Response to Reviewer 2

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 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.

General comments:

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 campaign using single column mode of CESM2 CAM6 model. They concluded that the simulations using aerosol-aware ice nucleation schemes and including SIP processes 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 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. The reviewer would recommend major revision for this manuscript in case the authors need 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.

Major comments:

1. 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.

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."

Following the reviewer's suggestion, we have replotted Figure 3 in which only ice particles larger than 100 microns are used from simulations, shown as Figure R1 below. The purpose 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 ICNC). The idea is, INPs represent the primary nucleated ice, and the difference between 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: "The purpose of this figure is to examine the relative importance between primary ice nucleation and SIP by comparing INP and ice crystal number concentrations. Therefore, all ice sizes are included in the simulation results".

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.

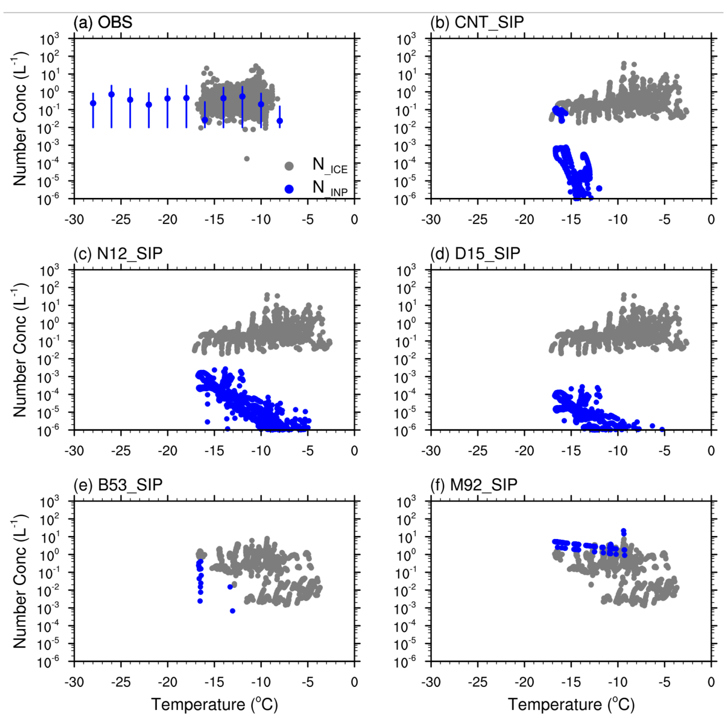

Figure R1. Same as Figure 3 but only shows ice particles with diameters larger than 100 µm from simulations.

3. Lines 199-203: "M-PACE observed ICNCs were scaled by a factor of 1/4", have the data collected by the authors been scaled by a factor to remove the shattering effect during the data quality control? Are the conclusions sensitive to this correction factor?

Reply: We thank the reviewer for the comment. The observed ICNC data we used in this study do not remove the shattering effect during the data quality control, since the ICNCs for M-PACE were measured before anti-shattering algorithms were developed to remove 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 ¼ to the M-PACE observed ICNCs.

We have conducted a sensitivity test with a scaling factor of 1/2 to the observed ICNCs, as 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 some discussions 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."

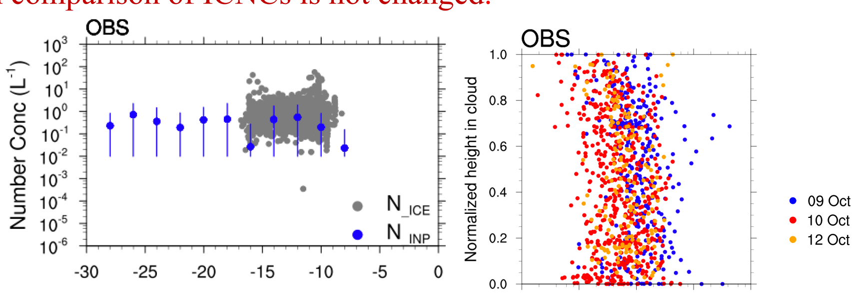

Figure R2. Same as Figure 3a and Figure 4a, but applied a correction factor of 1/2 to the measured ice crystal number concentrations for Figure 3a (left) and Figure 4a (right).

