1	Impacts of Secondary Ice Production on Arctic Mixed-Phase
2	Clouds based on ARM Observations and CAM6 Single-
3	Column Model Simulations
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10	Abstract. For decades, measured ice crystal number concentrations have been found to be orders
11	of magnitude higher than measured ice nucleating particle number concentrations in moderately
12	cold clouds. This observed discrepancy reveals the existence of secondary ice production (SIP) in
13	addition to the primary ice nucleation. However, the importance of SIP relative to primary ice
14	nucleation remains highly unclear. Furthermore, most weather and climate models do not represent
15	well the SIP processes, leading to large biases in simulated cloud properties. This study
16	demonstrates a first attempt to represent different SIP mechanisms (frozen raindrop shattering, ice-
17	ice collisional break-up, and rime splintering) in a global climate model (GCM). The model is run
18	in the single column mode to facilitate comparisons with the Department of Energy (DOE)'s

Atmospheric Radiation Measurement (ARM) Mixed-Phase Arctic Cloud Experiment (M-PACE)
observations.

21	We show the SIP importance in the four types of clouds during M-PACE (i.e., multilayer clouds,
22	single-layer stratus, transition and frontal clouds), with the maximum enhancement in ice crystal
23	number concentrations by up to 4 orders of magnitude in moderately supercooled clouds. We reveal
24	that SIP is the dominant source of ice crystals near the cloud base for the long-lived Arctic single-
25	layer mixed-phase clouds. The model with SIP improves the occurrence and phase partitioning of
26	the mixed-phase clouds, reverses the vertical distribution pattern of ice number concentrations, and
27	provides a better agreement with observations. The findings of this study highlight the importance
28	of considering the SIP in GCMs.

31 **1 Introduction**

32 Clouds play a critical role in the surface energy budget of the Arctic, thereby 33 affecting the Arctic sea ice and regional climate (Kay et al., 2009; Bennartz et al., 2013). 34 Clouds occur frequently in the Arctic (Beaufort Sea) with an observed annual mean 35 occurrence of 85%, a maximum of 97% in September, and a minimum of 63% in February 36 (Intrieri et al., 2002). Along with the occurrence frequency, the phase partitioning between 37 liquid and ice in mixed-phase clouds, i.e., the clouds where liquid and ice coexist at 38 subfreezing temperatures, is also important, since even a small amount of liquid content in 39 clouds can substantially change the radiative properties of the cloud (Shupe et al., 2004; 40 Cesana and Chepfer, 2013). Shupe et al. (2006) showed that over the Beaufort Sea, 59% 41 of observed clouds were mixed-phase, while another study indicated 90% over the western 42 Arctic Basin (Pinto, 1998). Cloud properties further play a key role in the Arctic climate 43 change through cloud feedbacks (Vavrus, 2004; Zhang et al., 2018; Tan and Storelymo, 44 2019).

Mixed-phase clouds are microphysically unstable. Even a small amount of cloud ice can glaciate the mixed-phase clouds in a few hours via the Wegener–Bergeron– Findeisen (WBF) mechanism (Morrison et al., 2012). Mixed-phase clouds in the Arctic are long-lived and characterized by a structure with liquid water at the cloud top and ice water underneath. Interaction and feedback among multiple processes, including longwave

50	radiative cooling, turbulence entrainment, and condensation of liquid water, provide
51	sufficient moistening and cooling at the cloud top. This sustains enough formation of liquid
52	mass against the depletion by the WBF process. In order to support the self-maintenance
53	of liquid water, low concentrations of small ice particles must be present near the cloud
54	base (Shupe et al., 2006; Korolev and Field, 2008). In this way, they are efficient in
55	sedimentation (Jiang et al., 2000) but less active in the WBF and vapor deposition
56	processes. Previous studies indicated that 90% of Arctic mixed-phase cloud temperatures
57	were between -25°C and -5°C from an annual mean perspective (Shupe et al., 2006),
58	indicating that ice exists in moderately supercooled clouds. However, the mechanisms
59	contributing to ice formation in these clouds are still unclear (Shupe et al., 2006; Morrison
60	et al., 2012). One objective of this study is to better understand the ice formation processes
61	in the Arctic mixed-phase clouds.

