Response to Referee #1

We thank reviewer 1 for thoughtful and constructive comments that helped us to improve the clarity and quality of our manuscript. Below we have provided a point-by-point response to each of the points raised by reviewer 1. The reviewer's comments are given in *italic black font* and our response in green with quotes in *italic font* style. The line numbers refer to the revised and highlighted manuscript file. Before addressing the specific points in detail, we would like to give a **general response** to comments given by both reviewers referring to the idealized nature of this study.

This work introduces the AC-1D model, which provides a novel framework to prognostically treat INP and ice crystal budgets while explicitly accounting for polydisperse aerosol inputs. A key capability of this model is the flexibility to apply different immersion freezing (IMF) parameterizations, including deterministic and time-dependent (CNT) schemes, to different aerosol types with unique particle size distributions.

By purpose, we chose to focus on INPs and ice crystal formation, and the role of supercooled water droplets in mixed-phase clouds is implicit to the model initialization of the thermodynamic state in the examined scenario. Cloud modeling has not yet been able to accurately determine the strength of the primary or secondary ice production (PIP or SIP) pathways. We believe this is in part due to the models' complexity trying to account for all different processes proceeding in the cloud. Here, we aim to assess the strength of the PIP in mixed-phase clouds since everything else follows from this. If we cannot constrain this aspect of the cloud sufficiently the remaining processes stay ambiguous.

During the development of this prognostic model, novel concepts and notions emerged including the concept of "activatable" INPs which reflect the maximum number of INPs available to form ice under given cloud conditions (not all activatable INPs necessarily form ice though) and the notion of an INP reservoir which is available to the cloud and, as we show, is defined by choice of freezing parameterization. This extends previous modeling studies.

In short, this bottom-up approach links specific polydisperse aerosol particle size distributions (PSDs) to their respective freezing behaviors, establishing the model as a robust testbed for investigating structural model uncertainties for user-provided cloud conditions (e.g., LES informed etc.). Specifically, the model architecture facilitates:

1. **Comprehensive prognostic treatment** of aerosol, INP, and ice crystal budgets, explicitly accounting for the loss of each property (e.g., via activation or sedimentation) to capture their temporal evolution.
2. **Flexible initialization** using realistic, polydisperse, and multi-component aerosol composition rather than simplified monodisperse inputs, as well as the ability to easily switch between prognostic and diagnostic modes or compare different freezing parameterizations simultaneously.

3. **Process-level diagnosis**, enabling the user to quantify detailed INP and ice crystal budgets (e.g., activation vs. sedimentation vs. entrainment) in response to user-defined perturbations in thermodynamic profiles, cooling rates, and microphysical parameters.

We developed this simplified model to isolate and quantify the structural uncertainty introduced by the choice of immersion freezing (IMF) parameterization. While we acknowledge that Arctic clouds are complex systems influenced by SIP and liquid-ice feedbacks (such as the Wegener-Bergeron-Findeisen process driving ice growth), including these processes in this initial study would obscure the primary signal we aim to investigate: the foundational uncertainty in ice crystal number concentrations due to different freezing parameterizations.

We make the following changes to the text to make these points clearer.

Abstract:

Line 15: We add the following statement:

"*We developed one-dimensional aerosol-cloud (AC-1D) model, which provides a novel framework to prognostically treat INP and ice crystal budgets while explicitly accounting for polydisperse and multicomponent aerosol that activate INPs following different freezing parameterizations.*"

We modified the text on line 155:

"Building on these identified uncertainties in primary ice production (PIP), we now focus in detail on the broader characteristics of Arctic aerosol. In this study, we employ the AC-1D model as a prognostic tool designed to isolate the structural uncertainties in PIP. The impact of liquid-ice feedbacks (such as the Wegener-Bergeron-Findeisen process driving ice growth) and secondary ice production (SIP) on the ice crystal budget depends crucially on the accurate description of the PIP. A key feature of this framework is its ability to conduct comprehensive sensitivity analyses by coupling polydisperse and multicomponent aerosol inputs directly to the INP and ice budgets. This setup allows for the simultaneous, prognostic evaluation of fundamentally distinct IMF parameterizations, while permitting user-defined adjustments to thermodynamic profiles and cloud system parameters. By generating detailed process-level data, such as explicit INP and ice crystal budgets, the model serves as a robust testbed to determine how the choice of parameterization dictates the PIP and evolution of the INP reservoir in Arctic mixed-phase clouds (Knopf et al., 2023; Arabas et al., 2025)."

