Response: We thank the reviewer for this helpful comment. Detailed explanations and supporting figures have already been provided in the previous responses. Here we briefly summarize the key points. We agree that the mechanisms illustrated in Figure 10 should be supported by explicit diagnostics rather than remaining conceptual. Following this suggestion, we have added a dedicated analysis of the key microphysical processes in Sect. 3.2.3, including both the WBF process and ice crystal sedimentation.

For the WBF process, we diagnose cloud droplet evaporation and vapor deposition under WBF-favorable conditions. The results show a clear enhancement of both rates, providing direct evidence of an accelerated transfer of water from the liquid phase to the ice phase.

For sedimentation, we analyze the ice crystal sedimentation flux. The results indicate enhanced sedimentation from ice clouds, together with a weaker increase below, suggesting partial retention and accumulation of ice crystals within mixed-phase cloud layers. This supports the proposed role of sedimentation in increasing ice crystal number concentration and reinforcing the WBF process in mixed-phase clouds.

These diagnostics provide direct process-level evidence supporting the mechanisms summarized in Figure 10.

In addition, we agree that Fig. 10 was not optimally placed in the original manuscript. The figure has now been moved from the *Conclusions* section to the *Results and Discussion* section, where it is directly linked to the corresponding process-based analysis.

The manuscript has been revised accordingly to better integrate the figure with the supporting evidence and to ensure that the proposed mechanisms are clearly substantiated.

Code and data availability: At minimum, the source data for the figures should be made available. Please also indicate the sources of the model code and parameterizations, even if they were modified by the authors.

Response: We thank the reviewer for this important comment regarding code and data availability.

Following the suggestion, we have uploaded the source data used to generate all figures to a Zenodo repository and provided access via a preview link. The repository is currently in preview mode because the manuscript is still under review and the dataset may be updated in response to further reviewer comments. As Zenodo records cannot be modified after being made public, we defer final publication of the dataset until the manuscript is finalized. A permanent public link will be provided upon

acceptance of the manuscript.

Regarding the model, this study is based on the official release of WRF-Chem v4.2, which is publicly available from the WRF model repository. The model was further developed in this study as described in Sect. 2. The modified model code is available upon request from the corresponding author.

We have revised the “Code and data availability” section in the manuscript (Lines 1007–1019) to clarify both the current data access and the source of the model code.

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