62 Previous studies have shown the important role of SIP in the Arctic clouds from observations (Schwarzenboeck et al., 2009) and small-scale model simulations 63 64 (Sotiropoulou et al., 2020a; Fu et al., 2019). Using a large-eddy simulation (LES) model 65 and a Lagrangian parcel model, Sotiropoulou et al. (2020a) found that a combination of 66 ice-ice collisional fragmentation and rime splintering provides a better agreement of the 67 simulated ice crystal number concentrations (ICNCs) with observations in the summer Arctic stratocumulus. They found a low sensitivity of SIP to prescribed number 68 concentrations of cloud condensation nuclei (CCN) and ice nucleating particles (INPs). Fu 69

et al. (2019) simulated an autumnal Arctic single-layer boundary-layer mixed-phase cloud using the Weather Research and Forecasting (WRF) model and showed that the model without considering SIP needs an increase of INP concentrations by two orders of magnitude to match the observed ICNCs. In comparison, the model that only considers the SIP through droplet shattering needs an INP increase of 50 times to match the observed ICNCs.

76 The roles of SIP have also been investigated in other geographical regions and for 77 other cloud types. Sotiropoulou et al. (2020b) simulated a summer boundary layer coastal 78 cloud in West Antarctica using the WRF model and found that the model with collisional 79 break-up between ice-phase particles can reproduce the observed ICNCs, which could not 80 be explained by the rime splintering or primary ice nucleation. Sullivan et al. (2017) used 81 a parcel model with rime splintering and graupel-graupel collisional break-up and found 82 that these two SIP processes can enhance the ICNCs by four orders of magnitude. Sullivan 83 et al. (2018a) showed that among the different SIP mechanisms, only ice-ice collisional 84 fragmentation contributes to a meaningful ice enhancement (larger than $0.002 L^{-1}$) in a 85 parcel model simulation. Other studies have shown the impact of SIP on ICNCs in a cold frontal rain band over the UK (Sullivan et al., 2018b), on surface precipitation of a tropical 86 87 thunderstorm (Connolly et al., 2006), and on the summertime cyclones (Dearden et al., 88 2016).

89	Previous modeling studies have used small-scale (e.g., parcel models and LES
90	models) and regional-scale models, to investigate the impacts of SIP on cloud properties.
91	There is still a lack of large-scale perspective based on global climate models (GCMs).
92	Moreover, the mechanisms contributing to ice production in the Arctic mixed-phase clouds
93	at moderately cold temperatures are still unknown. In this study, for the first time, we
94	implemented the representation of two new SIP mechanisms (i.e., raindrop shattering, ice-
95	ice collisional break-up) in a GCM. We tested the model performance by running the model
96	in the single column mode (SCM) and compared the SCM simulations of Arctic clouds
97	with observations. The objectives of this study are to examine the impact of SIP on different
98	types of the Arctic clouds and, ultimately to improve the model capability of representing
99	ice processes.
100	This paper is organized as follows. In section 2, we describe the GCM, associated
101	parameterizations, and the three SIP mechanisms represented in the model. In section 3,
102	we present the model experiments and observation data used for model evaluation. The
103	model results are presented in section 4. The main conclusions of this study and future
104	work are summarized in section 5.