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

We have added a note when we discuss about Figure 2 in the revised manuscript:
"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 than those from the B53 and M92 schemes. With these low INP concentrations, the single-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."

5. 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 measured by a CFDC on board an aircraft (Prenni et al., 2007) during the M-PACE single-layer mixed-phase cloud period. For the B53 scheme in the model, we use Equation 4 to calculate the immersion freezing rate, and diagnose INP number concentrations by multiplying the immersion freezing rate by the model timestep. The contact ice nucleation 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 observations. However, for the single-layer mixed-phase clouds, the immersion freezing is dominated, and the contributions from deposition and contact ice nucleation to total ice production are much smaller (see Figure R3 below).

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, 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."

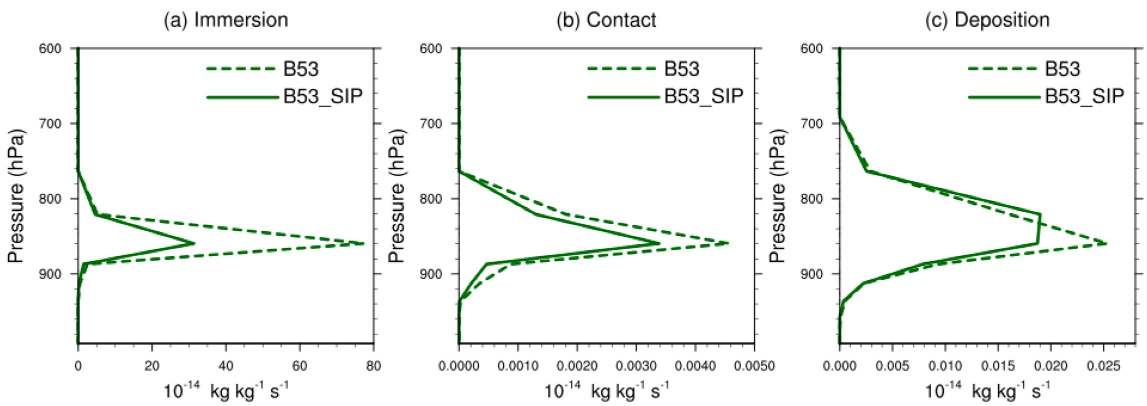

Figure R3. Vertical profiles of (a) ice production rate (unit: kg kg^{-1} s^{-1}) from immersion freezing of cloud water, (b) ice production rate (unit: kg kg^{-1} s^{-1}) from contact freezing of cloud water, and (c) ice production rate (unit: kg kg^{-1} s^{-1}) from deposition nucleation calculated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

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.

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 nucleation number from parameterization and ice particle number at current model time 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 Figure 10 are directly calculated by the CNT ice nucleation parameterization, which are the number of ice crystals nucleated from the parameterization times the initial mass of an ice particle (2.093×10^{-15} 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.

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

Reply: We thank the reviewer for the suggestion. Yes, the production rates in Figures 8-10 are for ice mass, which are calculated from ice production rates for ice number from the parameterizations multiplied by the initial mass of an ice particle (2.093×10^{-15} kg). We added a note in the text: "The ice mass production rates are calculated by multiplying ice number production rates from parameterizations by the initial mass of an ice particle (2.093×10^{-15} kg)."

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 and snow are separated artificially in the microphysics scheme in the model. IIC represents the process that snow particles collide with each other and produce smaller cloud ice 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 converted from snow to cloud ice in the IIC process, although the total ice mass is not changed.

Minor comments:

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

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

"The graupel mass mixing ratio (q_g) is diagnosed as the 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 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."

