Response to Reviewer 1

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

In this work, the authors contrasted several parameterizations of primary ice production (PIP), combined with a new set of parameterizations of secondary ice production (SIP) in the NCAR CESM2/CAM6 model. The model simulations are compared with observations from the DOE M-PACE campaign. The scientific questions include: What are the impacts 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.

Main comments:

1. About the comparison of ice crystal number concentration (ICNC) between observations 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, the simulations may overestimate ICNC when a wider size range is used. The reviewer suggests a revision on the simulation dataset to delete ice particles < 100 micron. In 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 around 1 to 1/4.5. Thus, using 1/4 seems to provide a lower end of ICNC from observations. 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.

Reply: We thank the reviewer for the suggestion. Following the reviewer's suggestion, we have replotted Figure 3 in which only ice particles larger than 100 microns are used, shown as Figure R1 below. The purpose of Figure 3 is to examine the relative importance between primary nucleation and SIP by comparing INP and ice number concentrations. The idea is, 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 crystal number concentrations between observations and simulations. Therefore, we used all sizes of ice crystals from the simulations in Figure 3 in the manuscript. 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 in the manuscript already uses the simulated ice larger than 100 microns.

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:

"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."

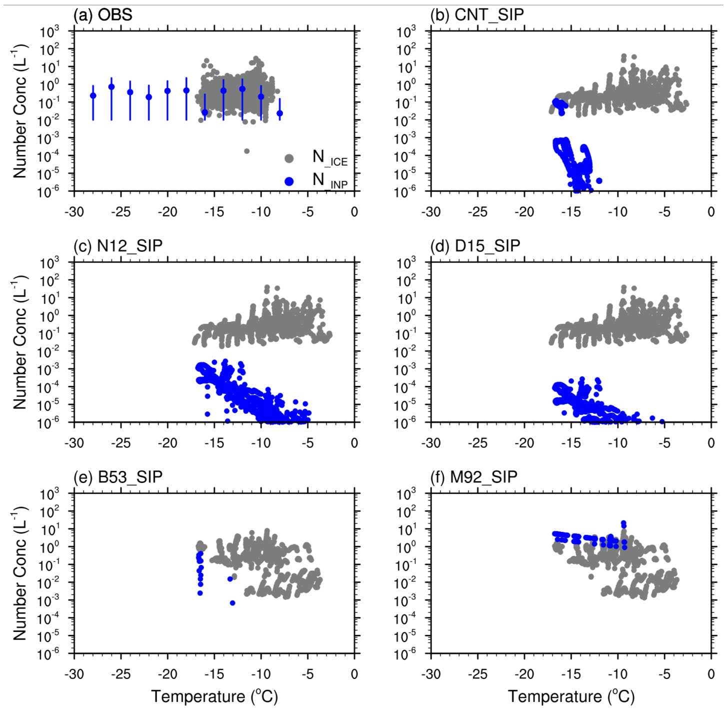

Figure R1. Same as Figure 3 but only shows ice particles with diameters larger than 100 μm from all the simulations.

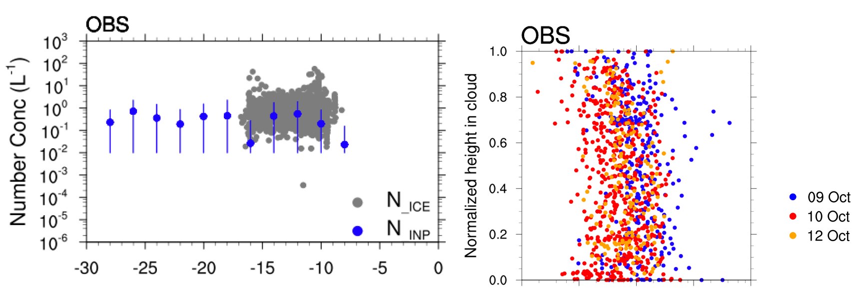

Figure R2. Same as Figure 3a and Figure 4a, but applied a correction factor of 1/2 to the measured ice crystal number concentrations for Figure 3a (left) and Figure 4a (right).

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?

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.

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".

