1 **Response to Reviewer 1**

- 2 We thank the anonymous reviewer for his/her careful reading and constructive review of
- 3 our paper. Our detailed responses to the comments follow. Reviewer's comments are in
- 4 blue color, our responses are in black color, and our corresponding revisions in the
- 5 manuscript are in red color.
- 6

Review of "Relative importance and interactions of primary and secondary ice
production in the Arctic mixed-phase clouds" by Zhao and Liu in ACPD, 2021.

9 In this work, the authors contrasted several parameterizations of primary ice production

10 (PIP), combined with a new set of parameterizations of secondary ice production (SIP) in

11 the NCAR CESM2/CAM6 model. The model simulations are compared with observations

12 from the DOE M-PACE campaign. The scientific questions include: What are the impacts

13 of SIP parameterizations on the simulation results? What are the effects of SIP on PIP?

- How does the PIP process influence SIP? As the authors mentioned, the interactions of SIP
 and PIP have not been carefully examined before, and the mechanisms of how they affect
 each other are still unclear.
- Overall, this is a well-written manuscript. It is very easy to follow the simulation
 experimental design since the logics are very clear and straightforward. The reviewer
 recommends that the paper be accepted after a minor revision on the following points.
- Reply: We thank the reviewer for the encouraging comments. We have revised our manuscript following your comments regarding the observation data and clarified the text to improve the quality of our paper.
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- 24

25 **Main comments:**

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1. About the comparison of ice crystal number concentration (ICNC) between observations 27 28 and simulations, the observations are restricted to > 100 micron, while the simulations use the entire size range from zero to infinity. Since ICNC is dominated by smaller ice particles, 29 the simulations may overestimate ICNC when a wider size range is used. The reviewer 30 suggests a revision on the simulation dataset to delete ice particles < 100 micron. In 31 32 addition, a scaling factor of 1/4 is applied to the observations due to potential ice shattering effect. But as the author mentioned, previous studies showed that the scaling factor may be 33 around 1 to 1/4.5. Thus, using 1/4 seems to provide a lower end of ICNC from observations. 34 35 If the authors apply another scaling factor, such as 1/2, how will it change the result? Some discussions on this sensitivity test can be added. 36

Reply: We thank the reviewer for the suggestion. Following the reviewer's suggestion, we 37 have replotted Figure 3 in which only ice particles larger than 100 microns are used, shown 38 as Figure R1 below. The purpose of Figure 3 is to examine the relative importance between 39 primary nucleation and SIP by comparing INP and ice number concentrations. The idea is, 40 41 INPs represent the primary nucleated ice, and the difference between INP and total ice number concentrations reflects the impact of SIP. Here, we are not comparing the ice 42 crystal number concentrations between observations and simulations. Therefore, we used 43 all sizes of ice crystals from the simulations in Figure 3 in the manuscript. We have added 44 45 a note in the Figure 3 caption: "The purpose of this figure is to examine the relative importance between primary ice nucleation and SIP by comparing INP and ice crystal 46 number concentrations. Therefore, all ice sizes are included in the simulation results". 47

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We have Figure 4 in the manuscript which is specifically aimed at comparing the simulated and the observed ice number concentrations. Figure 4 in the manuscript already uses the simulated ice larger than 100 microns.

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We agree with the reviewer that discussions on the observation sensitivity to a different scaling factor is necessary. We conducted a sensitivity test with a scaling factor of 1/2 as the reviewer suggested, to the observed ICNC, as shown in supplementary Figure S3 (attached below as Figure R2). We have added some discussions about this sensitivity test in the main text:

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59 "A different scaling factor of 1/2 is applied to the observed ICNCs, which increases the 60 observed ICNCs by a factor of 2 (Figure S3). The underestimation of ICNCs by the model 61 experiments with only ice nucleation (CNT, N12 and D15) is even worse and our 62 conclusion regarding model and observation comparison of ICNCs is not changed."





64 Figure R1. Same as Figure 3 but only shows ice particles with diameters larger than 100

 μ m from all the simulations.





Figure R2. Same as Figure 3a and Figure 4a, but applied a correction factor of 1/2 to the
measured ice crystal number concentrations for Figure 3a (left) and Figure 4a (right).

2. Another main comment is about the mechanism used to explain how introduction of SIP leads to weaker PIP. The authors described this mechanism around line 339 - 346, that is, "Since temperature and supersaturation are similar in these nudged simulations, the decreased cloud droplet number concentration with the introduction of SIP leads to weaker PIP in B53_SIP and M92_SIP". Can the authors clarify which variables in the SCAM simulation are nudged, such as temperature, U and V wind? Is the specific humidity nudged as well? The reviewer tries to understand why ice supersaturation is similar between the

two simulations. If there are more ice crystals produced by SIP, these ice crystals could provide more deposition of water vapor to ice phase, and thereby relaxing ice supersaturation back to ice saturation faster. Then it could lead to a suppression of PIP when ice supersaturation frequency and/or magnitude is reduced, since PIP requires a certain magnitude of ice supersaturation to occur. Also, are the ice crystals formed from SIP able to provide seeding for lower levels when they sediment? Can the seeding lead to suppression of PIP?

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Reply: We thank the reviewer for the great comment. In the SCAM simulations, wind (U and V) and temperature are nudged, while the specific humidity is not. We plotted the vertical distribution of the relative humidity with respect to ice (RHice) (Figure R3 below), and indeed the RHice is lower in M92_SIP than in M92, consistent with the reviewer's comment that more ice crystals produced by SIP should lead to more deposition of water vapor and reduce RHice in the SIP simulations. We also agree with the reviewer that this will further suppress the PIP in the SIP simulations.

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We modified the sentence in the revised manuscript as: "Since temperature is similar in these nudged simulations, the decreased cloud droplet number concentration and ice supersaturation (due to the deposition of water vapor on more ice crystals) with the introduction of SIP leads to weaker PIP in B53 SIP and M92 SIP".



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Figure R3. Relative humidity with respect to ice (RHice) from M92 and M92_SIP
 experiments averaged over the single-layer mixed-phase cloud period.

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Yes, ice crystals formed from SIP are able to provide seeding for lower level clouds when they sediment. The seeding can lead to suppression of PIP. We have added some sentences in the revised manuscript to discuss the contribution of the seeding effect. However, this effect may not be an important factor in the single-layer mixed-phase clouds, since PIP occurs in relatively higher cloud levels compared with SIP (Figure 8a and b), and low-level
 PIP may not contribute significantly to the ice formation. Ice seeding from multi-layer
 clouds is not important in this single-layer cloud period.

