Response to Reviewer 1

We thank the anonymous reviewer for his/her careful reading and constructive comments on 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.

This manuscript implements new parameterizations of secondary ice production (SIP) including frozen raindrop shattering, ice-ice collisional breakup and the classic Hallett-Mossop parameterization by Cotton et al. 1986 in the CAM6 single column model (SCM) to determine the impact of SIP on Arctic mixed-phase cloud properties compared to observations from the M-PACE field campaign. The authors conclude that SIP reduces the high supercooled liquid bias in Arctic single-layer stratiform mixed phase clouds and improves the low bias in ice crystals at relatively warm temperatures below the supercooled liquid clouds. They also find that the fragmentation of freezing droplets contributes most to ice production in single-layer boundary layer clouds, while ice-ice collisions and rime splintering contribute relatively less to ice production in frontal clouds and multilayer stratus clouds, respectively, and primary ice production is more important for cold multilayer and frontal clouds in CAM6 SCM.

The manuscript presents novel results, is important for the improvement of climate models and is very relevant to Atmospheric Chemistry and Physics. I would recommend publication after major revisions.

Reply: We thank the reviewer for the encouraging comments. We have revised the manuscript following your comments regarding the uncertainties of observation data and clarified the text to improve the paper.

My most major concern is regarding the M-PACE in situ data that was compared against the CAM6 SCM. These data did not correct for the shattering effect which is known to severely overestimate the ice number concentration by up to two orders of magnitude and likely cause misleading conclusions if not accounted for (Korolev et al. 2011, Korolev et al. 2013a, Korolev et al. 2013b, Korolev & Field 2014). I strongly recommend that the authors use data that have corrected for the shattering effect using both correction algorithms using interarrival time and data that have used antishattering tips.

Also, despite apparent better agreement with M-PACE (noting that the M-PACE data for ice number concentration are incorrect), the poor agreement with ice properties were not noted in the conclusions and abstract and underemphasized in the manuscript. Please revise accordingly.

Reply: We thank the reviewer for the constructive comment. We agree with the reviewer that the M-PACE in situ ice number data did not correct for the shattering effect, because the data were collected before the advent of shatter mitigating tips and before algorithms for removing the shattered particles had been developed. Thus, there were no corrections for the shattering in these data. We have discussed this issue with Greg McFarquhar who collected the M-PACE data. He suggested that we can get some estimates of the magnitude of the shattering effect on ice number concentrations from other campaigns, such as ISDAC, IDEAS-2011, and HOLODEC, which also used the 2DC cloud probe, but adopted anti-shattering tips and algorithms for removing the shattered particles.

Previous studies indicated a reduced ice number by 1-4.5 times and up to a factor of 10 depending on particle size for IDEAS-2011 and ISDAC after using the antishattering 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.