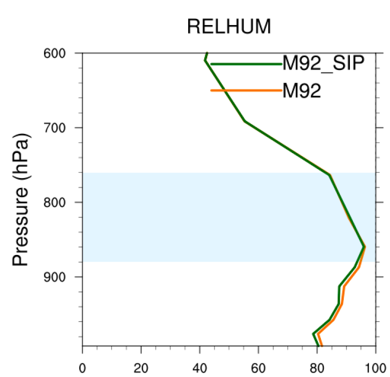

Figure R3. Relative humidity with respect to ice (RHice) from M92 and M92_SIP experiments averaged over the single-layer mixed-phase cloud period.

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.

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 single-layer mixed-phase clouds (Figure 8)."

3. Following the previous comments on Figure S7, some parts of this figure do not make 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 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 significantly increases in N12_SIP near the surface around 980 – 1000 hPa. This pressure level does not match with the location of changes seen in panel (d), and the increasing deposition doesn't explain the increase of accumulation mode dust at 880 – 700 hPa as mentioned above. The change of coarse mode deposition also doesn't match with the vertical locations of changes seen in coarse mode dust. Can the authors explain this figure 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.

Reply: We thank the reviewer for the helpful comment and suggestion. We have taken a more careful look at the changes of accumulation mode dust number concentration in 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 much less to primary ice nucleation than the coarse mode dust. Therefore, we have removed 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 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 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 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:

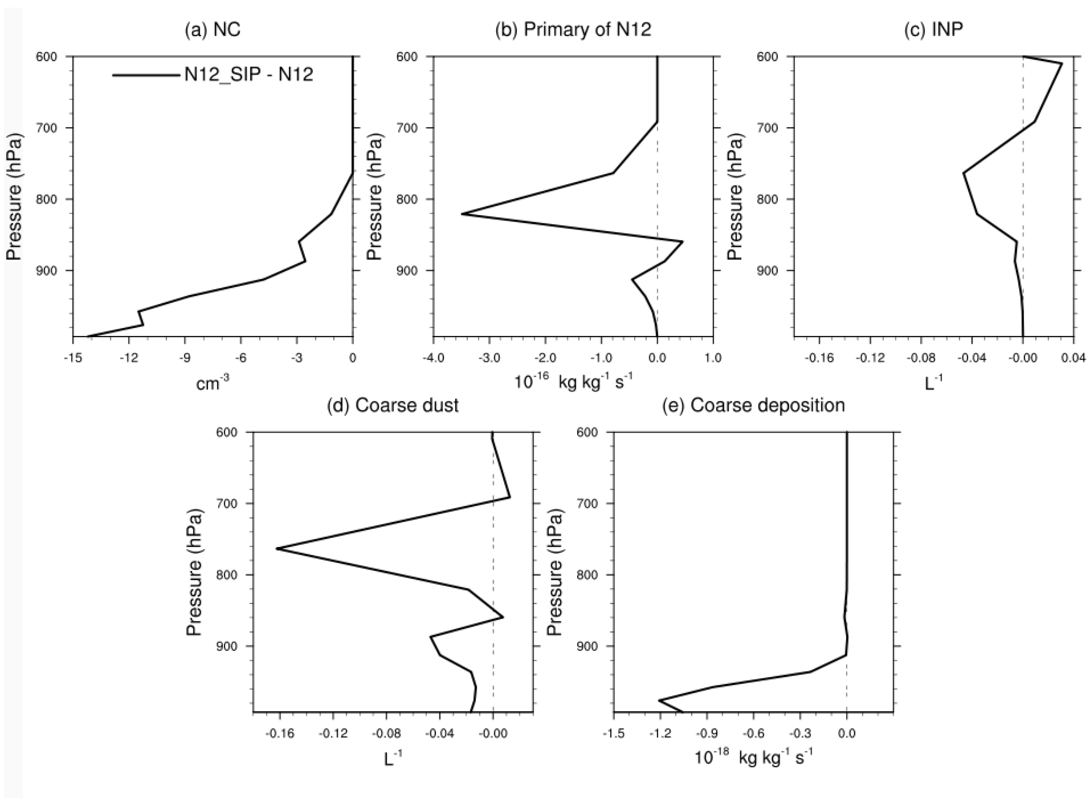

Figure R4. Vertical profiles of differences of (a) cloud droplet number concentration, (b) ice production rate from immersion freezing of cloud droplets, (c) immersion freezing INP number concentration, (d) interstitial dust number concentration in the coarse mode, and (e) wet deposition rate of interstitial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

4. In several analyses, the authors use relative altitude to the cloud layer, that is, 0 refers to cloud base and 1 refers to cloud top. There is no discussion about how this relative altitude is derived. Is it derived based on ground-based observations or in-situ observations? Please clarify.