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We have added in the revised manuscript: "The ice crystals formed from SIP are able to provide seeding for lower-level clouds when they sediment, further contributing to the suppression of PIP. However, this effect may not be an important factor for the suppression of PIP by SIP, considering that PIP occurs at higher levels relative to SIP in the singlelayer mixed-phase clouds (Figure 8)."

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3. Following the previous comments on Figure S7, some parts of this figure do not make 117 118 sense to the reviewer. For example, accumulation mode dust decreases at 880 – 1000 hPa, but increases at 880 – 700 hPa in N12 SIP compared with N12. Why does the accumulation 119 120 mode dust increase at 880 - 700 hPa in N12 SIP, if the mechanism of SIP is to increase wet deposition (line 341)? In addition, the accumulation mode deposition in panel (e) only 121 significantly increases in N12 SIP near the surface around 980 - 1000 hPa. This pressure 122 level does not match with the location of changes seen in panel (d), and the increasing 123 deposition doesn't explain the increase of accumulation mode dust at 880 - 700 hPa as 124 125 mentioned above. The change of coarse mode deposition also doesn't match with the 126 vertical locations of changes seen in coarse mode dust. Can the authors explain this figure 127 a bit more?

Some minor comments on Figure S7, the (d) panel x axis label is out of bound on the page.Also some x axes are suggested to use the same range for an easier comparison. For

example, c, d, and e can use the same scale and unit; f and g can use the same scale.

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Reply: We thank the reviewer for the helpful comment and suggestion. We have taken a 132 more careful look at the changes of accumulation mode dust number concentration in 133 134 Figure S7d, and found that the changes are actually neglectable (~1%) compared to its absolute concentrations (0.3 versus 30 L⁻¹). Also, the accumulation mode dust contributes 135 much less to primary ice nucleation than the coarse mode dust. Therefore, we have removed 136 137 the changes in the accumulation mode dust number concentration (Figure S7d) and in the deposition rate of accumulation mode dust (Figure S7f) in the revised manuscript. We 138 139 agree with the reviewer's comment that the change of coarse mode dust deposition does not match with the vertical locations of changes in coarse mode dust, since the changes of 140 141 aerosols are influenced by other processes, such as horizontal and vertical advection, in addition to wet/dry deposition. We plot only wet deposition rate of interstitial coarse mode 142

dust in Figure S7g (now Figure S8e). As can be seen, the stronger (more negative as a sink term) wet scavenging leads to less coarse mode dust at 850-1000 hPa. Changes of coarse mode dust at 820-700 hPa (which is above the cloud layers) are mainly due to other processes such as aerosol transport, and not to cloud processes. The purpose of Figure S7 (now Figure S8) is to explain that the weaker primary ice nucleation is caused by lower cloud droplet number and less INP. Less INP is mainly due to less coarse mode dust, and the stronger wet deposition can explain the decreases of coarse mode dust at 850-1000 hPa.

Following the reviewer's comment, we have used the same unit and scale for the x-axes of INP and coarse mode dust number concentrations in the revised Figure S8. The original panels (d and f) for accumulation mode dust are removed. Revised Figure S8 looks:



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Figure R4. Vertical profiles of differences of (a) cloud droplet number concentration, (b) ice production rate from immersion freezing of cloud droplets, (c) immersion freezing INP number concentration, (d) interstitial dust number concentration in the coarse mode, and (e) wet deposition rate of interstitial coarse mode dust between the N12_SIP and N12 experiments.

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161 4. In several analyses, the authors use relative altitude to the cloud layer, that is, 0 refers to

162 cloud base and 1 refers to cloud top. There is no discussion about how this relative altitude

163 is derived. Is it derived based on ground-based observations or in-situ observations? Please164 clarify.

165 **Reply:** The relative altitude (and the associated cloud base and cloud top) used in the 166 observational analysis (in Figures 4 and 6) are provided from McFarquhar et al. (2007), 167 which are derived based on in situ observations. For our model analysis, we assume that 168 clouds exist when the total cloud water LWC+IWC >10⁻⁶ kg kg⁻¹. From the model top to 169 the bottom, the first model layer with LWC+IWC >10⁻⁶ kg kg⁻¹ is assigned as the cloud 170 top, and similarly the last model layer with LWC+IWC >10⁻⁶ kg kg⁻¹ is assigned as the 171 cloud base.

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173 We have added the following note in the captions of Figures 4 and 6:

174 "The cloud base and cloud top used for (a) are provided from in situ observations 175 (McFarquhar et al., 2007), and those used for the model analyses are derived by searching 176 the model layers from the model top to the bottom with modeled total cloud water 177 LWC+IWC >10⁻⁶ kg kg⁻¹."

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179 Minor comments:

Several simulations, CNT, N12 and D15, as well as CNT_SIP, N12_SIP and D15_SIP,
 provide similar results to each other. Can the authors provide some explanations why these
 three PIP parameterizations provide very similar results? Is it because they were derived
 based on similar observation data?

Reply: We thank the reviewer for the comment. Although these three parameterization 184 schemes differ in details about temperature and aerosol dependences, CNT, N12, and D15 185 predict much lower INP concentrations for the M-PACE single-layer clouds compared with 186 the B53 and M92 schemes. With low INP concentrations, modeled clouds are 187 188 overwhelmingly dominated by liquid-phase. Therefore, it is not surprising to see the overall similar cloud features among the CNT, N12, and D15 simulations. In contrast, B53 and 189 M92 which are only dependent on temperature and not limited by aerosols predict much 190 higher INP concentrations. With these high INP concentrations, modeled clouds with the 191 192 B53 and M92 schemes are dominated by ice-phase.

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194 We have added a note when we discuss about Figure 2 in the revised manuscript:

- 195 "Although these schemes differ in details about temperature and aerosol dependences
- 196 (Figure 3), CNT, N12, and D15 predict much lower INP concentrations during M-PACE

- 197 than those from the B53 and M92 schemes. With these low INP concentrations, the single-
- 198 layer clouds modeled with the CNT, N12 and D15 schemes have similar cloud states (e.g.,
- dominated by liquid-phase) (Figures 1 and 2). In contrast, B53 and M92 which are only
 dependent on temperature and not limited by aerosols predict much higher INP
 concentrations. With these high INP concentrations, modeled clouds with the B53 and M92
 schemes are dominated by ice-phase."
- 203

204 2. Some of the analyses and figures are based on ground-based remote sensing observations
205 (such as Figure 1) while the other ones are based on in-situ aircraft observations. It would
206 be beneficial to clarify in Section 3, such as line 182, which type of observations is used in
207 a specific figure or analysis.