105 2 Model and Parameterizations

106 **2.1 Model description**

107 The Community Atmosphere Model version 6 (CAM6) used in this study is the 108 atmosphere component of the Community Earth System Model version 2 (CESM2). It 109 includes multiple physical parameterizations that are related to ice formation and evolution. 110 Cloud microphysics is described by a double-moment scheme (Gettelman and Morrison, 111 2015, hereafter as MG). The scheme considers homogeneous freezing of cloud droplets 112 (with temperatures below -40 °C), heterogeneous freezing of cloud droplets, the WBF 113 process, accretion of cloud droplets by snow, and the rime splintering. SIP from rime 114 splintering is parameterized based on Cotton et al. (1986). The condensation process is also 115 known as cloud macrophysics, which is governed by the Cloud Layers Unified by Binormals 116 (CLUBB) scheme, assuming that all the condensate is in the liquid phase (Golaz et al., 2002; 117 Larson et al., 2002). Furthermore, CLUBB also treats boundary layer turbulence and shallow 118 convection. In the mixed-phase clouds, heterogeneous ice nucleation is represented by the 119 classical nucleation theory (CNT), which relates ice nucleation rate to mineral dust and black 120 carbon aerosols (Wang et al., 2014). In cirrus clouds, where temperatures are below -37 °C, 121 heterogeneous immersion freezing on dust can compete with homogeneous freezing of 122 sulfate (Liu and Penner, 2005). The aerosol species involved in ice nucleation processes are 123 represented by the four-mode version of the Modal Aerosol Module (MAM4) (Liu et al., 124 2012; Liu et al., 2016).

125	In this study, we conducted the simulations using the SCM version of CAM6 (i.e.,
126	SCAM). SCAM is a one-column, time-dependent model configuration of CAM6 that
127	provides an efficient way to understand the behavior of model physical parameterizations
128	without the influence of nonlinear feedbacks from the large-scale circulation. In this way, the
129	biases of the modeled clouds can be exclusively identified from model evaluation against
130	observations.

2.2 Implementation of secondary ice production in CESM2 131

132 In addition to the existing SIP mechanism (i.e., rime splintering) in CAM6, we implemented two new mechanisms of SIP, including ice-ice fragmentation and droplet 133 134 shattering (Phillips et al., 2017a, 2018) that are parameterized based on theoretical and 135 measurement research.

136

a. An emulated bin framework

137 Ideally, a bin microphysics scheme is the most suitable model setup for the representation of SIP mechanisms in a model. However, running a GCM model with a bin 138 139 microphysics scheme is computationally too expensive under current computational 140 resources. To solve this problem, we developed an emulated bin framework for the existing bulk MG microphysics scheme to facilitate the collisions of ice hydrometeors and
raindrops. First, we selected the bin bounds for each hydrometeor, including cloud ice,
snow, and rain. A logarithmically equidistant size grid is adopted, that is,

 $D_{k+1} = CD_k, \tag{1}$

where $C = \sqrt[4]{2}$. The bin diameter ranges from 0.1 to 6 mm for raindrops and 0.1 to 50 mm 145 146 for snow and cloud ice particles. Based on the assumption of the particle size distribution, 147 the number concentration and mass mixing ratio of all hydrometeor types were calculated 148 in each temporary bin at each time step and grid point. The estimated particle size 149 distribution from the emulated bin framework served as inputs for the SIP schemes. The 150 SIP schemes were applied to each permutation of the bin during collisions of ice, snow, 151 and rain to calculate the secondary ice fragments. Finally, we summed up the fragment 152 from SIP over all pairs of bins.

153 The bin approach is only adopted in the SIP processes, while other processes, 154 including the existing collisions in the standard MG scheme, still use the bulk 155 microphysical approach. Thus, the modified MG scheme becomes a hybrid scheme that 156 combines the bulk and bin parameterizations. The advantage of this hybrid scheme is that the scheme can provide an accurate representation of the SIP processes while still maintains 157 158 a relatively high computational efficiency, which is very important for GCMs. The hybrid 159 schemes have been widely used. For example, previous studies used the bin approach for 160 the warm rain processes, while adopted the bulk approach for the ice-related processes

(Onishi and Takahashi, 2012; Grabowski et al., 2010; Kuba and Murakami, 2010). Other
previous studies used the bin approach for the sedimentation (Morrison, 2012) or look-up
tables for the collision processes in the bulk schemes (Feingold et al., 1998).