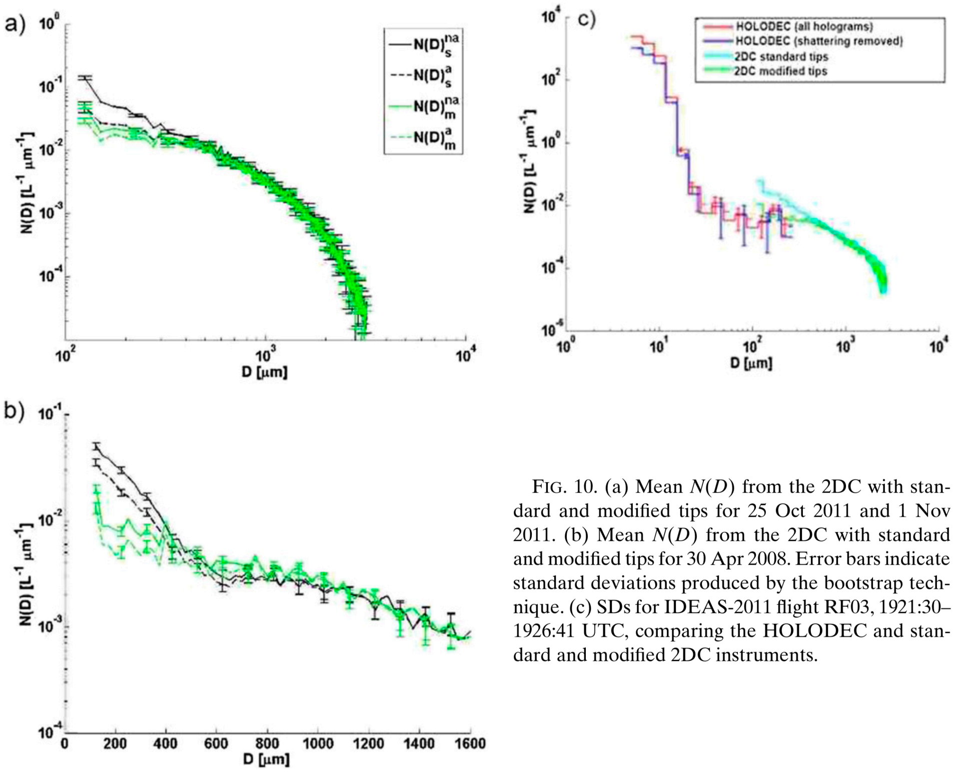


FIG. 10. (a) Mean $N(D)$ from the 2DC with standard and modified tips for 25 Oct 2011 and 1 Nov 2011. (b) Mean $N(D)$ from the 2DC with standard and modified tips for 30 Apr 2008. Error bars indicate standard deviations produced by the bootstrap technique. (c) SDs for IDEAS-2011 flight RF03, 1921:30–1926:41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

Based on the reviewer's comments, we added a discussion about the M-PACE observed ice number data in Section 3.2 as "However, the M-PACE data were collected before the advent of shatter mitigating tips and before algorithms for removing the shattered particles had been developed. Thus, there are no corrections for the shattering effect in these data. Previous studies indicated an averaged reduction of ice number concentrations by 1-4.5 times and up to a factor of 10 (for some data samples) in other field campaigns, such as Instrumentation Development and Education in Airborne Science 2011 (IDEAS-2011), Holographic Detector for Clouds (HOLODEC), and Indirect and Semidirect Aerosol Campaign (ISDAC), which also used the 2DC cloud probe, but adopted anti-shattering tips and algorithms for removing the shattered particles (Jackson and McFarquhar, 2014; Jackson et al., 2014).

In order to account for the anti-shattering effect, the observed ice number was scaled by a factor of 1/4 and 1/2, respectively to consider the possible range of the shattering effect. Furthermore, to be consistent with Figure 10 in Jackson et al. (2014), only ice particles with diameters larger than 100 μm are included and shown in our model-observation comparison."

We also added the following sentences in the section 4.3.2: "We note that the agreement between observed and modeled ice number concentrations is improved, as shown in Figures 10 and 11 with a correction factor of ¼ and in supplementary Figures S2 and S4 with a correction factor of 1/2, compared to that without the correction factor (Supplementary Figures S1 and S3). This is because all model simulations including SIP_PHIL underestimate the observed ice number concentrations without the correction of the shattering effect."

The original Figures 10 and 11 without the correction of observed ice number are moved to the supplement (now Supplementary Figures S1 and S3).