- 208 **Reply:** We thank the reviewer for the suggestion. We have added: "The ice water path (IWP) and liquid water path (LWP) are based on ground-based remote sensing 209 210 observations provided by Zhao et al. (2012) with uncertainties within one order of magnitude (Dong and Mace, 2003; Shupe et al., 2005; Deng and Mace, 2006; Turner et al., 211 2007; Wang, 2007; Khanal and Wang, 2015). The INP concentrations are based on in-situ 212 observations by a CFDC on board an aircraft (Prenni et al., 2007). The ICNCs and cloud 213 phase are based on in-situ observations and provided by McFarquhar et al. (2007)." in 214 215 Section 3 to clarify the types of observation data used in the analyses.
- 216

217 3. Please clarify how the variables related to "rate" are defined in the model. For example,

218 is the variable ice production rate describing the amount of ice crystals (in kg) being

219 produced in every unit mass of dry air (in kg) per second in the entire grid box, or only in 220 the in-cloud section of the grid box?

- Reply: We thank the reviewer for the comment. In the model, the unit of ice production rates is kg (ice crystals)/(kg dry air)/s, and all the ice production rates are grid box mean values. We have added the unit and a note to the captions of Figures 8 and 9.
- 224

4. Figure 2, is it possible to add sub-panels of observations to compare with the modelresults?

- 227 **Reply:** We thank the reviewer for the suggestion. We have added the observation data
- 228 (including standard deviations) in Figure 2, and the revised figure looks:



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Figure R5. Vertical profiles of LWC (left) and IWC (right) during the single-layer mixedphase cloud period (October 9-12) from CNT, CNT_SIP, N12, N12_SIP, M92, and M92_SIP experiments and from remote sensing retrievals (symbols). Horizontal gray lines represent standard deviations of retrieval data, and colored shadings represent standard deviations of model data. Note that N12 (N12_SIP) coincides with CNT (CNT_SIP) during the single layer stratus cloud period.

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5. Figure 3, since the INP concentrations in CNT, N12 and D15 are significantly lower
than the observations, can the authors apply a scaling factor to INPs in these
parameterizations to match with the observations better, and see how the results change?
Also, the reviewer wonders why with such low INP concentrations, these parameterizations
are able to produce quite a similar amount of ICNC compared with observations?

243 **Reply:** We thank the reviewer for the good suggestion. Applying a scaling factor to INPs means changing the INP parameterizations. Instead, we undertook another approach by 244 increasing dust aerosol concentrations used in the INP parameterizations, which will also 245 result in more INPs. We have conducted a sensitivity test using the CNT scheme with 246 increased dust concentrations by 100 times. This simulation shows overall similar cloud 247 properties, but the relative contribution of primary ice nucleation to total ice production is 248 increased by a factor of ~2 during M-PACE. We have added a discussion on this in Section 249 4.2 in the revised manuscript as: "Since the INP number concentrations in CNT, N12 and 250 D15 are significantly lower than the observations (Figure 3), a sensitivity test using the CNT 251 252 scheme with increased dust concentrations by 100 times shows overall similar cloud properties. However, the relative contribution of primary ice nucleation to total ice 253

production is increased by a factor of ~ 2 to 30% averaged for all the cloud types and to 20% for the single-layer mixed-phase clouds."

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For the reviewer's question: "why such low INP concentrations produce quite a similar amount of ICNC compared with observations?" The ICNCs in CNT, N12, and D15 experiments are actually 1-2 orders of magnitude lower than observed ICNCs as shown in Figure 4, not at a similar amount. The simulations with SIP (using CNT, N12, and D15) produce similar amounts of ICNCs at the lower portion of clouds compared with observations.

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6. Figure 5, please clarify how the normalization was calculated in this figure. It seems that the PDF is calculated by the number of samples of each bin divided by the total number of samples in each temperature bin (the sum of % in each temperature range equals one), instead of divided by the total number of samples of the entire temperature range. Is that correct? The reviewer wonders how this figure will change, if the latter type of normalization is also provided (i.e., the sum of % in all bins equals one).

Reply: We thank the reviewer for the question. We are sorry for the confusion. Figure 5 270 shows the probability of occurrence defined in terms of both temperature and ice number 271 272 concentration for Figure 5a (or enhancement ratio for Figure 5b-j), which means PDF is 273 calculated by the number of samples of each bin divided by the total number of samples of 274 the entire temperature range (i.e., the sum of % in all bins equals one). We have revised 275 the caption of Figure 5 as: "Figure 5. Bivariate joint probability density functions (PDF) in terms of both temperature and (a) ice crystal number concentration (L⁻¹) from the CNT 276 experiment, and (b)-(j) in terms of both temperature and enhancement ratio of ice crystal 277 278 number concentration from the respective experiment to that from the CNT experiment. A logarithmic scale is used for the x-axis." 279

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7. Figure 9, for the accretion rate of cloud water by snow, does cloud water include both
cloud droplets and rain? Some minor revisions on the sub-title of g and h are recommended.
For example, h can be "Droplet number" instead of "Cloud number", and h can be "Accrete
water by snow".