We thank the reviewer for the constructive criticism of our work. The comments provided valuable perspective that helped us significantly improve the manuscript, particularly regarding the contextualization of our aerosol inputs and the comparison of our model results with observational INP data.

MAIN COMMENTS:

(A) The case study is constructed using thermodynamic measurements from SHEBA and aerosol inputs from ISDAC and ICEALOT campaigns. While ISDAC and ICEALOT occurred in spring, it is not clarified to which season the SHEBA case corresponds to. The Arctic aerosol composition exhibits seasonal and spatial variability with long-range transport of dust and anthropogenic aerosols peaking in late winter–spring ("Arctic haze") and marine organic/sea-spray sources dominating in summer. This seasonal variability is also reflected in the INP composition/origin (Creamean et al. 2022).
Here, the prescribed dust load appears low, yet multiple observational studies show that dust intrusions into the Arctic can be important during certain periods, often linked to springtime transport from Asian or Saharan sources. Similarly, sea-spray emissions depend on open-water fraction and wind-driven surface conditions, which vary seasonally and geographically. Therefore, a short discussion on how the chosen PSDs and thermodynamic properties align in season and location would be useful, along with clarifications on the Arctic conditions that are represented by this case.

We appreciate the reviewer's insight regarding the seasonal and spatial variability of Arctic aerosol composition.

We have rewritten the text in Section 2.2 (Lines 245-274 of the original manuscript) to explicitly justify the composite aerosol initialization based on microphysical consistency with the SHEBA case while referring to the reviewer comment details. The revised sections now read:

"To evaluate the impact of different aerosol types on the INP reservoir, N_i and ice crystal formation rate, we examine the effects of mineral dust, organic, and SSA particles. While the thermodynamic profile is derived from the specific SHEBA case study (early May) to represent a typical Arctic mixed-phase boundary layer, the aerosol initialization represents a composite of spring Arctic conditions derived from ISDAC and ICEALOT (April).

Since size-resolved aerosol measurements were not available for the specific SHEBA case study, we approximated the total aerosol inputs based on the observed cloud droplet number concentration ($N_d \approx 200\ cm^{-3}$; Fridlind et al., 2012). Our model assumes that all in-cloud aerosols are activated.

To reconstruct a physically consistent aerosol population, we utilized data from the ISDAC campaign (for mineral dust and organic fractions) and the ICEALOT campaign (for SSA). The

ISDAC campaign is an appropriate proxy because it exhibited cloud microphysical properties very similar to SHEBA case, with measured N_d values ranging from 185 to 205 cm^{-3} (Savre et al., 2015).

To achieve quantitative alignment between the prescribed aerosol load and the SHEBA target ($N_d \approx 200\ cm^{-3}$), we selected aerosol inputs corresponding to the "clean case" defined in Earle et al. (2011). This case shows number concentrations of aerosol (N_{aer}) less than 250 cm^{-3}, while N_d is approximately $135 \pm 34\ cm^{-3}$ (Earle et al., 2011). Critically, this case exhibits a high activation efficiency, validating our model assumption that all in-cloud aerosols are activated .Within this total number constraint, the relative compositional fractions of mineral dust and organic aerosol were derived from single particle analyses performed during the same field campaign (Hiranuma et al., 2013). Although the PSDs are derived from the subsequent flight (Flight 31, April 27), Earle et al. (2011) classify both dates as the same meteorological regime, justifying this composite initialization.

While mineral dust is a major component of Arctic INPs during long-range transport events (Creamean et al., 2022; Böö et al., 2023), the background conditions defined for this specific sensitivity study maintain low dust number concentration compared to organic and SSA particles to match the microphysical constraints of the SHEBA case. High-load scenarios, e.g., increase in dust load, are explored separately in sensitivity runs. Similarly, while the SHEBA case occurred over pack ice where local SSA emission is suppressed, we include the ICEALOT SSA distribution (Quinn et al., 2017) to account for potential transport from open water leads and to establish a robust background state for testing competitive nucleation processes.

For each aerosol type, the applied aerosol particle size distributions (PSDs) are polydisperse consisting of two or three lognormal modes (Table 1). In addition to Aitken and accumulation modes, this framework includes a larger accumulation mode for aged aerosols and a source-specific SSA mode. Lastly, to reflect a more realistic aerosol population we combine the mineral dust, organic, and SSA PSD (composite PSD). Figure 1 displays the lognormal PSDs of the different aerosol particle types and the composite PSD, derived from the modal parameters specified in Table 1."