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

Reply: We thank the reviewer for the suggestion. We have modified the sentences as: "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 the M-PACE period after considering the SIP. In the SIP experiments with the B53 and M92 schemes, however, SIP has a minimal impact on the LWP/IWP. Second, the B53, B53_SIP, M92, and M92_SIP produce the largest IWP (~12 g m^{-2} averaged over the M-PACE period), followed by CNT_SIP, N12_SIP, and D15_SIP (~10 g m^{-2} averaged over the M-PACE period). CNT, N12, and D15 experiments produce the smallest IWP (~5 g m^{-2} averaged over the M-PACE period). These characteristics are also evident in the vertical profiles of LWC and IWC in Fig. 2 and Fig. S2. It indicates that the B53 and M92 nucleation schemes are highly efficient in forming ice; in comparison, the SIP simulations using CNT/N12/D15 ice nucleation schemes show the lower ice production capabilities. B53, B53_SIP, M92, and M92_SIP experiments generate the closest IWP (~12 g m^{-2} averaged over the M-PACE period) compared with the observation (~64 g m^{-2}). However, these four experiments also show substantially low biases of LWP (~40 g m^{-2} compared with 126 g m^{-2} in the observation averaged over the M-PACE period). As shown in Fig. 1 and Fig. S1, the mixed-phase clouds are almost fully glaciated during the single layer stratus period. Therefore, the CNT_SIP, N12_SIP, and D15_SIP experiments give the best simulation results in terms of LWP and IWP during the M-PACE. Adding the SIP does not change the modeled LWP/LWC and IWP/IWC with the B53 and M92 ice nucleation schemes. On the contrary, SIP decreases the LWP/LWC by 38% and doubles the IWP/IWC with the CNT, N12, and D15 ice nucleation schemes."

3. Lines 233-234: "appears an inversely linear relationship", "this relationship is not 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 nucleation schemes (e.g., N12 and D15), there appears an inversely relationship between $\log_{10}(N_{INPs})$ and temperature".

4. Lines 234-238: Please quantify the analyses, e.g., "reduces dramatically", "much 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 ºC, from ~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"

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?

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 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 4.3 (Figure 10). The reason for the inactive SIP in B53_SIP/M92_SIP is because of the competition between PIP and SIP (Figure 10). Too strong primary ice nucleation in 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 explanation in Section 4.3 and Figure 10).

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."

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)."

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".

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

9. Lines 294-308: It is confusing whether the authors talked about ice number or mass in Figure 7. If the authors talked about ice mass in Figure 7, how do IIC contribute to ice 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 (2.093×10^{-15} 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 (2.093×10^{-15} kg)."

IIC represents the process that bigger snow particles collide with each other and produce smaller ice particles due to fragmentation. In the model, IIC process rate indicates a mass 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.

10. Line 328: Please quantify "substantially weakened"

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

11. Lines 342-343: Based on Eq. (5), M92 seems dependent on supersaturation not 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.

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

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 M92_SIP compared to CNT_SIP (with the peak value of 10 kg kg^{-1} s^{-1}) is a result of smaller graupel mass mixing ratio in M92_SIP (with the peak value of 1.4 mg kg^{-1} in M92_SIP versus 5.2 mg kg^{-1} in CNT_SIP) (Fig. 10g). As the graupel mass is diagnosed from the cloud water mass, snow mass, and temperature, smaller mass mixing ratios of cloud water (with the peak value of 8 versus 125 mg kg^{-1} in Fig. 10h) and snow (with the peak value of 1.4 versus 2.3 mg kg^{-1} in Fig. 10i) in M92_SIP eventually lead to a smaller graupel mass mixing ratio and a smaller graupel-related IIC rate. Similar results can be found with the other ice nucleation schemes."