Reply: We thank the reviewer for the question and suggestion. Figure 9h is the accretion rate of cloud droplets by snow, and the cloud water does not include rain. We have revised the sub-titles of Figure 9g and h as you suggested to "Cloud droplet number" and "Accrete

cloud water by snow", respectively. The revised Figure 9 looks:



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Figure R6. Vertical profiles of (a) ice production rate (unit: kg kg⁻¹ s⁻¹) from immersion 290 freezing of cloud water, (b) ice production rate (unit: kg kg⁻¹ s⁻¹) from contact freezing of 291 cloud water, (c) ice production rate (unit: $kg kg^{-1} s^{-1}$) from homogeneous and heterogeneous 292 deposition nucleation, (d) immersion freezing INP number concentration, (e) cloud-borne 293 dust number in the accumulation mode, (f) cloud-borne dust number in the coarse mode, 294 (g) cloud droplet number concentration, (h) accretion rate of cloud droplets by snow, and 295 296 (i) WBF process rate from CNT and CNT SIP experiments averaged over the single-layer mixed-phase cloud period. Light blue shadings indicate the ice nucleation regime. Ice 297 production rates are grid-box means. 298

299 **Response to Reviewer 2**

- 300 We thank the anonymous reviewer for his/her careful reading and constructive review of
- 301 our paper. Our detailed responses to the comments follow. Reviewer's comments are in
- 302 blue color, our responses are in black color, and our corresponding revisions in the
- 303 manuscript are in red color.
- 304
- Review of Manuscript # acp-2021-686 in ACPD: "Relative importance and interactions of
 primary and secondary ice production in the Arctic mixed-phase clouds" by Zhao and Liu.
- 306 primary and secondary ice production in the Arctic
- 307 General comments:
- 308 The authors examined five different ice nucleation schemes and secondary ice production (SIP) processes in the simulations of Arctic mixed-phase clouds during the M-PACE 309 310 campaign using single column mode of CESM2 CAM6 model. They concluded that the simulations using aerosol-aware ice nucleation schemes and including SIP processes 311 312 resemble the observed single-layer mixed-phase clouds during the M-PACE. In these simulations, SIP plays a key role, and there is a competition between ice nucleation and 313 314 SIP. Overall, the manuscript is well organized, and the logic is clear. However, there are several concerns that should be clarified before considering the manuscript for publication. 315 The reviewer would recommend major revision for this manuscript in case the authors need 316
- 317 more time for revision.
- Reply: We thank the reviewer for the positive comments. We have revised the manuscript following your suggestions regarding the quantitative analyses and clarified the text to improve the quality of our paper.
- 321

322 Major comments:

Analyses: The analyses in the manuscript are full of qualitative phrases. Some
examples are listed in the minor comments. Please conduct quantitative analyses.

- Reply: We thank the reviewer for the suggestion. We have conducted quantitative analyses
 and improved the qualitative phrases in the revised manuscript.
- 327

2. How did the authors attain the simulated ice crystal number concentration (ICNC) for comparison with observations? Did the authors consider snow particles? Because observations should include all types of ice particles, the authors should include all ice categories for comparison. Meanwhile, in the comparison only the observed ICNC with sizes larger than 100 microns are considered, while the entire size range of simulated ICNC is used. So, the comparison is also unfair. Please use the same size range of all types of ice particles for comparison. Reply: We thank the reviewer for the comments. The simulated ice crystal number concentration (ICNC) includes both cloud ice and snow particles, for a consistent comparison with observations. We have added a sentence in the revised manuscript as: "Since the measurements cannot distinguish snow from cloud ice, the simulated ICNC, IWP, and IWC all include the snow component for the comparison with observations."

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Following the reviewer's suggestion, we have replotted Figure 3 in which only ice particles 341 larger than 100 microns are used from simulations, shown as Figure R1 below. The purpose 342 343 of Figure 3 is to examine the relative importance between primary ice nucleation and SIP by comparing INP and ice number concentrations (not comparing simulated and observed 344 ICNC). The idea is, INPs represent the primary nucleated ice, and the difference between 345 346 INP and total ice number concentrations reflects the contribution of SIP. Therefore, we used all sizes of ice crystals in Figure 3. We have added a note in the Figure 3 caption: 347 "The purpose of this figure is to examine the relative importance between primary ice 348 nucleation and SIP by comparing INP and ice crystal number concentrations. Therefore, 349 350 all ice sizes are included in the simulation results".

351

We have Figure 4 in the manuscript which is specifically aimed at comparing the simulated and the observed ice number concentrations. Figure 4 already uses the simulated ice larger than 100 microns, so we do not modify Figure 4.



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Figure R1. Same as Figure 3 but only shows ice particles with diameters larger than 100
 μm from simulations.

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360 3. Lines 199-203: "M-PACE observed ICNCs were scaled by a factor of 1/4", have 361 the data collected by the authors been scaled by a factor to remove the shattering effect 362 during the data quality control? Are the conclusions sensitive to this correction factor? 363 **Reply:** We thank the reviewer for the comment. The observed ICNC data we used in this 364 study do not remove the shattering effect during the data quality control, since the ICNCs 365 for M-PACE were measured before anti-shattering algorithms were developed to remove 366 the shattered particles for the 2DC cloud probes. We contacted the data collector Dr.

- McFarquhar to confirm this. At his suggestion, we applied a factor of ¹/₄ to the M-PACE
 observed ICNCs.
- 369

370 We have conducted a sensitivity test with a scaling factor of 1/2 to the observed ICNCs, as 371 shown in supplementary Figure S3 (attached below as Figure R2). The conclusion of model and observation comparison of ICNCs is not sensitive to this correction factor. We added 372 some discussions about this sensitivity test in the main text: "A different scaling factor of 373 374 1/2 is applied to the observed ICNCs, which increases the observed ICNCs by a factor of 375 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 376 377 observation comparison of ICNCs is not changed."



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Figure R2. Same as Figure 3a and Figure 4a, but applied a correction factor of 1/2 to the measured ice crystal number concentrations for Figure 3a (left) and Figure 4a (right).

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382 4. The reviewer was surprised as the results shown in Figures 1 and 2. N12 (N12_SIP)
383 seems to be the same as CNT (CNT_SIP), but their INPs are obviously different in Figure
384 3. Why?

Reply: We thank the reviewer for the great comment. Although these three schemes differ in details about temperature (and aerosol) dependences (Figure 3), CNT, N12, and D15 predict much lower INP concentrations for the M-PACE single-layer clouds than those from the B53 and M92 schemes. With these low INP concentrations, modeled clouds are overwhelmingly dominated by liquid-phase (Figures 1, 2, and 6). Therefore, it is not surprising to see the overall similar cloud states among CNT, N12, and D15. For comparison, B53 and M92 which are only dependent on temperature and not limited by aerosols predict much higher INP concentrations. With these high INP concentrations, modeled clouds with the B53 and M92 schemes are dominated by ice-phase.