(B) The authors mainly show results related to the activatable INPs and ice crystal number. I think it would be very useful to show results related to activated INPs and compare to the vast literature that has been recently published on Arctic INPs (e.g. Wex et al. 2019, Creamean et al. 2022, etc). I suspect that the CNT formulations might predict INP concentrations that are outside (above) the observed range, suggesting that these two parameterizations are not suitable for Arctic conditions.

We appreciate this excellent suggestion. To address this, we have added a new figure (Fig. 3) to the beginning of Section 3.1, comparing our model's predicted INP concentration against a comprehensive set of recent Arctic field observations.

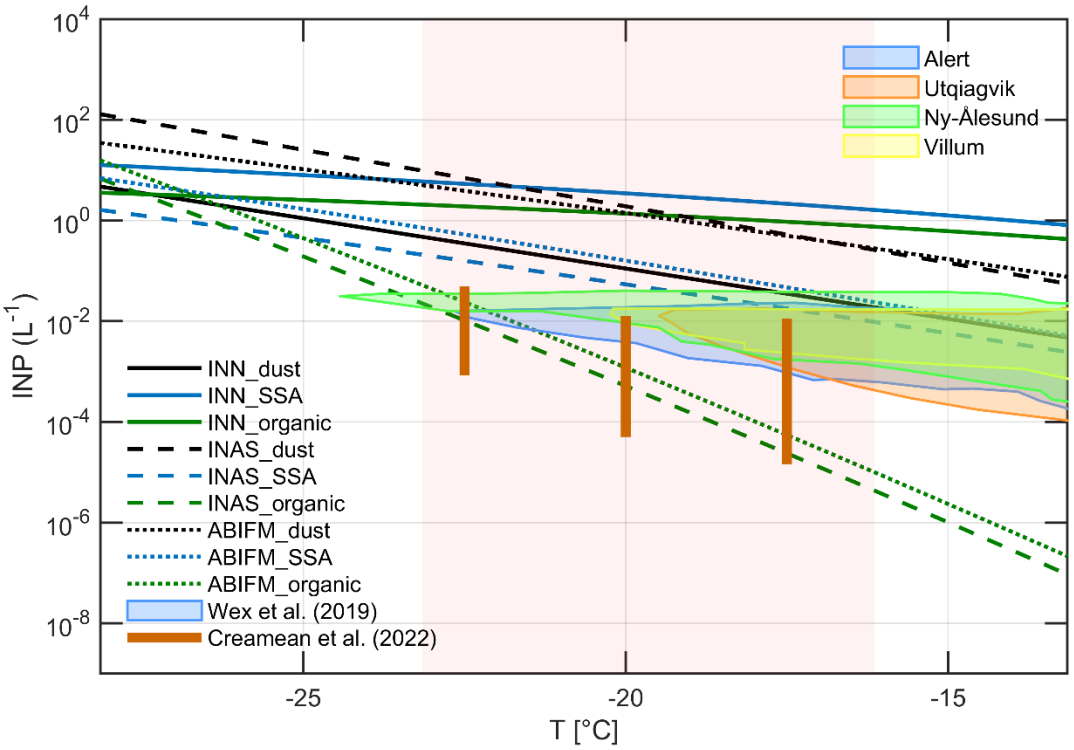

Figure 3. Predicted activated INP concentrations from immersion freezing parameterizations are shown for mineral dust (black lines), SSA (blue lines), and organic (green lines) aerosols, calculated with their respective PSD from Table 1. Lines indicate the different schemes: INN (solid), INAS (dashed), and ABIFM (dotted). Note that the INN schemes use DeMott et al. (2015) for mineral dust and DeMott et al. (2010) for SSA and organic aerosol particles. Model predictions are compared against a composite of Arctic field observations from Wex et al. (2019) (colored shaded regions reflect Alert, Utqiaġvik, Ny-Ålesund, Villum) and Creamean et al. (2022) (vertical orange bars). The vertical pink background shading indicates the temperature range in the simulation domain.

We have inserted the following text to validate the parameterizations against observations before discussing the simulation results at line 492:

"As a crucial step for model evaluation, we first place the chosen IMF parameterizations into an observational context. This provides a baseline for interpreting the prognostic simulations that follow. To this end, we compare the activated INP concentrations predicted by each IMF parameterization (initialized with the specific aerosol PSDs from Table 1) against recent Arctic field observations.