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

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

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

Minor comments:

1. 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 schemes differ in details about temperature and aerosol dependences, CNT, N12, and D15 predict much lower INP concentrations for the M-PACE single-layer clouds compared with the B53 and M92 schemes. With low INP concentrations, modeled clouds are 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 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."

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

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 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., 2007; Wang, 2007; Khanal and Wang, 2015). The INP concentrations are based on in-situ observations by a CFDC on board an aircraft (Prenni et al., 2007). The ICNCs and cloud phase are based on in-situ observations and provided by McFarquhar et al. (2007)." in Section 3 to clarify the types of observation data used in the analyses.

3. Please clarify how the variables related to "rate" are defined in the model. For example, is the variable ice production rate describing the amount of ice crystals (in kg) being produced in every unit mass of dry air (in kg) per second in the entire grid box, or only in 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.

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

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

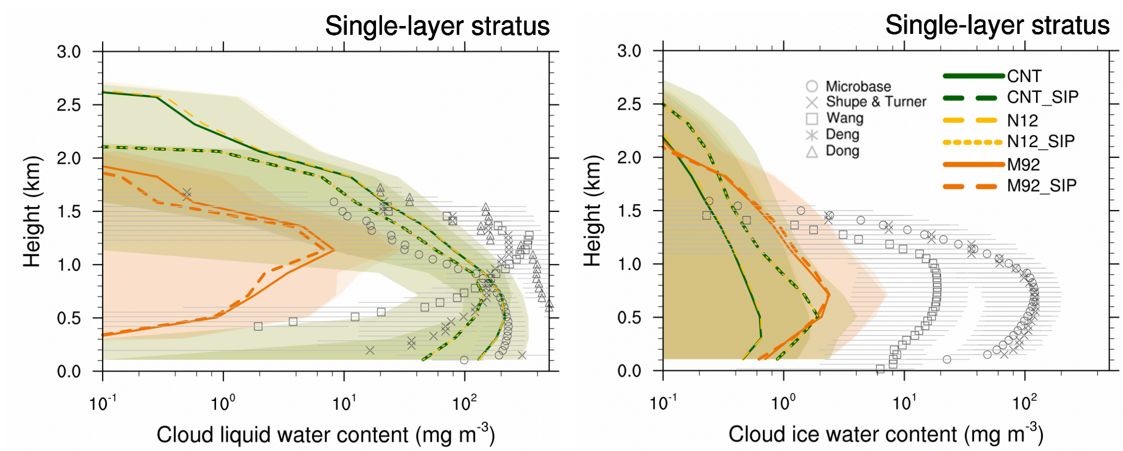

Figure R5. Vertical profiles of LWC (left) and IWC (right) during the single-layer mixed-phase cloud period (October 9-12) from CNT, CNT_SIP, N12, N12_SIP, M92, and M92_SIP experiments and from remote sensing retrievals (symbols). Horizontal gray lines represent standard deviations of retrieval data,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

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?

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 increasing dust aerosol concentrations used in the INP parameterizations, which will also result in more INPs. We have conducted a sensitivity test using the CNT scheme with increased dust concentrations by 100 times. This simulation shows overall similar cloud properties, but the relative contribution of primary ice nucleation to total ice production is increased by a factor of ~2 during M-PACE. We have added a discussion on this in Section 4.2 in the revised manuscript as: "Since the INP number concentrations in CNT, N12 and D15 are significantly lower than the observations (Figure 3), a sensitivity test using the CNT scheme with increased dust concentrations by 100 times shows overall similar cloud properties. However, the relative contribution of primary ice nucleation to total ice 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."

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.

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 shows the probability of occurrence defined in terms of both temperature and ice number concentration for Figure 5a (or enhancement ratio for Figure 5b-j), which means PDF is calculated by the number of samples of each bin divided by the total number of samples of the entire temperature range (i.e., the sum of % in all bins equals one). We have revised 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 experiment, and (b)-(j) in terms of both temperature and enhancement ratio of ice crystal number concentration from the respective experiment to that from the CNT experiment. A logarithmic scale is used for the x-axis."