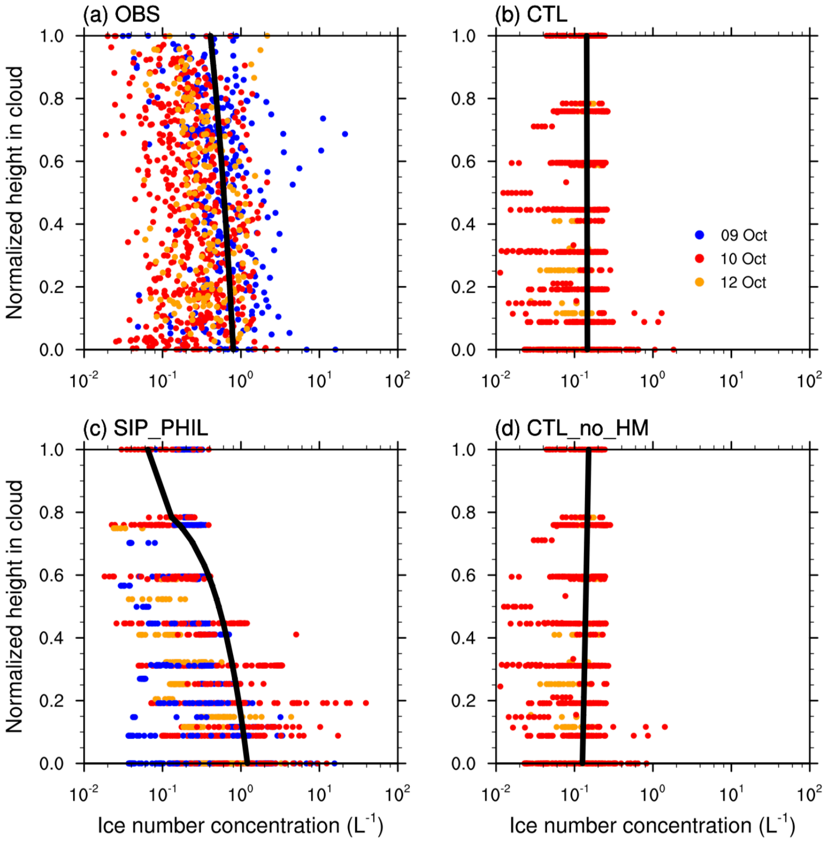


Figure 10. Ice number concentrations as a function of normalized cloud height from cloud base from (a) observation, (b) CTL, (c) SIP_PHIL, and (d) CTL_no_HM. Black solid lines show the linear regression between ice number concentration and height. Only ice particles with diameters larger than 100 μm from observations and model simulations are included in the comparison. A correction factor of ¼ is applied to the observed ice number concentrations in (a).

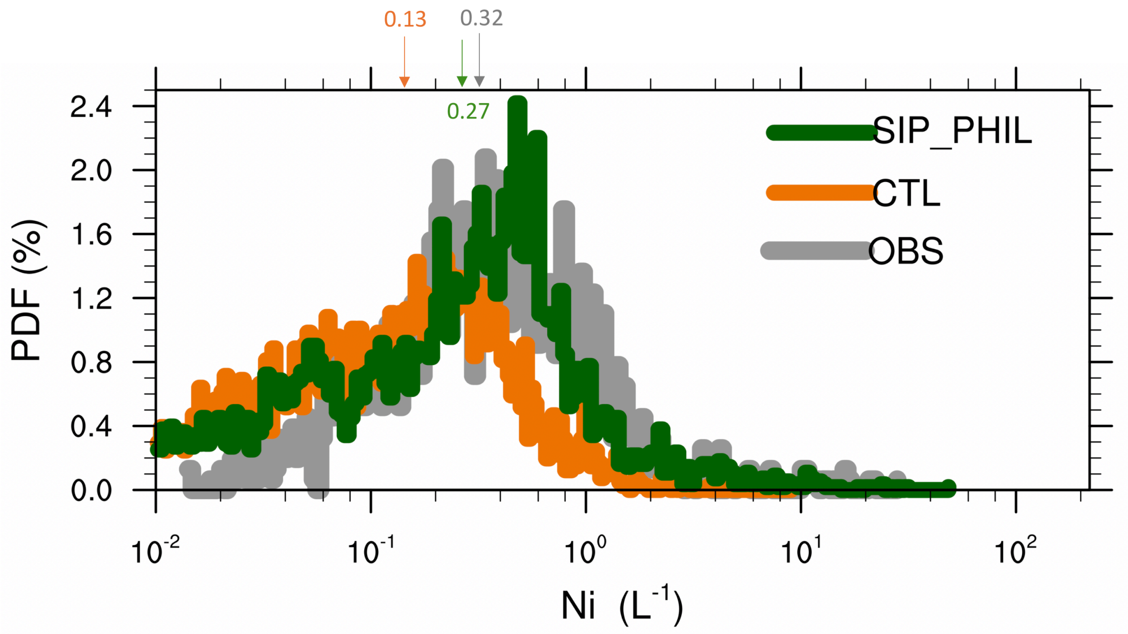


Figure 11. The probability density function (PDF) of ice crystal number concentrations from observation (gray line), CTL (orange line), and SIP_PHIL simulations (green line). The arrow indicates the median of each distribution which means that the set of values less (or greater) than the median has a probability of 50%. Only ice particles with diameters larger than 100 μm from observations and model simulations are included in the comparison. A correction factor of ¼ is applied to the observed ice number concentrations.

Minor revisions:

- Section 4.1.1: A discussion of why the ice properties are so poorly represented in the model is much needed. Please include.