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:

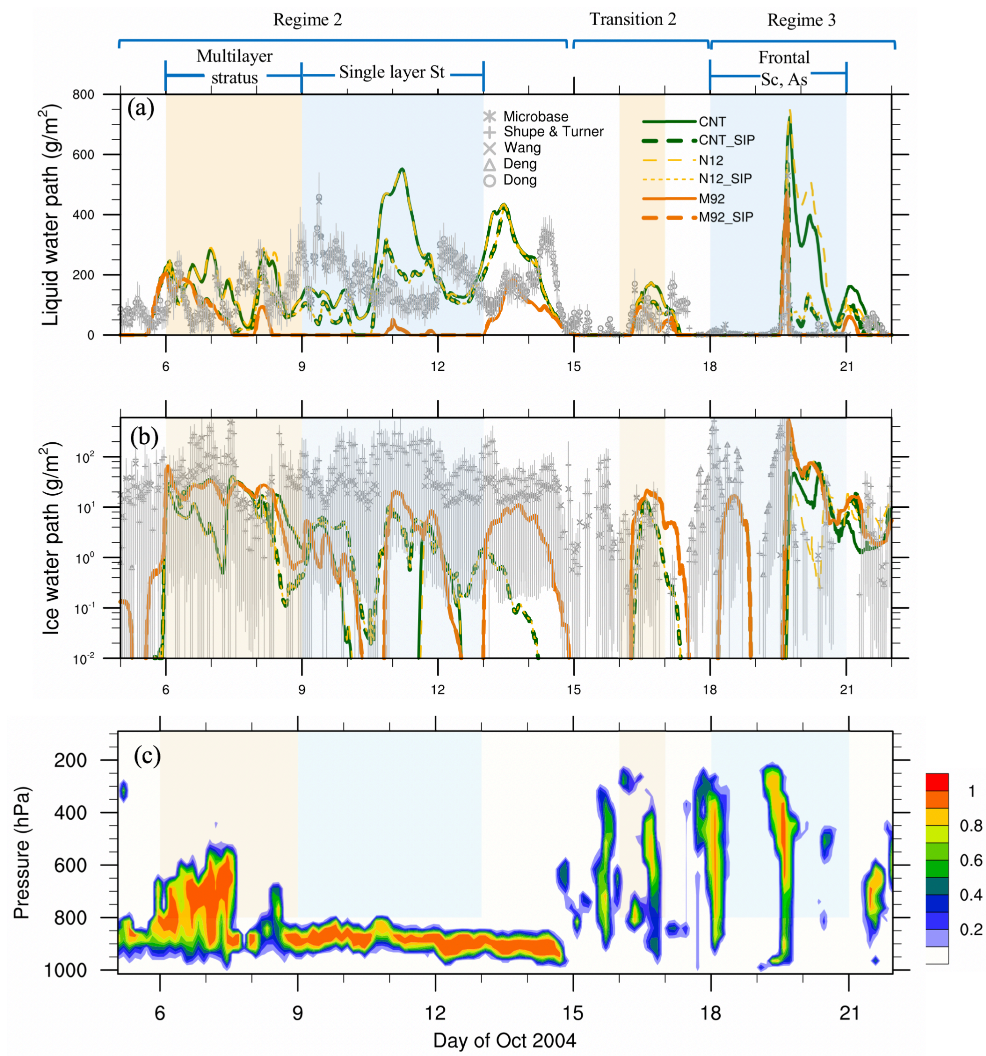

Figure R4.

