Response to Reviewer 3

We thank the anonymous reviewer for his/her careful reading and constructive review of our paper. Our detailed responses to the comments follow. Reviewer's comments are in blue color, our responses are in black color, and our corresponding revisions in the manuscript are in red color.

Review for "Relative importance and interactions of primary and secondary ice production in the Arctic mixed-phase clouds" by Zhao & Liu

This manuscript compares the impacts of primary ice production (PIP) and secondary ice production (SIP) as well as their interactions on the simulation of multiple Arctic mixed-phase cloud microphysical and macrophysical properties observed during the M-PACE field campaign. The authors design a set of 10 simulations, 5 of which differ only in their treatment of ice nucleation schemes and the other 5 which utilize the same 5 aforementioned ice nucleation schemes but with representations of SIP via the ice-ice collisional breakup (IC) and rain droplet fragmentation (FR) mechanisms in addition to the Hallett-Mossop process which is represented in all 10 simulations. The authors find that 3 of the ice nucleation schemes that are aerosol-aware (CNT, N12 and D15) exhibit similar behaviour to each other in terms of their simulated ice crystals number concentration vertical profiles, supercooled liquid fraction (SLF), IWP, LWP and relative contributions from primary and SIP rates to the total ice production rate. They also find that these variables are also similar to each other for the other two ice nucleation schemes (B53 and M92). One of the main is that PIP and SIP actively influence each other. The authors also conclude that the aerosol-aware ice nucleation schemes with the IC and FR mechanisms represented best represent the single-layer mixed-phase clouds observed during M-PACE.

This is an interesting and valuable study at the forefront of effort to improve cold cloud microphysics in global climate models and their impact on cloud properties. There are however, a number of ways that the manuscript can be improved, particularly pertaining to the writing including the description of the model used and the experimental design, description of the observations and grammar. Overall, I recommend major revisions that are provided below.

Reply: We thank the reviewer for the positive comments. We have revised the manuscript following your comments regarding the writing including the description of the model used, the experimental design, and the observations to improve the quality of our paper.

Major revisions:

- The title is wordy and unclear. Perhaps revise to something like "primary and secondary ice production: interactions and their relative importance"?

Reply: We thank the reviewer for the suggestion. We changed the title as: "Primary and Secondary Ice Production: Interactions and Their Relative Importance" as the reviewer suggested.

- An interesting conclusion of this manuscript is the interaction between SIP and PIP which compete with one another. The suppression of SIP via PIP is intuitive, however, the suppression of PIP via SIP is less intuitive since one would initially expect that more ice crystals nucleated via PIP would allow more SIP. The explanation for the latter phenomenon provided in the manuscript relates to the lack of precipitation particles in B53 and M92 due to the enhanced glaciation of mixed-phase clouds. A description of the graupel scheme (which seems to be diagnostic based on line 364) the authors implemented would help the readers more clearly understand the mechanism instead of referring to Zhao et al. 2021. The mechanism of SIP and PIP suppression could also be summarized in the Abstract. Also, the discussion on lines 73-78 in the Introduction can also be elaborated on in this aspect when describing the work of Phillips et al. 2017b.

Reply: We thank the reviewer for the suggestion. The same as the reviewer, we initially expected that stronger PIP would allow more SIP. However, the model shows the suppression of SIP via PIP due to complex interactions between cloud microphysics processes resulting in the reduction of precipitation particles (rain and graupel).

Following the reviewer's comment, we added a description of the graupel scheme as:
"The graupel mass mixing ratio (q_g) is diagnosed as precipitation ice mass (currently snow, q_s) multiplied by the rimed mass fraction Ri (Zhao et al., 2017),

$$q_g = q_s \times Ri \tag{6}$$

The rimed mass fraction Ri is calculated as:

$$Ri = \frac{m_{rimed}}{m_{rimed}+m_{unrimed}} \approx \frac{1}{1+\frac{6\times10^{-5}}{q_c(q_i+q_s)^{0.17}}} \tag{7}$$

q_c, q_i, and q_s in (7) are modeled cloud water, cloud ice, and snow mixing ratio (kg kg^{-1}), respectively. The graupel number is assumed to have the same ratio to snow number as the ratio of graupel mass to snow mass."