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We have added a note when we discuss about Figure 2 in the revised manuscript:

396 "Although these schemes differ in details about temperature and aerosol dependences (Figure 3), CNT, N12, and D15 predict much lower INP concentrations during M-PACE 397 398 than those from the B53 and M92 schemes. With these low INP concentrations, the singlelayer clouds modeled with the CNT, N12 and D15 schemes have similar cloud states (e.g., 399 dominated by liquid-phase) (Figures 1 and 2). In contrast, B53 and M92 which are only 400 dependent on temperature and not limited by aerosols predict much higher INP 401 402 concentrations. With these high INP concentrations, modeled clouds with the B53 and M92 schemes are dominated by ice-phase." 403

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It is not clear that how the authors attained the INP number concentrations from
observations and simulations especially for B53 scheme. Did the author conduct a fair
comparison between them? Did the authors include all types of ice nucleation for
comparison? Please provide a more detailed description.

- **Reply:** We thank the reviewer for the questions. The INP number concentrations were 409 measured by a CFDC on board an aircraft (Prenni et al., 2007) during the M-PACE single-410 layer mixed-phase cloud period. For the B53 scheme in the model, we use Equation 4 to 411 calculate the immersion freezing rate, and diagnose INP number concentrations by 412 multiplying the immersion freezing rate by the model timestep. The contact ice nucleation 413 414 is based on Young (1974), and deposition ice nucleation on Meyers et al. (1992) in the model simulation. We include all these types of ice nucleation for the comparison with 415 observations. However, for the single-layer mixed-phase clouds, the immersion freezing is 416 dominated, and the contributions from deposition and contact ice nucleation to total ice 417 production are much smaller (see Figure R3 below). 418
- 419

420 We have provided a more detailed description in section 3: "The N12, D15, B53, and M92

421 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, or based on Meyers et al. (1992) and Young (1974), respectively in the B53
and M92 experiments."

In section 4 (Results) we added: "The contributions from deposition and contact ice
nucleation to total ice production are much smaller compared to the immersion freezing
for the single-layer mixed-phase clouds during M-PACE."

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Figure R3. Vertical profiles of (a) ice production rate (unit: kg kg⁻¹ s⁻¹) from immersion freezing of cloud water, (b) ice production rate (unit: kg kg⁻¹ s⁻¹) from contact freezing of cloud water, and (c) ice production rate (unit: kg kg⁻¹ s⁻¹) from deposition nucleation calculated in the B53 and B53_SIP experiments.

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6. "Section 4.3 Interactions between PIP and SIP": SIP suppressed the PIP. Did the
authors consider whether some setups in the microphysics scheme lacking physical
meaning result in or enhance this suppression? For example, suppression is due to
decreasing difference between total ice nucleation number from parameterization and
increasing ice particle number. Please provide a discussion.

442 **Reply:** We thank the reviewer for the good question. We understand that the reviewer is talking about the ice nucleation tendency calculated as the difference between total ice 443 nucleation number from parameterization and ice particle number at current model time 444 445 step. This tendency is reduced when the current time step ice particle number is increased due to SIP. However, the ice production rates (for ice mass) from ice nucleation shown in 446 Figure 10 are directly calculated by the CNT ice nucleation parameterization, which are 447 the number of ice crystals nucleated from the parameterization times the initial mass of an 448 449 ice particle $(2.093 \times 10^{-15} \text{ kg})$. As we explain in the text, the suppression of PIP by SIP is due to lower number concentrations of INPs and cloud droplets after considering SIP. 450

452 7. Some "rate"s in the manuscript are confusing. If the reviewer understood correctly, 453 the production rates in the manuscript are mainly for ice mass based on Figures 8-10. The 454 question is how IIC increases ice mass? The "ice" in the manuscript all means "cloud ice" 455 and does not include "snow"? If yes, following comment #2, different categories of ice are 456 defined artificially in microphysics schemes, and it might not be true in observations. The 457 authors should clarify it. The reviewer would recommend conducting analyses including 458 simulated snow particles.

- 459 **Reply:** We thank the reviewer for the suggestion. Yes, the production rates in Figures 8-10 460 are for ice mass, which are calculated from ice production rates for ice number from the 461 parameterizations multiplied by the initial mass of an ice particle $(2.093 \times 10^{-15} \text{ kg})$. We 462 added a note in the text: "The ice mass production rates are calculated by multiplying ice 463 number production rates from parameterizations by the initial mass of an ice particle 464 $(2.093 \times 10^{-15} \text{ kg})$."
- 465

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466 In all analyses for the comparison of modeled ICNCs, IWP, and IWC with observations, modeled cloud ice and snow are added together. We agree with the reviewer that cloud ice 467 and snow are separated artificially in the microphysics scheme in the model. IIC represents 468 the process that snow particles collide with each other and produce smaller cloud ice 469 470 particles due to the snow fragmentation. In Figure 10c, the IIC process rate indicates an increase in cloud ice mass from the fragmentation of colliding snow particles. Ice mass is 471 converted from snow to cloud ice in the IIC process, although the total ice mass is not 472 changed. 473

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475 **Minor comments:**

476 1. Lines 107-108: Please describe how the graupel mass and number are diagnosed in477 the scheme briefly.

478 **Reply:** We thank the reviewer for the comment. We diagnose the graupel mass based on
479 cloud water, cloud ice, and snow mass mixing ratio. We have added the diagnostic method
480 in the revised manuscript as:

481 "The graupel mass mixing ratio (q_g) is diagnosed as the precipitation ice mass (currently 482 snow, q_s) multiplied by the rimed mass fraction *Ri* (Zhao et al., 2017),

483

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

484 The rimed mass fraction *Ri* is calculated as:

485
$$Ri = \frac{m_{rimed}}{m_{rimed} + m_{unrimed}} \approx \frac{1}{1 + \frac{6 \times 10^{-5}}{q_c(q_i + q_s)^{0.17}}}$$
(7)

486 $q_c, q_i, and q_s in (7)$ are modeled cloud water, cloud ice, and snow mixing ratios (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."

489

Lines 209-225: Please quantify the analyses, e.g., percentage of enhancement,
reduction, "largest", "smallest", "modest", "closest", "significantly
decreases/increases", ...