Reply: We thank the reviewer for the suggestion. We have added a detailed discussion of why the ice properties are so poorly represented in the model in Section 5 conclusions as: "The representation of ice properties is highly simplified in the current model. First of all, ice particles in nature are featured with continuous size distributions with complex shapes and a wide range of densities. In contrast, the current model artificially classifies them into two categories (i.e., cloud ice and snow) with fixed densities, e.g., densities of 500 kg m^{-3} for cloud ice and of 250 kg m^{-3} for snow. Moreover, the shape of all ice particles is assumed to be spherical. The parameters, *a* and *b* in the relationship of terminal velocity and diameter (V-D, $V=aD^b$) are fixed values for cloud ice and snow. These assumptions cannot represent the complexities of ice properties (e.g., size distribution, density, shape, and fall speed) in the measurement. Furthermore, the riming intensity of ice particles changes as ice collides with supercooled liquid, leading to significant changes in density and fall speed of ice. This evolution of ice properties is currently not represented in the model. A promising method is to represent the ice-phase microphysics with varying ice properties (Morrison and Milbrandt, 2015; Zhao et al., 2017)."

- What are the initialization and forcing conditions of the model?

Reply: We thank the reviewer for the suggestion. We have added a brief explanation about the initialization and forcing data in Section 3.3 as: "The model is initialized and driven by the large-scale forcing data at every 3 hours. The forcing data which were developed by Xie et al. (2006) include divergences and advections of moisture and temperature as well as the surface flux."

- The title would be more accurate if "CAM6 single column model" is used in place of CESM2. Please modify.

Reply: We thank the reviewer for the suggestion. We have modified the title as: "Impacts of Secondary Ice Production on Arctic Mixed-Phase Clouds based on ARM Observations and CAM6 Single-Column Model Simulations"

- Please include more information about the formulation of the Hallett-Mossop parameterization.

Reply: We thank the reviewer for the suggestion. We have added more detailed description of the Hallett-Mossop parameterization in the revised manuscript as:

"The ice number production rate is based on the parameterization of Cotton et al. (1986), which is given as:

$$N_{HM} = C_{sip\_HM} \times p_{sacws} \tag{11}$$

where p_{sacws} is the riming rate of cloud droplets by snow and is expressed as:

$$p_{sacws} = \frac{\pi \times a_{vs} \times \rho \times N_{0s} \times E_{ci} \times \Gamma(b_{vs}+3)}{4 \times \lambda^{b_{vs}+3}} \tag{12}$$

in which E_{ci} is the collection efficiency for the riming of cloud droplets by snow based on Thompson et al. (2004), a_{vs} and b_{vs} are the fall speed parameters for snow particles, $b_{vs} = 0.41$, and $a_{vs} = 11.72 \times \frac{\rho_{850}}{\rho}$, ρ and ρ_{850} are the air density and the typical air density at 850 hPa, respectively, and N_{0s} and λ are the parameters for the snow particle size distribution.

The conversion coefficient C_{sip_HM} in Equation (11) depends on temperature T_c in ºC:

$$C_{sip\_HM} = \frac{3.5 \times 10^8 \times (-3 - T_c)}{2}, \text{ when } -5 < T_c < -3, \text{ and} \tag{13}$$

$$C_{sip\_HM} = \frac{3.5 \times 10^8 \times (T_c - (-8))}{3}, \text{ when } -8 < T_c < -5 \tag{14}$$

The production rate for cloud ice mixing ratio is given as:

$$P_{HM} = N_{HM} \delta m_{ice} \tag{15}$$

in which δm_{ice} is mass for a single ice particle in the HM process, prescribed as 2.09×10^{-15} kg."

- Lines 35-39: not only is the cloud radiative effect important but also the impact of Arctic cloud properties in climate change scenarios: Vavrus 2004, Zhang et al. (2018),Tan & Storelvmo 2019.

  Reply: We agree with the reviewer's comment, and have added a sentence as: "Cloud properties further play a key role in the Arctic climate change through cloud feedbacks (Vavrus, 2004; Zhang et al., 2018; Tan and Storelvmo, 2019)."

- Line 422: there are more references related to this than the single ones mentioned for the CAM3/CAM5 model: e.g. Klein et al. 2009, Cesana et al. 2015, Tan & Storelvmo 2016, Zhang et al. 2019, Tan & Storelvmo 2019.