We have added the mechanism of SIP and PIP suppression in the abstract: "SIP is not only a result of ice crystals produced from ice nucleation, but also competes with the ice nucleation by reducing the number concentrations of cloud droplets and cloud-borne dust INPs. Conversely, strong ice nucleation also suppresses SIP by glaciating mixed-phase clouds and thereby reducing the amount of precipitation particles (rain and graupel)."

- An 80% contribution of SIP to total ice formation seems very large. Are these any observations to gauge how realistic this value is? Similarly, on lines 297-301, are there any observations to gauge how realistic these contributions are? Otherwise, this should be declared in the main text.

Reply: We thank the reviewer for the comment. We agree with the reviewer that an 80% contribution of SIP seems a large fraction. So far, we do not have observations to directly verify the contribution of SIP to total ice formation. However, observations have reported that ice crystal number concentrations are often a few orders of magnitude higher than INP number concentrations, as we discussed in the abstract. A recent study by Luke et al. (2021; PNAS) found that "the occurrence frequency of secondary ice events averaging to <10% over the 6 years ground-based radar measurements in the Arctic, but SIP has a significant impact in a local region when they do occur, with up to a 1,000-fold enhancement in ice number concentration." In our study, we compare observed INP number concentrations with observed ice number concentrations to identify the SIP process, as shown in Figure 3. We note that ice number concentrations are three orders of magnitude higher than INP number concentrations from the model simulations, and are two orders of magnitude higher from the observation, suggesting the dominant contribution of SIP to total ice formation.

We have added a declaration in Section 5 (Summary and conclusions) as: "More observation data are needed to identify the frequencies and conditions of SIP occurrence in cold clouds and its contribution to total ice formation so that the impact of SIP can be better quantified by the models."

- In addition to the graupel implementation mentioned above, the description of the ice nucleation schemes could also be described in more detail. All ice nucleation schemes appear to be implemented as immersion freezing schemes --- please confirm. How are deposition, condensation, and contact freezing represented? To be consistent with the other naming conventions used in the manuscript, I would also recommend changing "CNT" scheme to reflect the reference that was used (was it Wang et al. 2014 or Hoose et al. 2010)? The description of this scheme also does not include the equation and the units of all equations that are provided are missing. For N12, is the dry diameter of dust particles predicted by MAM4? For the D15 scheme, please include more information on the instruments that were used for the measurements and the location where the observations were taken from. To be clear, are marine organic aerosols and sea salt not included as INPs in any of the parameterizations? Please include in the description.

Reply: We thank the reviewer for the suggestions. The CNT scheme represents immersion, contact, and deposition nucleation separately with different equations. With many equations involved in the CNT scheme, we prefer not to include them in the paper, but refer the readers to Wang et al. (2014) and Hoose et al. (2010). The CNT scheme is formulated based on Hoose et al. (2010) and implemented in CAM5 by Wang et al. (2014) with further improvements of using a PDF of contact angle instead of a single contact angle in Hoose et al. (2010). We prefer keeping the name "CNT" in the paper since it is called in our previous studies (Shi and Liu, 2019; Zhao et al., 2021).

We have modified the sentence as: "CNT is formulated based on Hoose et al. (2010) and implemented in CAM by Wang et al. (2014) with further improvements of using a probability density functions (PDF) of contact angle instead of a single contact angle in Hoose et al. (2010)."

The N12, D15, B53, and M92 are empirical schemes for the immersion freezing of cloud droplets. Thus, for the D15 and N12 experiments, the deposition and contact ice nucleation are still represented by the CNT scheme. For the B53 and M92 experiments, the deposition ice nucleation is represented by M92 and the contact ice nucleation by the Young (1974) scheme. We understand that there is an inconsistency in the representation of deposition and contact ice nucleation in these experiments. However, for the single-layer mixed-phase clouds, immersion freezing is dominated, and the contributions from deposition and contact ice nucleation to total ice production are much smaller (Figure 9).