493 **Reply:** We thank the reviewer for the suggestion. We have modified the sentences as:

494 "In the SIP experiments with the CNT, N12, and D15 ice nucleation schemes, simulated IWP is increased from 5 to 10 g m⁻² and LWP is decreased from 156 to 97 g m⁻² averaged over 495 the M-PACE period after considering the SIP. In the SIP experiments with the B53 and M92 496 497 schemes, however, SIP has a minimal impact on the LWP/IWP. Second, the B53, B53 SIP, M92, and M92 SIP produce the largest IWP (\sim 12 g m⁻² averaged over the M-PACE period), 498 followed by CNT SIP, N12 SIP, and D15 SIP (~10 g m⁻² averaged over the M-PACE 499 period). CNT, N12, and D15 experiments produce the smallest IWP (~5 g m⁻² averaged over 500 the M-PACE period). These characteristics are also evident in the vertical profiles of LWC 501 and IWC in Fig. 2 and Fig. S2. It indicates that the B53 and M92 nucleation schemes are 502 highly efficient in forming ice; in comparison, the SIP simulations using CNT/N12/D15 ice 503 nucleation schemes show the lower ice production capabilities. B53, B53 SIP, M92, and 504 M92 SIP experiments generate the closest IWP (~12 g m⁻² averaged over the M-PACE 505 period) compared with the observation (~64 g m⁻²). However, these four experiments also 506 show substantially low biases of LWP (~40 g m⁻² compared with 126 g m⁻² in the observation 507 averaged over the M-PACE period). As shown in Fig. 1 and Fig. S1, the mixed-phase clouds 508 are almost fully glaciated during the single layer stratus period. Therefore, the CNT SIP, 509 N12 SIP, and D15 SIP experiments give the best simulation results in terms of LWP and 510 IWP during the M-PACE. Adding the SIP does not change the modeled LWP/LWC and 511 IWP/IWC with the B53 and M92 ice nucleation schemes. On the contrary, SIP decreases the 512 513 LWP/LWC by 38% and doubles the IWP/IWC with the CNT, N12, and D15 ice nucleation schemes." 514

515

516 3. Lines 233-234: "appears an inversely linear relationship", "this relationship is not 517 as clear", do they have statistical significance?

Reply: We thank the reviewer for the comment. The purpose of this figure is to compare N_{INPs} with ICNCs, not to derive a relationship between N_{INPs} and temperature. We have removed the word "linear" and revised the related sentence as: "With the empirical ice

521 nucleation schemes (e.g., N12 and D15), there appears an inversely relationship between 522 $\log_{10}(N_{INPs})$ and temperature".

523

524 4. Lines 234-238: Please quantify the analyses, e.g., "reduces dramatically", "much
525 higher", ...

Reply: We thank the reviewer for the suggestion. We have revised the sentences as: "However, this relationship is not as clear with the CNT and B53 schemes, and N_{INPs} reduces rapidly at temperatures warmer than -15 °C, from $\sim 10^{-1}$ L⁻¹ at -17° C to $< 10^{-5}$ L⁻¹ at -13° C (Fig. 3b, e). In contrast, N_{INPs} with the aerosol-independent M92 scheme is less variable with temperature, and is 1-7 orders of magnitude higher than that with the aerosol-aware schemes"

531

5. Lines 253-264: Why is SIP not active in B53_SIP and M92_SIP? Is there a maximum threshold of ICNCs in the microphysics scheme?

534 **Reply:** We thank the reviewer for the comment. We understand the reviewer's concern that the inactivity of SIP in B53 SIP/M92 SIP might be caused by a maximum threshold of 535 536 ICNCs imposed in the microphysics scheme. However, this is not the case in the two model experiments. We have conducted in-depth analyses and given an explanation in Section 537 4.3 (Figure 10). The reason for the inactive SIP in B53 SIP/M92 SIP is because of the 538 539 competition between PIP and SIP (Figure 10). Too strong primary ice nucleation in 540 B53 SIP and M92 SIP consumes available liquid cloud water, which results in less graupel in clouds. With less graupel amount, SIP through IIC is suppressed (see detailed 541 542 explanation in Section 4.3 and Figure 10).

543

544 6. Line 269: Please quantify "slightly higher"

Reply: We thank the reviewer for the suggestion. We have calculated the vertically integrated ice number to be 1.649×10^6 and 1.646×10^6 m⁻² in the N12 and D15 experiments, respectively. So, ice number concentrations in N12 and D15 are very similar. We have removed: "even though the N12 experiment has a slightly higher ice enhancement ratio compared with the D15 experiment."

550

551 7. Lines 281 and 283: Please quantify "overestimate", "predominantly"

Reply: We thank the reviewer for the suggestion. We have provided quantitative numbers and revised the sentences as: "The CNT, N12, and D15 experiments share the similar cloud phase distribution and all overestimate the SLF in clouds with the vertically averaged SLF of 96.25%, 96.28%, and 96.26% in CNT, N12, and D15, respectively, compared to 64.35% from the observation. On the contrary, the B53 and M92 experiments with more efficient ice nucleation show predominantly ice phase clouds with the vertically averaged SLF of 17.62%

and 16.43%, respectively, which agrees with previous findings (Liu et al., 2011)."

559

560 8. Lines 287-288: How about the TWC in these simulations?

Reply: We thank the reviewer for the suggestion. The TWP is reduced with decreased LWP (and SLF) and increased IWP in these simulations, as shown in Table R1 below. We have added a sentence in the revised manuscript: "The TWC is reduced with the total water path (TWP = LWP + IWP) decreased from 218.5, 219.2, and 219.1 in CNT, N12, and D15 to 132.6, 131.0, and 130.8 in CNT_SIP, N12_SIP, and D15_SIP, respectively".

566

567 Table R1. LWP, IWP, and TWP in different experiments for the single layer mixed-phase

568 clouds period.

	LWP	IWP	TWP
Obs	190.19	74.66	264.85
CNT	217.62	0.93	218.55
N12	218.30	0.95	219.25
D15	218.12	0.97	219.09
CNT_SIP	129.98	2.58	132.55
N12_SIP	128.40	2.61	131.01
D15_SIP	128.19	2.62	130.81

569

570 9. Lines 294-308: It is confusing whether the authors talked about ice number or mass
571 in Figure 7. If the authors talked about ice mass in Figure 7, how do IIC contribute to ice
572 mass?

Reply: We thank the reviewer for the suggestion. We are sorry for the confusion. Figure 7 shows the relative contribution from different processes to the total ice (mass) production rate. We output PIP and SIP number process rates, and multiple them by the initial mass of an ice particle $(2.093 \times 10^{-15} \text{ kg})$ to calculate the ice (mass) production rates used in Figure 7. We added a note in the manuscript: "The ice mass production rates are calculated by multiplying ice number production rates from parameterizations by the initial mass of an ice particle $(2.093 \times 10^{-15} \text{ kg})$."