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:

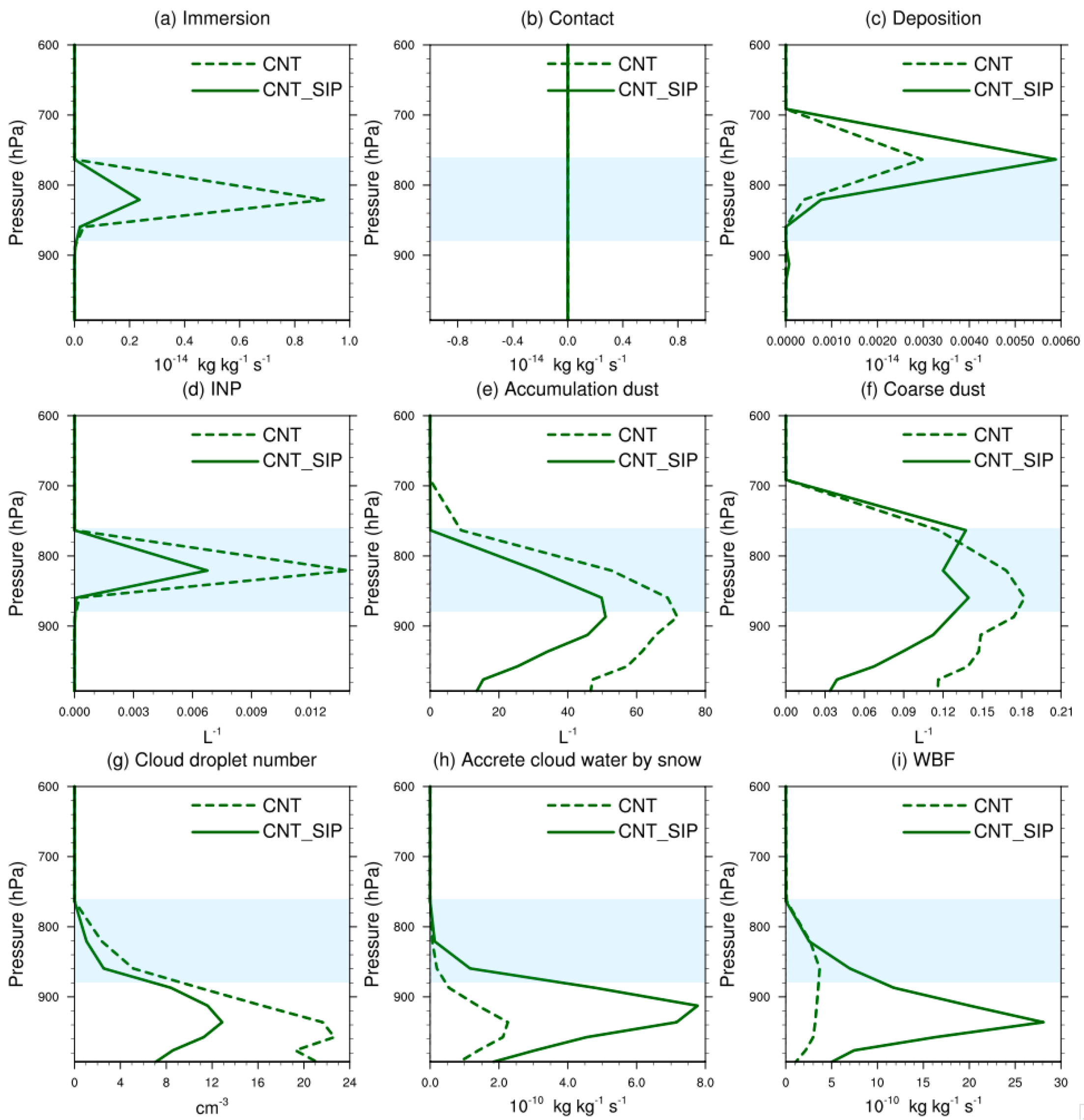

Figure R6. Vertical profiles of (a) ice production rate (unit: kg kg^{-1} s^{-1}) from immersion freezing of cloud water, (b) ice production rate (unit: kg kg^{-1} s^{-1}) from contact freezing of cloud water, (c) ice production rate (unit: kg kg^{-1} s^{-1}) from homogeneous and heterogeneous deposition nucleation, (d) immersion freezing INP number concentration, (e) cloud-borne dust number in the accumulation mode, (f) cloud-borne dust number in the coarse mode, (g) cloud droplet number concentration, (h) accretion rate of cloud droplets by snow, and (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 production rates are grid-box means.