We have provided a more detailed description in section 3: "The N12, D15, B53, and M92 experiments are the same as the CNT experiment except using the respective ice nucleation scheme to replace the CNT scheme for the immersion freezing (section 2.2). The deposition and contact ice nucleation are still based on the CNT scheme in the N12 and D15 experiments, and based on Meyers et al. (1992) and Young (1974) in the B53 and M92 experiments."

We have included the units in all equations of the ice nucleation schemes.

Yes, for N12, the dry diameter of dust particles is predicted by MAM4.

For the D15 scheme, we have added descriptions for instruments and measurement locations as: "D15 was developed as a combination of field campaign and laboratory data measured by the continuous flow diffusion chamber (CFDC) and the Aerosol Interactions and Dynamics of the Atmosphere (AIDA) cloud chamber. The field campaign data were obtained during the 2007 Pacific Dust Experiment (PACDEX) on the NSF/NCAR G-V aircraft over the Pacific Ocean basin (Stith et al., 2009), and the 2011 Ice in Clouds Experiment – Tropical (ICE-T) on the NSF/NCAR C-130 aircraft flown from St. Croix, US Virgin Islands (Heymsfield and Willis, 2014)."

No, marine organic aerosols and sea salt are not included as INPs in any of the parameterizations. We have added at the end of section 2.2 as the reviewer suggested: "Marine organic aerosols and sea salt are not included as INPs in any of the above ice nucleation parameterizations".

- Lines 96-97: It would be better to clarify that this is the case for the default CAM6 model with MG2 microphysics.

Reply: We thank the reviewer for the suggestion. We have revised the sentence as: "Graupel is not considered in the default CAM6 model with MG2 microphysics."

- More on the model description: line 165: What were the aerosols initialized with in SCAM and what are the aerosol types that are represented? Line 168: what aerosol-cloud interactions are represented? g. Twomey, Albrecht, glaciation indirect effect, etc.? Lines 171-172: can the cloud-borne aerosols released as interstitial aerosols be reactivated? Were the simulations not free-running or nudged to MPACE meteorology?

Reply: We thank the reviewer for the comments. The SCAM is initialized with monthly averaged aerosol concentration profiles for a given location, which are derived from a present-day CAM6 climatological simulation. The initialized aerosols and precursor gases include dust, sea salt, black carbon (BC), sulfate, particulate organic matter (POM), secondary organic aerosol (SOA), SO_2, dimethyl sulfide (DMS), and a lumped condensable organic gas species (SOAG).

In the model, Twomey, Albrecht, and INP glaciation indirect effects are represented in the model (Liu et al., 2012; Ghan et al., 2012). Yes, the cloud-bore aerosols released as interstitial aerosols can be reactivated when clouds form. The simulations are nudged to M-PACE meteorology.

We have made the corresponding changes in the revised manuscript: "In SCAM, aerosols are initialized with monthly averaged profiles for different aerosol types (sulfate, BC, particulate organic matter, secondary organic aerosol, dust, sea salt) at a given location, which are derived from a present-day CAM6 climatological simulation."
"The cloud-borne aerosols will be released to the interstitial aerosols once cloud droplets evaporate, which can be re-activated when cloud droplets are nucleated."

- Line 194: please cite the original source of the observations. The ground-based observations are not directly comparable with the model and should be stated.

Reply: We thank the reviewer for the comment. We added the original sources of the observations: "Dong and Mace, 2003; Shupe et al., 2005; Deng and Mace, 2006; Turner et al., 2007; Wang, 2007; Khanal and Wang, 2015"; "We note that these data may not be directly comparable with the model outputs" in the revised manuscript.