164

165 **b. Ice-ice fragmentation**

Phillips et al. (2017a, b) developed a scheme for SIP during an ice-ice collision based on the principle of energy conservation. This scheme relates the fragment numbers to particle initial kinetic energy and ice particle habits (i.e., ice morphology), which can be explained in terms of environmental temperature, particle size, and riming intensity of ice particles (Fig. 1). The production of new ice particles per collision is calculated as:

171

172
$$\mathcal{N} = \alpha A \left[1 - e^{-\left(\frac{Ck_0}{\alpha A}\right)^{\gamma}} \right]$$
(2)

173

in which α is the surface area of ice particle, i.e., the equivalent spherical area in a unit of m², $\alpha = \pi D^2$; *A* is the number density of breakable asperities of ice particles, which is related to riming intensity and ice particle size; *C* is the asperity-fragility coefficient, prescribed to be 10815 for dendrites and 24780 for spatial planar; γ is a parameter related to riming intensity (*rim*), $\gamma = 0.5 - (0.25 \times rim)$, and *rim* is assumed to be 0.1; k_0 is the initial kinetic energy, which is given as:

180
$$k_0 = \frac{1}{2} \frac{m_1 m_2}{m_1 + m_2} (v_1 - v_2)^2$$
(3)

181 in which m_1 and m_2 are the particle masses of two colliding particles, and v_1 and v_2 182 are the terminal velocities of the two colliding particles.

In this method, three types of collision are identified based on the type of collision particles: (1) cloud ice/snow collide with hail/graupel; (2) cloud ice/snow collide with cloud ice/snow; (3) hail/graupel collide with hail/graupel (not included currently, since CESM2-CAM6 does not treat graupel currently); For each collision type, different values of parameters α , A, C, and γ in Eq. (2) are yielded based on the measured relationship between fragment number and collisional kinetic energy (Phillips et al., 2017a).

189 Under the emulated bin framework, the new fragment production rate for each190 permutation of a bin is written as:

191
$$N_{iic} = \mathcal{N} E_c \delta N_1 \delta N_2 \pi (r_1 + r_2)^2 |v_1 - v_2|$$
(4)

in which E_c is the accretion efficiency, assumed to be 0.5 to be consistent with the MG microphysics scheme, and δN_1 and δN_2 are the particle number concentrations in the two bins with particle sizes of r_1 and r_2 , respectively.

- 195 The ice production rate for cloud ice mixing ratio is:
- $P_{iic} = N_{iic} \delta m_{ice} \tag{5}$

197 in which δm_{ice} is mass for single ice particle, prescribed as 2.09×10⁻¹⁵ kg.

198 c. Droplet shattering during rain freezing

199	Phillips et al. (2018) proposed numerical formulations for ice multiplication during
200	the raindrop freezing. They suggested two modes of droplet break-up during the rain
201	freezing based on the relative weight of raindrop and ice particle (Fig. 2).

202 In mode 1, the freezing of rain is triggered by a collision with less massive ice 203 crystals or with INPs. By fitting to the laboratory data, Phillips et al. (2018) derived an 204 empirical formulation for the number of ice fragments per frozen raindrop as a function of 205 drop diameter and temperature. A Lorentzian distribution as a function of temperature was 206 adopted to represent the number of ice fragments per frozen raindrop. There are two types 207 of raindrop fragmentation: shattering to form "big" fragments and "tiny" splinters. The 208 total (big plus tiny) and big ice fragments per frozen raindrop emitted in the mode 1 of 209 droplet shattering are given in Eqs. (6) and (7), respectively:

210
$$\mathcal{N}_T = F(D)\Omega(T) \left[\frac{\zeta \eta^2}{(T - T_0)^2 + \eta^2} + \beta T \right]$$
(6)

211
$$\mathcal{N}_B = \min\left\{F(D)\Omega(T)\left[\frac{\zeta_B \eta_B^2}{(T - T_{B0})^2 + \eta_B^2}\right], \mathcal{N}_T\right\}$$
(7)

where the parameters ζ , η , β , ζ_B , η_B , T_0 , T_{B0} are derived by fitting the formulations to a collection of laboratory data. Further details about empirical formulae can be found in Phillips et al. (2018). F(D) and $\Omega(T)$ are the interpolating functions for the onset of fragmentation and *T* is the temperature in K. The mass of a big fragment is $m_B = \chi_B m_{rain}$, 216 in which $\chi_B = 0.4$, and the mass of a small fragment is $m_S = \frac{\pi \rho_i}{6} D^3$, in which $\rho_i = 217 \quad 500 \ kg \ m^{-3}$.