580

581 IIC represents the process that bigger snow particles collide with each other and produce 582 smaller ice particles due to fragmentation. In the model, IIC process rate indicates a mass 583 transfer from snow to cloud ice. It is true, the total ice mass is not changed, but ice mass is transferred from snow to cloud ice in the model, which separates the total ice into cloud ice and snow categories.

586

587 10. Line 328: Please quantify "substantially weakened"

588 Reply: We thank the reviewer for the comment. We have revised the sentence as: "The
589 immersion ice nucleation is weakened by a factor of 4.5 (Fig. 9a) ..."

590

591 11. Lines 342-343: Based on Eq. (5), M92 seems dependent on supersaturation not
592 temperature and cloud droplet number concentration.

Reply: We thank the reviewer for the comment. We are sorry for the confusing. M92 is dependent on ice supersaturation. Since the model microphysics assumes saturation vapor pressure with respect to liquid in mixed-phase clouds to calculate ice supersaturation (i.e., $(e_{sl}-e_{si})/e_{si}$, e_{sl} and e_{si} are the saturation vapor pressures with respect to liquid and to ice, respectively), M92 is indirectly dependent on temperature. In the model, if there are no cloud droplets, ice nucleation will not occur. Thus, M92 also depends indirectly on cloud droplet number concentration.

600

601

1 12. Lines 362-367: Please quantify the analysis.

602 **Reply:** We thank the reviewer for the suggestion. we have revised the related sentences as "A smaller graupel-related IIC rate (with the peak value of 2 kg kg⁻¹ s⁻¹) (Fig. 10f) in 603 M92 SIP compared to CNT SIP (with the peak value of 10 kg kg⁻¹ s⁻¹) is a result of smaller 604 graupel mass mixing ratio in M92 SIP (with the peak value of 1.4 mg kg⁻¹ in M92 SIP 605 versus 5.2 mg kg⁻¹ in CNT SIP) (Fig. 10g). As the graupel mass is diagnosed from the cloud 606 water mass, snow mass, and temperature, smaller mass mixing ratios of cloud water (with 607 the peak value of 8 versus 125 mg kg⁻¹ in Fig. 10h) and snow (with the peak value of 1.4 608 versus 2.3 mg kg⁻¹ in Fig. 10i) in M92 SIP eventually lead to a smaller graupel mass mixing 609 ratio and a smaller graupel-related IIC rate. Similar results can be found with the other ice 610 611 nucleation schemes."

612

613 13. Figure 1: Please provide uncertainties of these observations.

Reply: We thank the reviewer for the suggestion. We have revised Figures 1, 2, S1, and S2
to include uncertainties (standard deviations) of these observations. The revised Figures 1
and 2 look:



617 618 Fa



620 Figure R5.

619

621 14. Figure 4: How did the authors determine the cloud top and cloud base for 622 observations and simulations?

623 **Reply:** We thank the reviewer for the suggestion. The observation data are from 624 McFarquhar et al. (2007), and they have already determined the cloud top and cloud base 625 for observation data we use in this study. More information can be found in the data 626 description paper (McFarquhar et al., 2007). For our model analysis, we assume that clouds 627 exist when LWC+IWC >10⁻⁶ kg kg⁻¹. From the model top to bottom, the first model layer 628 with LWC+IWC >10⁻⁶ kg kg⁻¹ is the cloud top, and similarly the last model layer with 629 LWC+IWC >10⁻⁶ kg kg⁻¹ is assigned as the cloud base.

- 630
- 631 15. Figure 5: x-axis in (h), "CTL" -> "CNT"? What are the bin sizes for x and y
- 632 variables?

633 **Reply:** We thank the reviewer for catching the typo in Figure 5h, which we have corrected.

We have used 25 bins for x and y variables. The bin size for temperature is 2 degree, and the bin size for the ice number/ice enhancement is calculated by (maximum value minimum value)/25.



- 637
- 638 Figure R6.
- 639

640 16. Figure 7: "total ice production rate", is the "production rate" for mass or number? 641 **Reply:** We thank the reviewer for the suggestion. We output PIP and SIP number process 642 rates, and multiple them by the initial mass of an ice particle $(2.093 \times 10^{-15} \text{ kg})$ to calculate 643 the ice mass production rates used in Figure 7. We added a note in the text: "The ice mass 644 production rates are calculated by multiplying ice number production rates from 645 parameterizations by the initial mass of an ice particle $(2.093 \times 10^{-15} \text{ kg})$." Figure 9: "(h) accretion rate of cloud water by snow", how about the accretion of
rainwater by snow? **Reply:** We thank the reviewer for the comment. Figure 9h only shows the accretion of

649 **Reply:** We thank the reviewer for the comment. Figure 9h only shows the accretion of 650 cloud water by snow, and does not include the accretion of rain by snow, since the purpose 651 of this figure is to illustrate that a stronger "accretion rate of cloud water by snow" (8 vs. 2 652 kg kg⁻¹ s⁻¹) results in a lower cloud water amount (13 mg kg⁻¹) in the CNT_SIP experiment 653 compared with that (23 mg kg⁻¹) in the CNT experiment.

654

655 18. Figure 10: (c), (e), (F), IIC influences the mass mixing ratio of ice particles?

- 656 **Reply:** We thank the reviewer for the suggestion. This IIC process transfers ice mass from
- snow to cloud ice, although the total ice mass does not change.
- 658

659 **Response to Reviewer 3**

660 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
- 663 manuscript are in red color.
- 664

665 Review for "Relative importance and interactions of primary and secondary ice production

666 in the Arctic mixed-phase clouds" by Zhao & Liu

667

668 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-669 670 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 671 672 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 673 674 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 675 3 of the ice nucleation schemes that are aerosol-aware (CNT, N12 and D15) exhibit similar 676 677 behaviour to each other in terms of their simulated ice crystals number concentration 678 vertical profiles, supercooled liquid fraction (SLF), IWP, LWP and relative contributions 679 from primary and SIP rates to the total ice production rate. They also find that these 680 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 681 also conclude that the aerosol-aware ice nucleation schemes with the IC and FR 682 683 mechanisms represented best represent the single-layer mixed-phase clouds observed during M-PACE. 684

685

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.