Figure 3 compares these model predictions against a composite of Arctic data. The shaded regions represent the annual variability observed at four ground-based Arctic stations (Alert, Utqiaġvik, Ny-Ålesund, and Villum Research Station) utilizing filter samples for ice nucleation

experiments analyzed with a cooling rate of 1 °C min⁻¹ (Wex et al., 2019). Additionally, we show INP measurements recorded during the Multidisciplinary drifting Observatory for the Study of Arctic Climate (MOSAiC) expedition onboard the research vessel Polarstern (Creamean et al., 2022). These particle samples were analyzed using a cooling rate of 0.33 °C min⁻¹. The vertical bars indicate the minimum and maximum INP concentrations observed during the full annual cycle at -17.5, -20, and -22.5 °C, selected to match the temperature range of our simulated cloud layer. For the time-dependent ABIFM (CNT) parameterization, a nucleation time period is required to derive the cumulative activated INP number concentration. We apply $t = 1$ min (following Alpert et al. (2022)) to approximate the nucleation time scales for the experiments.

As shown in Figure 3, the agreement depends on both the parameterization choice and the assumed aerosol type. The deterministic INAS scheme, when applied to SSA and organic aerosol PSDs, shows good agreement within the range of field observations. The ABIFM (CNT) predictions for organic aerosol also align well with the annual range observed during the MOSAiC campaign. While the deterministic INN scheme and dust parameterizations tend to predict concentrations at the upper end of the observational range, they generally fall within the total variability spanning the different stations and seasons. Except for a pure dust case, INAS and CNT parameterizations predict INP concentrations similar to the ones observed in the Arctic regions."

[The original text starting at Line 521: *"The choice of IMF parameterization fundamentally dictates..."* follows here.]

(C) I think that the highly idealized nature of these simulations is underdiscussed. Mixed-phase microphysical processes are complex and the impact of processes like WBF, riming and aggregation can largely affect the ice crystal size distribution and eventually the available ice crystal budget. Taking into account these processes could likely change the relative contribution of each IMF parameterization to the ice number and affect their interactions with other processes, like sedimentation. Moreover, there is increasing evidence that secondary ice production (SIP) is important in Arctic

We acknowledge that Arctic mixed-phase clouds are complex systems where processes such as the Wegener-Bergeron-Findeisen process, riming, and SIP can play significant roles. However, the simplification of the AC-1D model, specifically the exclusion of liquid-phase feedbacks and SIP, is an intended design choice to isolate the role of PIP. Please see also our general response above.

The fundamental objective of this work is to quantify the uncertainty in INP and ice crystal budgets arising directly from the choice of immersion freezing parameterization. If the representation of PIP remains uncertain, the simulation of secondary processes that depend on this primary ice become highly speculative.

Therefore, this study provides a necessary bottom-up investigation, treating INPs prognostically based on the polydisperse aerosol population. This allows us to establish a baseline of uncertainty for PIP before adding the complexity of secondary processes.

It is important to note that not every mixed-phase cloud is prone to SIP as discussed in Fridlind et al. (2018) (chapter7, section 2.5 and 5.3). For example, a 6-year ground-based remote sensing study in the Arctic found that SIP events occurred in less than 10% of observed slightly supercooled clouds (temperatures > -10°C), even though this temperature range is considered optimal for the Hallett-Mossop rime splintering process (Luke et al., 2021). For the specific SHEBA case, observations indicated a low liquid water path, no precipitation, and sparse unrimed ice crystals (Fridlind et al., 2012), conditions under which SIP and riming were not dominant factors. We have expanded the Introduction and Discussion sections to explicitly frame this model as a robust prognostic framework designed to disentangle the fundamental structural uncertainties of primary ice formation. By offering a transparent environment to track the INP budget, this tool serves as an essential prerequisite step to more complex microphysical simulations.

We have revised the manuscript in three specific locations to explicitly frame the model as a prognostic testbed for PIP and to justify the exclusion of secondary processes for this specific case study.