- Line 200: Dividing by a factor of 4 seems very approximate to account for shattering effects. I would suggest using a dataset that has been revised according to the interarrival times for more accurate comparisons (Korolev et al. 2015)

Reply: We thank the reviewer for the constructive comment. We agree with the reviewer that "Dividing by a factor of 4 seems very approximate to account for shattering effects". We adopted this method since the M-PACE data were collected before the advent of shatter mitigating tips and before algorithms for removing the shattered particles had been developed. Thus, there were no corrections for the shattering effects on these data. We discussed this issue with Greg McFarquhar who collected the M-PACE data. He suggested that we can get some estimates of the magnitude of the shattering effect on ice number concentrations from other campaigns, such as ISDAC, IDEAS-2011, and HOLODEC, which also used the 2DC cloud probe, but adopted anti-shattering tips and algorithms for removing the shattered particles.

Previous studies indicated a reduced ice number concentrations by 1-4.5 times and up to a factor of 10 depending on particle size for IDEAS-2011 and ISDAC after using the anti-shattering tips (Jackson and McFarquhar, 2014; Jackson et al., 2014). Figure 10 in Jackson et al. (2014) below indicates that the shattering effect increases the ice number by 1-4.5 times, and the effect is stronger for smaller ice than larger ice.

To address the reviewer's concern, we did a sensitivity test with a scaling factor of 1/2 to the observed ICNC, as shown in supplementary Figure S3. We added some discussion about this sensitivity test in the main text:

"A different scaling factor of 1/2 is applied to the observed ICNCs, which increases the observed ICNCs by a factor of 2 (Figure S3). The underestimation of ICNCs by the model experiments with only ice nucleation (CNT, N12 and D15) is even worse and our conclusion regarding model and observation comparison of ICNCs is not changed."

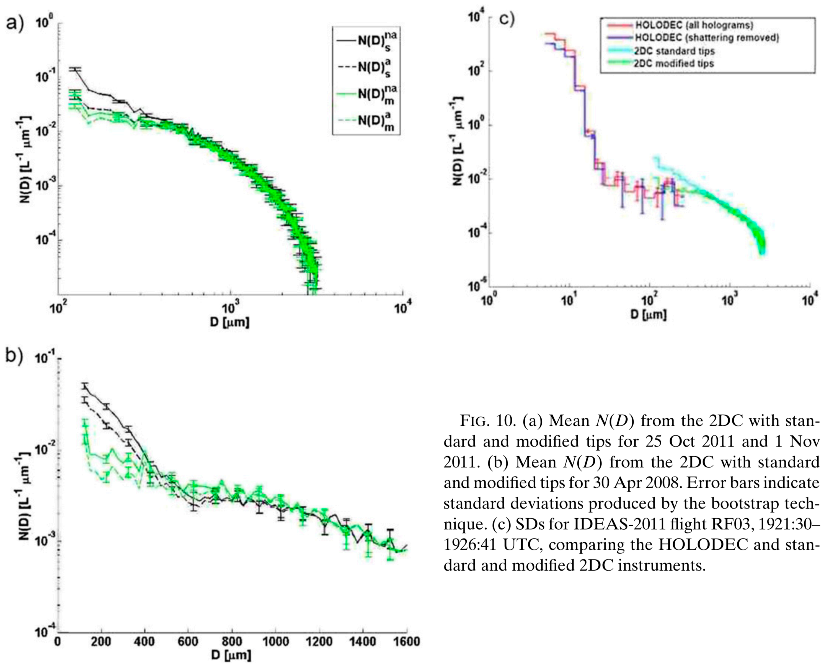
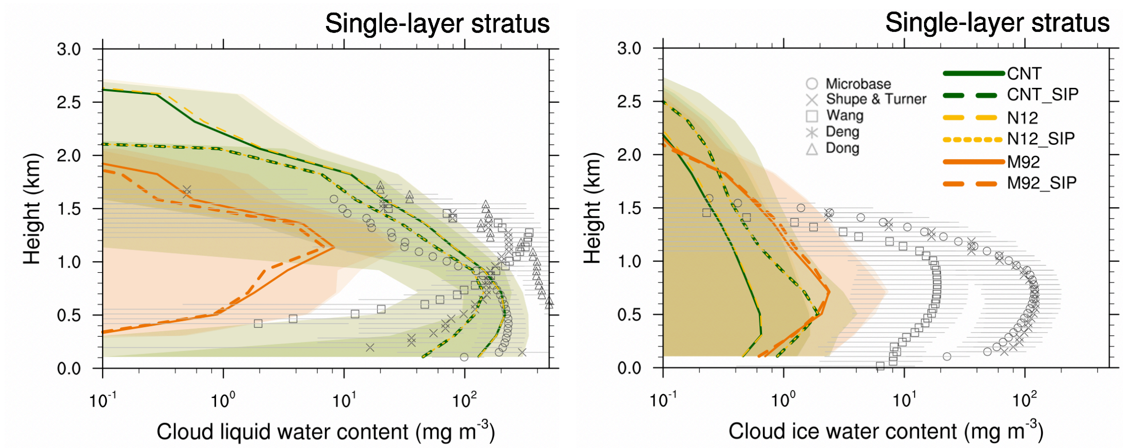