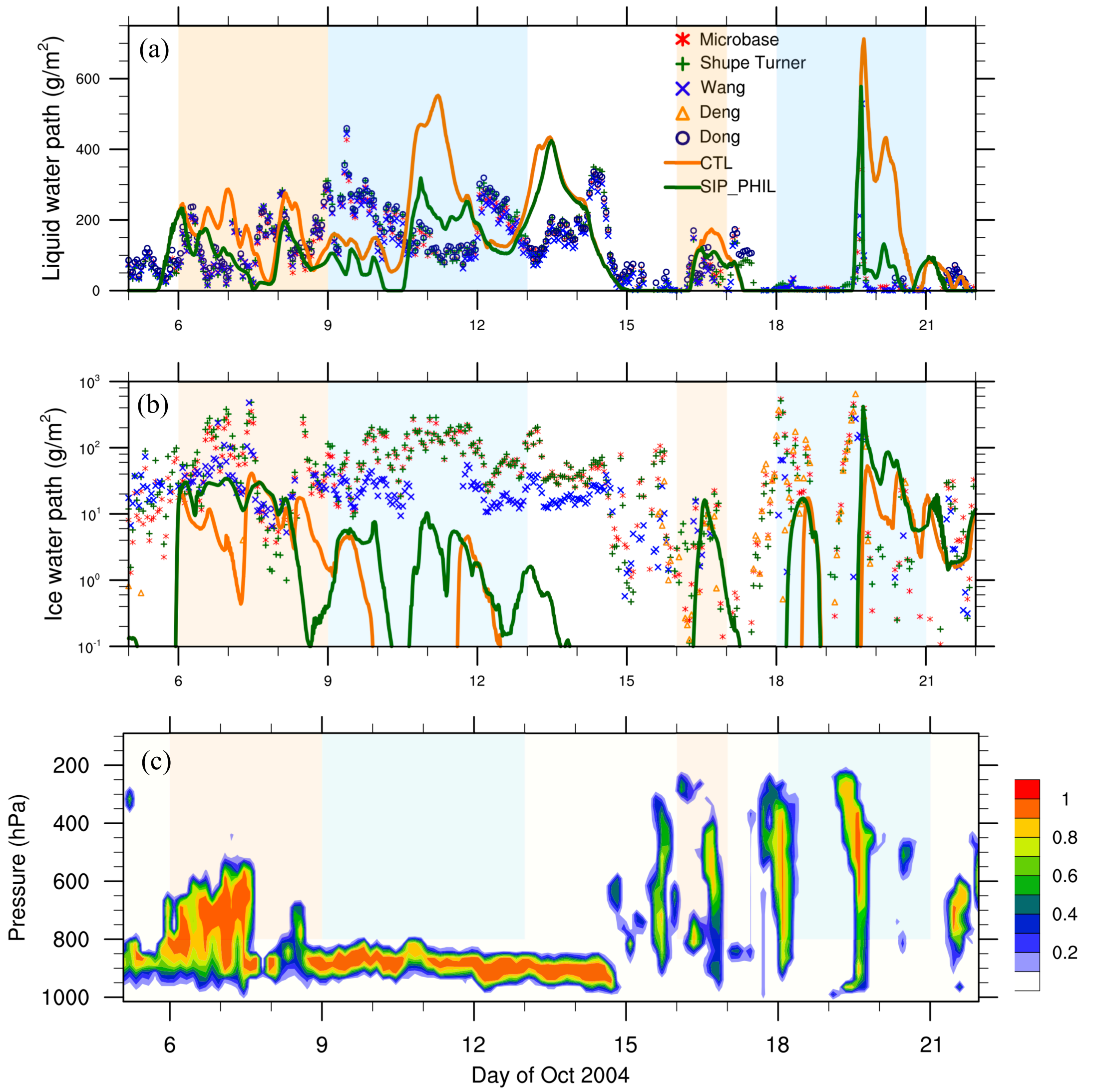
  Reply: We thank the reviewer for the suggestion. We have added more references based on your comments as: "CAM5 showed an underestimation of the liquid fraction (Liu et al., 2011; Cesana et al., 2015; Tan and Storelvmo, 2016; Zhang et al., 2019; Tan and Storelvmo, 2019),"

- Figure 4a: Why does LWP decrease in the SIP_PHIL experiment? Is this related to the Bergeron-Findeisen process?

  Reply: We thank the reviewer for the suggestion. Yes, as you suggested, the decrease of LWP is related to the Bergeron-Findeisen process. Even though SIP is not the direct reason for the LWP decrease, it triggers a chain of microphysical process interactions. We have examined the SIP-induced LWP and IWP changes based on the budget analyses of cloud hydrometeors in Zhao and Liu (2021, submitted).