First, in the Introduction (Line 155 at Page 5), we revised the text:

“Building on these identified uncertainties in primary ice production (PIP), we now focus in detail on the broader characteristics of Arctic aerosol. In this study, we employ the AC-1D model as a prognostic tool designed to isolate the structural uncertainties in PIP. The impact of liquid-ice feedbacks (such as the Wegener-Bergeron-Findeisen process driving ice growth) and secondary ice production (SIP) on the ice crystal budget depends crucially on the accurate description of the PIP. A key feature of this framework is its ability to conduct comprehensive sensitivity analyses by coupling polydisperse and multicomponent aerosol inputs directly to the INP and ice budgets. This setup allows for the simultaneous, prognostic evaluation of fundamentally distinct IMF parameterizations, while permitting user-defined adjustments to thermodynamic profiles and cloud system parameters. By generating detailed process-level data, such as explicit INP and ice crystal budgets, the model serves as a robust testbed to determine how the choice of parameterization dictates the PIP and evolution of the INP reservoir in Arctic mixed-phase clouds (Knopf et al., 2023; Arabas et al., 2025).”

Second, in the methods section (Section 2.3), We added the following paragraph at lines 331:

"In this setup, PIP and ice crystal sedimentation are the dominant ice microphysical processes tracked. Processes such as the Wegener-Bergeron-Findeisen (WBF) process, riming, and SIP are deliberately excluded to isolate the uncertainty in INP and ice crystal budgets arising directly from the choice of the immersion freezing parameterization."

Third, we added the following text at the end of section 4.1 (line 924):

"Finally, the distinct ice crystal evolution trends observed here highlight the sensitivity of the system to the representation of PIP. By disentangling this process from liquid-phase feedbacks and SIP, we demonstrate that the choice of freezing parameterization constitutes a foundational source of uncertainty in the simulated ice crystal budget."

MINOR COMMENTS:

Line 185: you probably refer to Figure 2. What is the liquid water path range?

We have corrected the reference to Figure 2. We have also added the liquid water path range observed for this case, which is approximately 5–10 g m^{-2} (Fridlind et al., 2012), to Section 2.1 (Line 214).

Line 216: Here it is mentioned that SSA PSDs are based on measurements above sea, while the SHEBA case corresponds to pack-ice conditions (see main comment A)

We acknowledge the distinction pointed out by the reviewer. As discussed in detail in our response to Main Comment A, the SHEBA case indeed occurred over pack ice where local sea spray emission is suppressed. However, we intentionally included the ICEALOT SSA distribution to represent a "complete" Arctic background state that accounts for potential transport from open water leads or the marginal ice zone. This allows the model to serve as a robust testbed for competitive nucleation.

Please refer to Response to Comment A for the full revision of Section 2.2. Specifically addressing this concern, we have added the following clarification to the text in Section 2.2 (Line at 267):

"Similarly, while the SHEBA case occurred over pack ice where local SSA emission is suppressed, we include the ICEALOT SSA distribution (Quinn et al., 2017) to account for potential transport from open water leads and to establish a robust background state for testing competitive nucleation processes."

Line 218: composite. Figure 1 should be 2

Corrected. We also corrected the figure numbering. To ensure figures appear in the order they are mentioned in the text, we have renumbered the first two figures. The thermodynamic profiles figure (discussed in Section 2.1) is now Figure 1, and the figure showing the aerosol particle size distributions (discussed in Section 2.2) is now Figure 2. The text was updated to reflect changes.

Section 2.3: Are primary ice production and sedimentation the only microphysical processes accounted in the model? This should clearly stated that other important processes are ignored (e.g. WBF, riming, etc) Also are there any aerosol processes accounted for?

That is correct. We have rewritten the text in Section 2.3 (Lines 331–336) to clearly define the active and excluded processes.

"In this setup, PIP and ice crystal sedimentation are the dominant ice microphysical processes tracked. Processes such as the Wegener-Bergeron-Findeisen (WBF) process, riming, and SIP are deliberately excluded to isolate the uncertainty in INP and ice crystal budgets arising directly from the choice of the immersion freezing parameterization. Regarding aerosol physics, the model treats the aerosol population prognostically, explicitly accounting for the depletion of INPs via activation and changes due to transport. However, internal aerosol microphysical processes (e.g., coagulation, condensation growth) are not simulated."

Line 278: Could you provide references for dust being negligible in the Arctic? There are many studies that do not support this claim (e.g. Boo et al. 2023; Creamean et al 2022)

We have revised this statement to avoid generalization. We do not claim dust is universally negligible in the Arctic; rather, we clarify that in the specific background PSDs selected for this sensitivity study as given in response to general comments A and B, mineral dust number concentrations are low compared to organic and sea spray aerosols. This was based on the study by Earle et al. (2011)) where dust number concentrations are low compared to organic and SSA particles. We have added references (including Creamean et al. (2022)) to acknowledge that dust can be a major component during transport events, but maintaining the low-dust case to align with droplet number concentration of the SHEBA case study.