FIG. 10. (a) Mean $N(D)$ from the 2DC with standard and modified tips for 25 Oct 2011 and 1 Nov 2011. (b) Mean $N(D)$ from the 2DC with standard and modified tips for 30 Apr 2008. Error bars indicate standard deviations produced by the bootstrap technique. (c) SDs for IDEAS-2011 flight RF03, 1921:30–1926:41 UTC, comparing the HOLODEC and standard and modified 2DC instruments.

From Jackson et al. (2014), Figure 10.

- Why don't B53, B53_SIP, D15 and D15_SIP not appear in Figs. 1 and 2? Please include. Please also include the observations in Fig. 2.

Reply: We thank the reviewer for the suggestion. We have put B53, B53_SIP, D15, and D15_SIP results in Figs. S1 and S2 in the manuscript. Otherwise, Figs. 1 and 2 will be too busy, as current Figs. 1 and 2 have already had 6 lines and five makers. We have added the observations in Fig. 2 as:

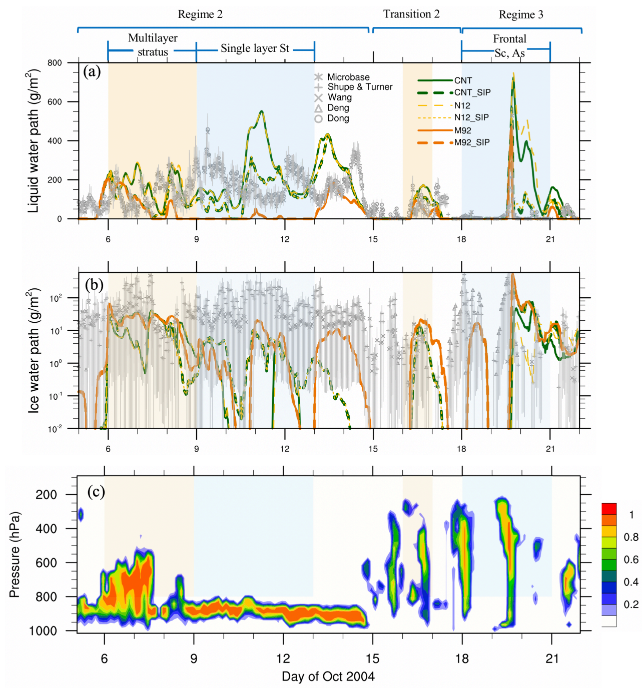


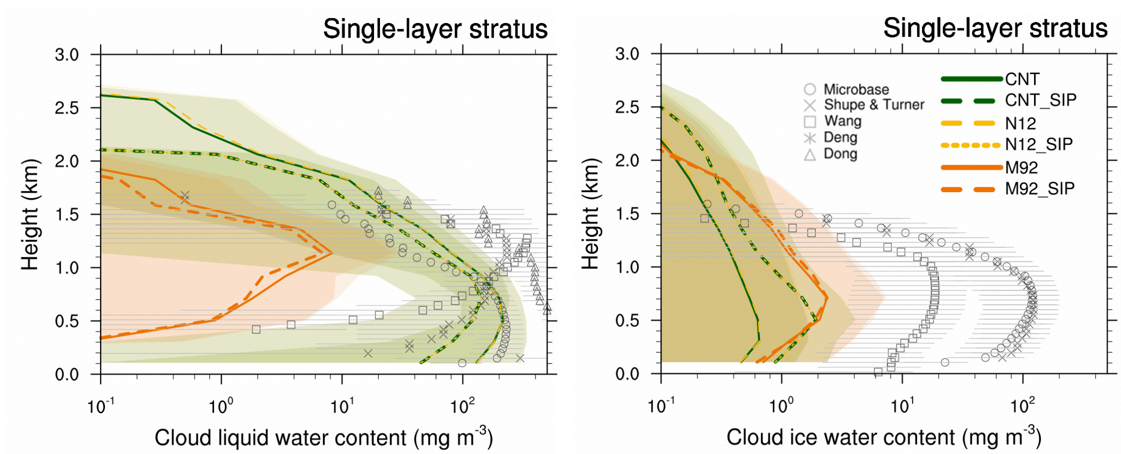
- Fig 5: I find the "enhancement ratio" confusing because the relative enhancement in Figures b-j are compared relative to Figure a, but they all use the same colour bar. Wouldn't it make more sense to use a separate colour scheme for b-j?

Reply: We thank the reviewer for the comment. We however, find that it is hard to include two color schemes in Fig. 5. Since we are plotting the bivariate joint probability density functions (PDF) for all the panels, we think that it would be cleaner to use the same color scheme and thus keep Figure 5 unchanged.

- Please include error bars in the observations and all simulations.

Reply: We thank the reviewer for the suggestion. We have added error bars in the observations and all simulations in Figures 1 and 2, which are shown below.





Minor revisions:

- Line 12: "of" needed after "importance"