The observational data used for the formulations of raindrop freezing by mode 1 was limited to drop diameter of 1.6 mm and a temperature range between -4 °C to -25 °C. Phillips et al. (2018) linearly extrapolated their algorithm for larger particles and other temperatures in the mixed-phase cloud regime. As shown in Fig. 2a, b, mode 1 of the droplet shattering is most effective near -15°C.

In mode 2, a theoretical approach is adopted which is based on the assumption that the number of fragments generated when a drop collides with a more massive ice particle is controlled by the initial kinetic energy and surface energy (Fig. 2c). The number of fragments generated per frozen drop in mode 2 is given as:

227
$$\mathcal{N}_{fr2} = 3\Phi(T) \times [1 - f(T)] \times \max(DE - DE_c), \tag{8}$$

228 where *DE* is the dimensionless energy and is expresses as:

$$DE = \frac{k_0}{s_e},\tag{9}$$

where k_0 is the initial kinetic energy which is given in Eq. (3), S_e is the surface energy, expressed as $S_e = \gamma_{liq} \pi D^2$ (for $D > 150 \ \mu m$), γ_{liq} is the surface tension of liquid water which is 0.073 J m⁻². DE_c in Eq. (8) is set to be 0.2. f(T) is the frozen fraction (Phillips et al., 2018), and is given as:

234
$$f(T) = \frac{-C_w T}{L_f}$$
 (10)

where C_w is the specific heat capacity of liquid water (4200 J kg⁻¹ K⁻¹) and L_f is the specific latent heat of freezing (3.3×10⁵ J kg⁻¹), $\Phi(T) = 0.5$ at -1°C and $\Phi(T) = min [4f(T), 1]$.

237 d. Rime splintering

The MG microphysics already includes the SIP associated with rime splintering, which is also known as Hallet-Mossop (HM) process. In this process, secondary ice particles are generated during the accretion of cloud droplets by snow, and a part of rimed mass is converted to cloud ice. The ice number production rate is based on the parameterization of Cotton et al. (1986), which is given as:

243
$$N_{HM} = C_{sip_HM} \times p_{sacws}$$
(11)

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

245
$$p_{sacws} = \frac{\pi \times a_{vs} \times \rho \times N_{0s} \times E_{ci} \times \Gamma(b_{vs}+3)}{4 \times \lambda^{b_{vs}+3}}$$
(12)

in which E_{ci} is the collection efficiency for the riming of cloud droplets by snow, 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 air density and typical air density at 850 hPa, respectively, and N_{0s} and λ are the parameters for the snow particle size distribution.

250 The conversion coefficient $C_{sip_{HM}}$ in Equation (11) depends on temperature T_c in

251 °C:

252
$$C_{sip_HM} = \frac{3.5 \times 10^8 \times (-3-T)}{2}$$
, when $-5 < T_c < -3$, and (13)

253
$$C_{sip_{HM}} = \frac{3.5 \times 10^8 \times (T - (-8))}{3}$$
, when $-8 < T_c < -5$ (14)

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

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

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

kg. The rime splintering rate as a function of p_{sacws} and temperature is shown in Fig. 3.