692 **Reply:** We thank the reviewer for the positive comments. We have revised the manuscript

693 following your comments regarding the writing including the description of the model used,

694 the experimental design, and the observations to improve the quality of our paper.

- 695 Major revisions:
- 696
- 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.

702

703 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 704 suppression of PIP via SIP is less intuitive since one would initially expect that more ice 705 706 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 707 and M92 due to the enhanced glaciation of mixed-phase clouds. A description of the 708 graupel scheme (which seems to be diagnostic based on line 364) the authors implemented 709 710 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 711 Abstract. Also, the discussion on lines 73-78 in the Introduction can also be elaborated on 712 713 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).

718

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,

721 q_s) multiplied by the rimed mass fraction *Ri* (Zhao et al., 2017),

724

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

723 The rimed mass fraction *Ri* is calculated as:

$$Ri = \frac{m_{rimed}}{m_{rimed} + m_{unrimed}} \approx \frac{1}{1 + \frac{6 \times 10^{-5}}{q_C(q_i + q_s)^{0.17}}}$$
(7)

 $q_c, q_i, and q_s in (7)$ are modeled cloud water, cloud ice, and snow mixing ratio (kg kg⁻¹),

726 <u>respectively. The graupel number is assumed to have the same ratio to snow number as the</u>

- 727 ratio of graupel mass to snow mass."
- 728

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)."

734

• 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% 739 740 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 741 742 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; 743 744 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 745 impact in a local region when they do occur, with up to a 1,000-fold enhancement in ice 746 747 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. 748 749 We note that ice number concentrations are three orders of magnitude higher than INP 750 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. 751

752

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."

757

• 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, 770 contact, and deposition nucleation separately with different equations. With many 771 equations involved in the CNT scheme, we prefer not to include them in the paper, but 772 773 refer the readers to Wang et al. (2014) and Hoose et al. (2010). The CNT scheme is 774 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 775 776 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). 777

778

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)."

783

784 The N12, D15, B53, and M92 are empirical schemes for the immersion freezing of cloud 785 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 786 ice nucleation is represented by M92 and the contact ice nucleation by the Young (1974) 787 scheme. We understand that there is an inconsistency in the representation of deposition 788 and contact ice nucleation in these experiments. However, for the single-layer mixed-phase 789 clouds, immersion freezing is dominated, and the contributions from deposition and contact 790 791 ice nucleation to total ice production are much smaller (Figure 9).

792

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."

799

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

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

802

For the D15 scheme, we have added descriptions for instruments and measurement 803 locations as: "D15 was developed as a combination of field campaign and laboratory data 804 805 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 806 obtained during the 2007 Pacific Dust Experiment (PACDEX) on the NSF/NCAR G-V 807 aircraft over the Pacific Ocean basin (Stith et al., 2009), and the 2011 Ice in Clouds 808 809 Experiment - Tropical (ICE-T) on the NSF/NCAR C-130 aircraft flown from St. Croix, US Virgin Islands (Heymsfield and Willis, 2014)." 810

811

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".

816

• 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."

821

• 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), SO₂, 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 toM-PACE meteorology.

838

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,

842 which are derived from a present-day CAM6 climatological simulation."

843 "The cloud-borne aerosols will be released to the interstitial aerosols once cloud droplets844 evaporate, which can be re-activated when cloud droplets are nucleated."

845

• 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.

852

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 856 that "Dividing by a factor of 4 seems very approximate to account for shattering effects". 857 We adopted this method since the M-PACE data were collected before the advent of shatter 858 mitigating tips and before algorithms for removing the shattered particles had been 859 860 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 861 that we can get some estimates of the magnitude of the shattering effect on ice number 862 863 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 864 removing the shattered particles. 865

866

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:

876

872

"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."

- 881
- 882



883 884

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

886

• 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

890 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

892 observations in Fig. 2 as:



894

Fig 5: I find the "enhancement ratio" confusing because the relative enhancement in 895 • Figures b-j are compared relative to Figure a, but they all use the same colour bar. Wouldn't 896 it make more sense to use a separate colour scheme for b-j? 897

Reply: We thank the reviewer for the comment. We however, find that it is hard to include 898 two color schemes in Fig. 5. Since we are plotting the bivariate joint probability density 899 functions (PDF) for all the panels, we think that it would be cleaner to use the same color 900 scheme and thus keep Figure 5 unchanged. 901

902

Please include error bars in the observations and all simulations. 903 •

Reply: We thank the reviewer for the suggestion. We have added error bars in the 904 observations and all simulations in Figures 1 and 2, which are shown below. 905



917 crystals that fell from overlying cirrus clouds.

918	Reply: We thank the reviewer for the suggestion. We have added a sentence to discuss the		
919	seeding effect as: "Ice crystals that fall from overlying cirrus clouds can provide another		
920	source of ice in mixed-phase clouds."		
921			
922	• Lines 42-43: Ice crystal fall speed is a cloud microphysical process that is also quite		
923	important for mixed-phase cloud properties such as SLF according to the CAM5 model		
924	shown by Tan & Storelvmo 2016.		
925	Reply: We thank the reviewer for the suggestion. We have modified the sentence as: "In		
926	addition, other microphysical processes such as rain formation, ice growth, and ice		
927	sedimentation are important for mixed-phase cloud properties (Mülmenstädt et al., 2021;		
928	Tan and Storelvmo, 2016)".		
929			
930	• Line 70: "Albeit these studies, how" is grammatically incorrect.		
931	Reply: We thank the reviewer for the suggestion. We have revised the sentence as: "Despite		
932	the above progress, many questions remain unexplored for the Arctic mixed-phase stratus		
933	clouds, e.g., whether PIP always promotes the SIP and how SIP influences the PIP."		
934			
935	• Line 188: "rather than" I think should be "in addition to" since Hallett-Mossop is		
936	included in all simulations?		
937	Reply: Corrected. Thanks.		
938			
939	• Line 248: suggest replacing "in accompany with" with "accompanied by" and again		
940	on line 409.		
941	Reply: Corrected. Thanks.		
942			
943	• Line 370: add "rate" after "nucleation"		
944	Reply: Added. Thanks.		
945			
946	• Lines 423-426: Not necessary to discuss here since there is no associated figure and		
947	discussion and not central to the manuscript?		
948	Reply: We thank the reviewer for the suggestion. These sentences are removed in the		
949	revised manuscript.		
950			
951			