Reply: Thanks. We have changed the sentence to: "the interactions between primary and SIP processes and their relative importance…"

- Line 32-34: another source of ice particles in mixed-phase cloud could be from ice crystals that fell from overlying cirrus clouds.

Reply: We thank the reviewer for the suggestion. We have added a sentence to discuss the seeding effect as: "Ice crystals that fall from overlying cirrus clouds can provide another source of ice in mixed-phase clouds."

- Lines 42-43: Ice crystal fall speed is a cloud microphysical process that is also quite important for mixed-phase cloud properties such as SLF according to the CAM5 model shown by Tan & Storelvmo 2016.

Reply: We thank the reviewer for the suggestion. We have modified the sentence as: "In addition, other microphysical processes such as rain formation, ice growth, and ice sedimentation are important for mixed-phase cloud properties (Mülmenstädt et al., 2021; Tan and Storelvmo, 2016)".

- Line 70: "Albeit these studies, how…" is grammatically incorrect.

Reply: We thank the reviewer for the suggestion. We have revised the sentence as: "Despite the above progress, many questions remain unexplored for the Arctic mixed-phase stratus clouds, e.g., whether PIP always promotes the SIP and how SIP influences the PIP."

- Line 188: "rather than" I think should be "in addition to" since Hallett-Mossop is included in all simulations?

Reply: Corrected. Thanks.

- Line 248: suggest replacing "in accompany with" with "accompanied by" and again on line 409.

Reply: Corrected. Thanks.

- Line 370: add "rate" after "nucleation"

Reply: Added. Thanks.

- Lines 423-426: Not necessary to discuss here since there is no associated figure and discussion and not central to the manuscript?

Reply: We thank the reviewer for the suggestion. These sentences are removed in the revised manuscript.