Section 2.4: The description of sensitivity simulations is a bit confusing. E.g. CCR=0.3 is listed as sensitivity test, while based on the caption of figure 2, I would assume that the same CCR is applied in the CTRL simulation. If CCR is zero in CTRL simulation then this should be listen Table 4. If green profiles in Figure 2 concern only the sensitivity test and not CTRL case, this should be clearly explained in the caption

We have clarified the description in Table 4 and Section 2.4. The CTRL run has a Cloud Cooling Rate (CCR) of 0. The CCR = 0.3 case is a specific sensitivity experiment. The caption for Figure

1 has been updated to explicitly state that the green profiles represent the evolved conditions under the specific sensitivity test (CCR = 0.3) after 10 hours, while the blue profiles represent the initial/CTRL conditions:

Table 4: Parameter choices of the different aerosol-cloud 1D model simulation. Imm_CTRL: The CTRL run with the baseline settings for all IMF parameterizations, no perturbations, used as a reference. h_res_t and h_res_z: Simulation applying higher resolution with doubly refined vertical resolution (5 m) and much smaller time step (1 s).

Figure 1: Thermodynamic conditions applied in the minimalistic 1D aerosol-cloud model. From left to right: The temperature (T), the relative humidity (RH), supersaturation with respect to the ice (S_{ice}) and Δa_w. Blue and green lines represent the initial (t = 0 h) thermodynamic conditions and the thermodynamic conditions with cloud cooling rate of 0.3 °C h^{-1} (CCR = 0.3 °C h^{-1} sensitivity run) after 10 h, respectively. The blue shaded area denotes the cloud layer.

Line 285-287: Also it should be explicitly stated that CTRL simulation is run with a single aerosol type

We have revised the text in Section 2.4 (Lines 446–449) to clarify this point:
"We examine various simulation setups as given in Table 4. The thermodynamic conditions and cloud parameters from the LES baseline results of the SHEBA case study serve as the control run (hereafter referred to as CTRL) while applying the three different aerosol PSDs individually. Note that the baseline CTRL simulations are initialized with single aerosol types to isolate their specific freezing behaviors."

Line 308: also shown in Figure 2(?)
The reviewer is correct. We have revised the text in Section 2.4 (Page 16, Line 473) to include this reference (note the change in figure number): *"The evolution of the temperature profiles for CTRL, and CCR = 0.3 are presented in Figure 1 and Fig. S2."*

Line 310: clarify that each PSD corresponds to different aerosol type
We have revised the text in Section 2.4 (Page 16, Lines 471–472 of the original manuscript) to read:
"This results in 24 cases consisting of three aerosol PSDs (mineral dust, organic, and SSA respectively), four freezing parameterizations and two sets of cases..."

Line 503: Why organic INPs are more sensitive to temperature changes?

We have added an explanation to Section 3.3.1 (Page 28, Lines 681–683) to clarify this mechanism:
"...especially for organic aerosols which exhibit high sensitivity of N_{INP} to temperature changes. This heightened sensitivity arises because the INAS density formulation for organic particles

(China et al., 2017) exhibits a significantly steeper slope with respect to temperature compared to the mineral dust parameterization in this temperature range."

Line 576: black instead of brown

We have corrected the color descriptions in the text and captions to accurately match the figures (Page31, Line 756): *"applying dust, organic and SSA particles, given as brown, green, and blue lines, respectively."*

Fig 7 is confusing. There is a light solid brown line in panels a and b, not include in the legend. Also dashed blue line is not visible in panel a

We have redesigned Figure 7 (now Fig. 8) to improve legibility. We ensured that all plotted lines correspond to the legend, and improved the visibility of the dashed lines (representing entrainment).