3 Case Description, Observations, and Model Experiments

259 **3.1 M-PACE case**

260 In this study, we focus on the Arctic mixed-phase clouds observed during the 261 Department of Energy (DOE)'s Atmospheric Radiation Measurement (ARM) Mixed-262 Phase Arctic Cloud Experiment (M-PACE). The M-PACE campaign was conducted over 263 the North Slope of Alaska (NSA) during the autumn from 27 September to 22 October 2004. 264 Various types of clouds were observed during M-PACE, including multilayer clouds, 265 boundary layer mixed-phase stratus, cirrus, and altostratus clouds associated with the frontal 266 system (Verlinde et al., 2007; Liu et al., 2007; Xie et al., 2008; Liu et al., 2011). Single-267 layer mixed-phase clouds were formed under moderately supercooled conditions with the cloud temperature at around -10 °C (Verlinde et al., 2007; McFarquhar et al., 2007), 268 269 providing a favorable condition for studying the influence of SIP on cloud evolution (Field 270 et al., 2016).

271	The synoptic-scale systems regulated the properties of clouds observed during the M-
272	PACE campaign. Hence, Verlinde et al. (2007) divided the M-PACE period into three
273	synoptic regimes and two transition periods based on the synoptic weather conditions. The
274	first synoptic regime began on 24 September and lasted until 1 October, 2004, when a well-
275	developed trough dominated aloft with several low-pressure systems that influenced the
276	surface. Followed by the first transition period between 2 and 3 October, the second synoptic
277	regime occurred between 4 and 14 October (Fig. 4), which was controlled by a pronounced
278	high-pressure system. The second transition period was from 15-17 October. By 18 October,
279	a fast-developing strong frontal system controlled the cloud formation over the NSA in the
280	third synoptic regime (Fig. 4). During M-PACE, the surface flux of water vapor, sensible
281	heat, and latent heat played different roles in the cloud formation. For example, clouds
282	formed in response to a strong surface forcing during the second regime, while clouds formed
283	under a relatively weak surface forcing during the third regime. In this study, we evaluate the
284	modeled cloud properties with M-PACE observations in the second and third synoptic
285	regimes focusing on the boundary layer mixed-phase stratus during 9-12 October in the
286	second regime.

3.2 Observation data

The observed cloud occurrence data at Barrow (located at 71.3° N, 156.6° W) are
from the ARM Climate Modeling Best Estimate product (Xie et al., 2010). The liquid water

290	path (LWP) and ice water path (IWP) data are obtained from Zhao et al. (2012).
291	Specifically, the Shupe and Turner's data are based on the retrievals of cloud properties
292	measured by the ARM Millimeter-Wavelength Cloud Radar (Shupe et al., 2005) and the
293	Microwave Radiometer (MWR) (Turner et al., 2007), with the uncertainties for liquid water
294	content (LWC) within 50% and for ice water content (IWC) within a factor of 2. For
295	Wang's data, IWP is retrieved from the combined ARM Millimeter-Wavelength Cloud
296	Radar and Micropulse lidar measurements (Wang and Sassen, 2002) with an uncertainty
297	of 35% (Khanal and Wang, 2015). LWP is retrieved from the ARM MWR measurements
298	with an uncertainty of 50% (Wang, 2007). For Deng's data, IWC is retrieved based on the
299	Millimeter-Wavelength Cloud Radar measurements with a retrieval error within 85%
300	(Deng and Mace, 2006). For Dong's data, LWC is retrieved from the MWR measurements
301	with an uncertainty within 113% (Dong and Mace, 2003). Note that measured IWC and
302	IWP cannot distinguish cloud ice from the snow. The simulated IWP and IWC therefore
303	include the snow component which is consistent with observations used in this study.
304	The ICNC was measured during the M-PACE single-layer mixed-phase stratus
305	period. The data includes 53 profiles measured in four flights over Barrow and Oliktok Point
306	(located at 70.5° N, 149.9° W) by the University of North Dakota Citation aircraft. By
307	combining measurements from different probes, McFarquhar et al. (2007) provided cloud
308	particle size distributions over a continuous size range. The forward scattering spectrometer
309	probe (FSSP) measured particle number concentrations with particle diameters between 3 to