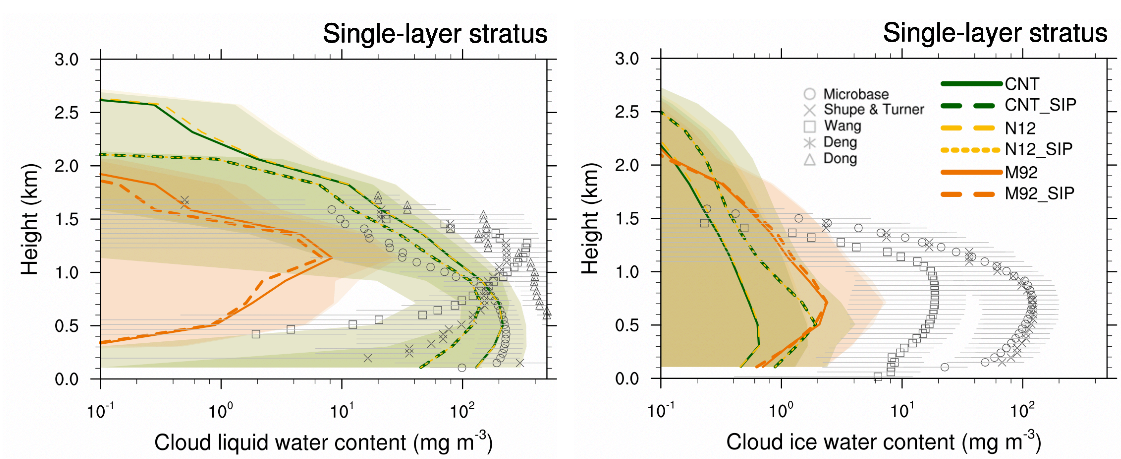

Figure R5.

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

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

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

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.

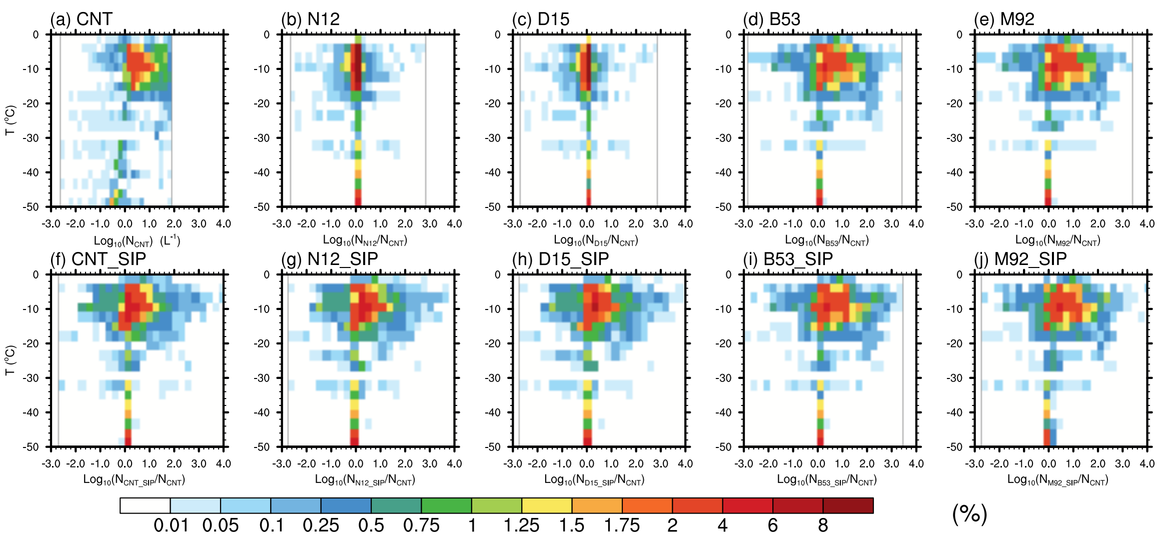

Figure R6.

16. Figure 7: “total ice production rate”, is the “production rate” for mass or number?

Reply: We thank the reviewer for the suggestion. We output PIP and SIP number process rates, and multiple them by the initial mass of an ice particle (2.093×10^{-15} kg) to calculate the ice mass production rates used in Figure 7. We added a note in the text: “The ice mass production rates are calculated by multiplying ice number production rates from parameterizations by the initial mass of an ice particle (2.093×10^{-15} kg).”

17. 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 cloud water by snow, and does not include the accretion of rain by snow, since the purpose of this figure is to illustrate that a stronger "accretion rate of cloud water by snow" (8 vs. 2 kg kg^{-1} s^{-1}) results in a lower cloud water amount (13 mg kg^{-1}) in the CNT_SIP experiment compared with that (23 mg kg^{-1}) in the CNT experiment.

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

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.