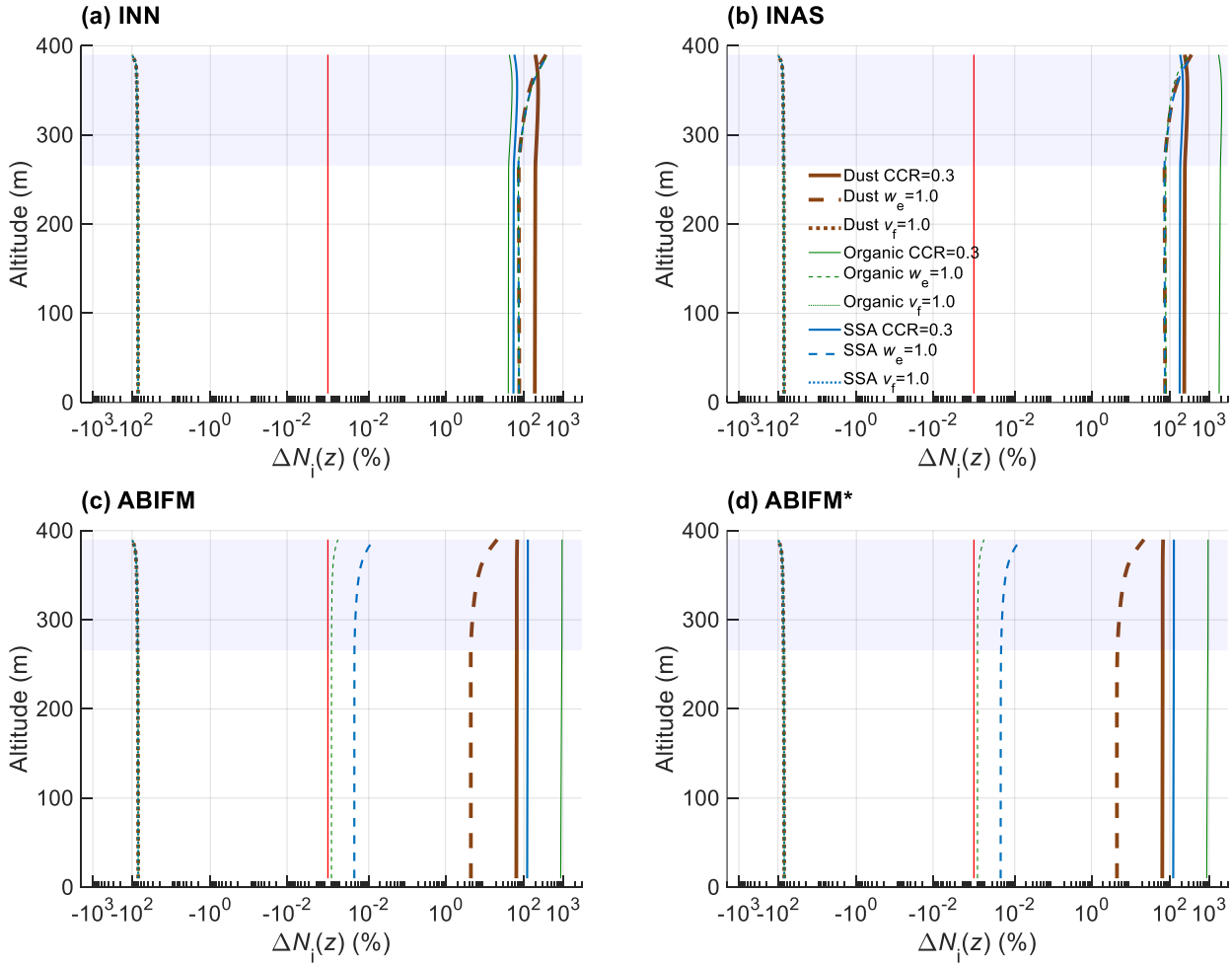

Figure 8: Vertical profiles of the change in number concentration of activatable INP ($\Delta N_{INP}(z)$ in %) averaged over entire 10 h of simulation time. ΔN_{INP} differs compared to the respective CTRL runs due to the change of cloud parameters (cloud cooling rate, cloud-top entrainment rate) applying dust, organic and SSA particles, given as black, green, and blue lines, respectively. Different immersion freezing parameterizations are applied including (a) ice nucleation number based (INN),

(b) ice-nucleation active sites (INAS), (c) water-activity based immersion freezing model (ABIFM), and (d) ABIFM enabling subsaturated freezing (d) ABIFM*. Simulation results for changing cloud cooling rate (solid lines) and cloud-top entrainment rate (dashed lines) are shown. The blue shaded area denotes the cloud layer and the red line in the middle highlights the value of 0.

Fig 8 is confusing. Is it the logarithmic scale that inhibits the demonstration of the whole vertical profile?