- Figure 4b: consider using a nonlinear scale to improve visibility of small values.

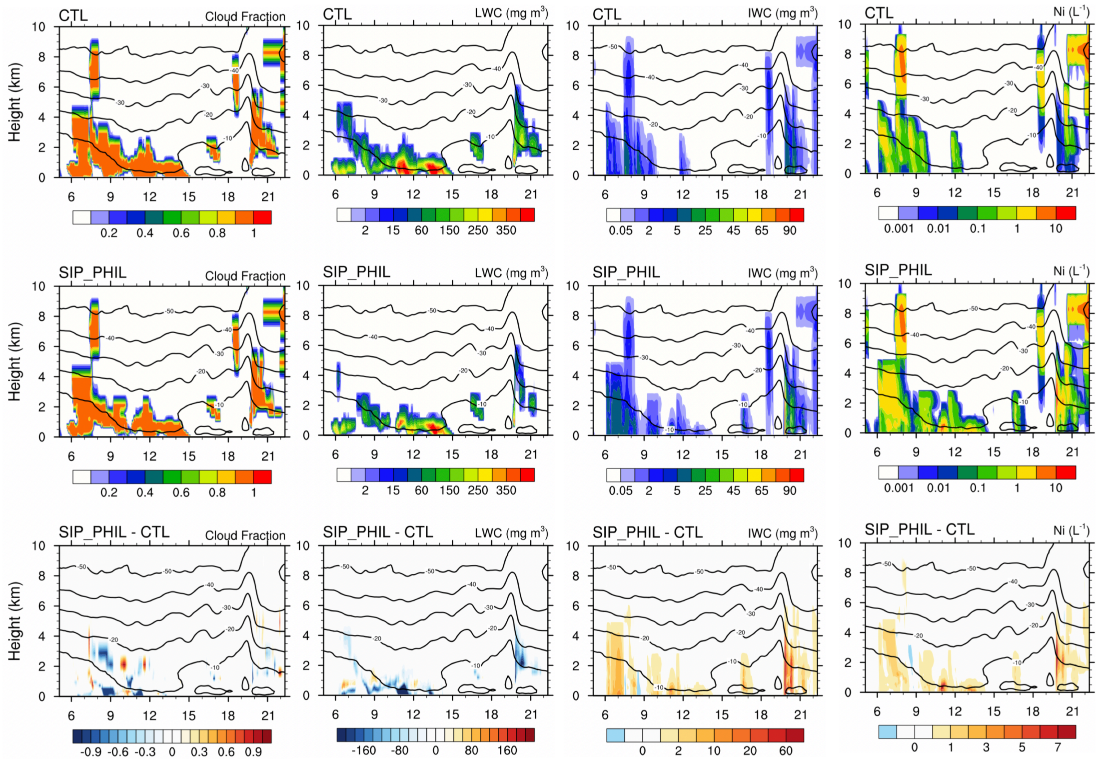
Reply: We thank the reviewer for the suggestion. We have revised Figure 4b according to your suggestion as:



- Figure 5 and lines 352-353: To my eye, it is not clear that the "decoupling" is much improved in the SIP simulation; also has a typo in bottom row, should be "LWC", CF is not labelled in first column.

Reply: We thank the reviewer for the suggestion. We have revised the sentence by removing "decoupling" in the sentence: "This bias is alleviated in SIP_PHIL during 8-12 October (Fig. 5)."

We have also corrected the typo and added the "Cloud Fraction" label in the first column of Figure 5:



- I suggest Figure 12 go to the Supplementary Info.

Reply: Following the reviewer's suggestion, we have moved Figure 12 to the Supplementary Information as Figure S5.

- In terms of the writing style, in general, there are too many short subsections that might be better combined into a broader section. Also, the grammar could improve.

Reply: We thank the reviewer for the suggestion. Following your comment, we have combined section 4.1.1, section 4.1.2, section 4.1.3, section 4.1.4, and section 4.1.5 into a single section 4.1 in the revised manuscript.

References:

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Zhao, X., Lin, Y., Peng, Y., Wang, B., Morrison, H. and Gettelman, A.: A single ice approach using varying ice particle properties in global climate model microphysics, Journal of Advances in Modeling Earth Systems, 9(5), 2138–2157, https://doi.org/10.1002/2017MS000952, 2017.