Figure 8, now Fig. 9 in revised manuscript, shows ice crystal formation rate which can only proceed in the cloud layer when assuming saturated conditions. Except for panel (d) where freezing is allowed to proceed at slightly subsaturated conditions, i.e., from diluted aqueous solutions, a formation rate exists for below the cloud layer.

To prevent confusion, we have updated the figure caption to explicitly state this physical constraint. (Note: We have also updated the figure to make it clearer as discussed in the previous comment).

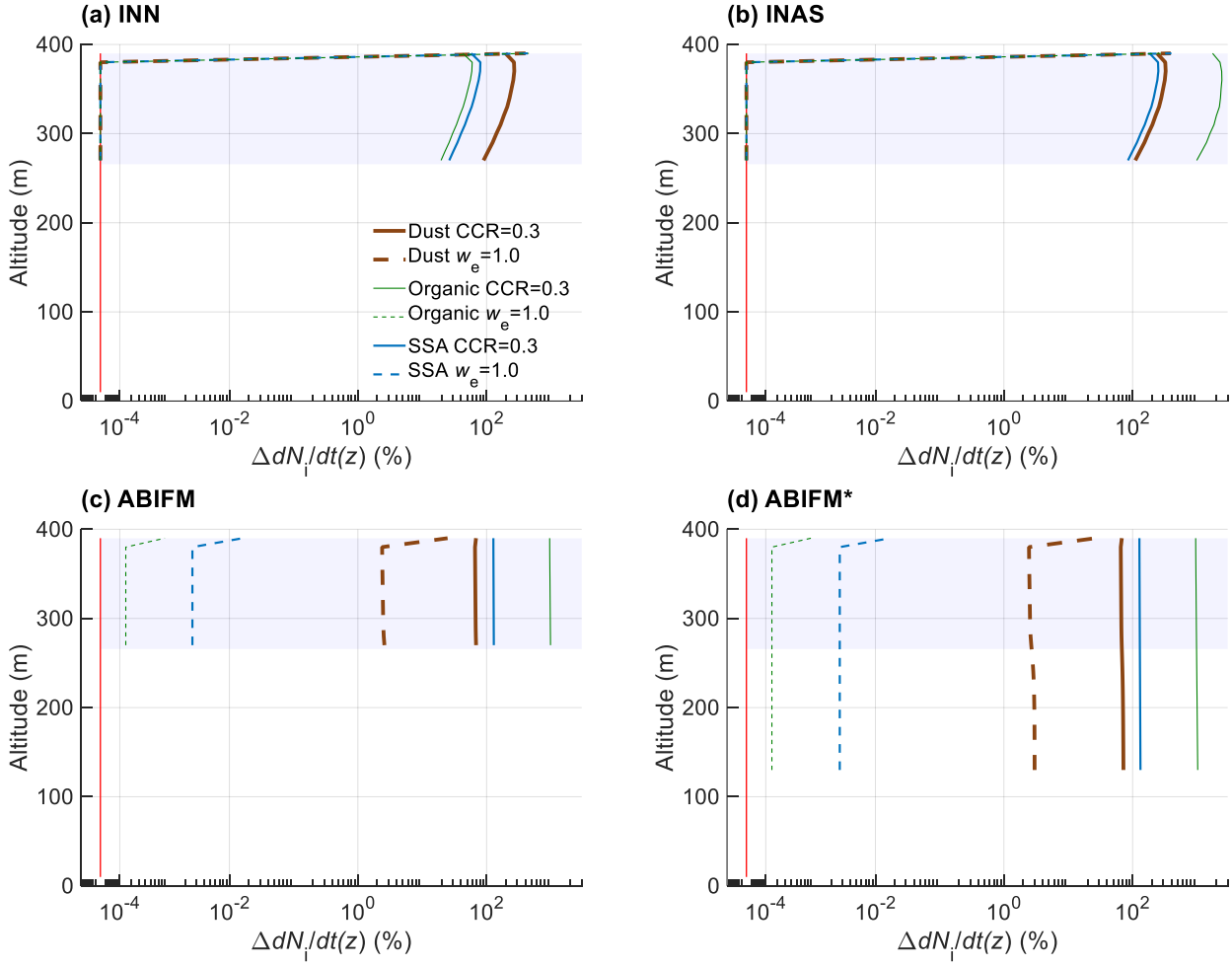

Figure 9: As in Figure 7 but for the change in ice crystal formation rate ($\Delta dN_i/dt(z)$).

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