310	53 μ m, while the one-dimensional cloud probe (1DC) counted cloud particles ranging from
311	20 to 620 μ m. The two-dimensional cloud probe (2DC) covered particle sizes from 30 to 960
312	μ m, while the high-volume precipitation sampler (HVPS) sampled particles from 0.4 to 40
313	mm. The data were collected every 10 seconds but were averaged to 30s^{-1} to ensure adequate
314	statistical sampling. The cloud phase was identified by detecting the presence of supercooled
315	droplets by the Rosemount Icing Detector (RICE). In mixed-phase clouds, any particles
316	larger than 125 μm are identified as ice particles, and cloud particles smaller than 53 μm are
317	counted as liquid-phase particles. Particles with a diameter ranging from 53 to 125 μm are
318	counted as a liquid when there is drizzle, and as ice, if there is no drizzle. A more detailed
319	description of the particle phase identification algorithm can be found in McFarquhar et al.
320	(2007). When comparing the simulated ICNC with the observations, we only consider ice
321	particles larger than 53 μ m, as the observations were limited to ice particles larger than 53
322	μm.

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 the Instrumentation Development and Education in Airborne Science 2011 (IDEAS-2011), the Holographic Detector for Clouds (HOLODEC), and the Indirect and Semidirect Aerosol Campaign (ISDAC), which

330	also used the 2DC cloud probe, but adopted anti-shattering tips and algorithms for
331	removing the shattered particles (Jackson and McFarquhar, 2014; Jackson et al., 2014). In
332	order to account for the anti-shattering effect, observed ice number concentrations were
333	scaled by a factor of 1/4 and 1/2, respectively, to consider the possible range of the
334	shattering effect. Furthermore, to be consistent with Figure 10 in Jackson et al. (2014), only
335	ice particles with diameters larger than 100 μ m are included in our model and observation
336	intercomparisons.

337 **3.3 Model set up and description of model experiments**

338 In this study, we run SCAM with 32 vertical layers from the surface up to 3 hPa. The 339 model is initialized and driven by the large-scale forcing data at every 3 hours. The forcing 340 data developed by Xie et al. (2006) include the divergences and advections of moisture and 341 temperature as well as the surface flux. The simulation period is from 5 to 22 October 2004 342 and covers the second and third synoptic regimes and the transition period between them. 343 A detailed description of model experiments along with SIP mechanisms in these 344 experiments is provided in Table 1. The control experiment (CTL) uses the default CAM6 345 model that only includes the SIP due to the HM process. The impacts of two new SIP 346 mechanisms, including the ice-ice collision break-up and rain freezing fragmentation based on Phillips et al. (2017a, 2018) are addressed in the SIP PHIL experiment. To examine the 347

impact of rime splintering in the CTL experiment, we conducted CTL_no_HM experimentthat is similar to CTL but without the HM process.

350

351 **4 Results**

352 4.1 SIP impacts on different types of clouds during M-PACE

353 Figure 4 shows the temporal evolution of LWP, IWP, and cloud fractions from two 354 model simulations (CTL and SIP PHIL) and their comparison to observations. The model 355 simulations cover the second and third synoptic regimes as well as the transition period 356 between them. Two different types of clouds were formed in response to the strong surface 357 forcing during the second synoptic regime from 4 to 14 October. As shown in Fig. 4c, 358 multilayer stratus occurred from 5 to 8 October, and the clouds extended from 950 hPa up to 359 500 hPa. Between 9 and 14 October, single-layer boundary layer stratus occurred between 360 800 and 950 hPa. Because of the dramatic change in cloud types in the second regime, we 361 further separate the second regime into two time periods. Then, we select typical days in the 362 four time periods for our analysis in this study, as shown in Fig. 4. The period from 6 to 8 363 October is selected as the "multilayer stratus" period. The period from 9 to 12 October is 364 selected as the "single-layer stratus" period, followed by the transition period marked on 16 365 October. The period between 18 and 20 October is selected to represent the "frontal cloud"366 type during the third regime.

367	Figure 4 shows that the simulated IWP is systematically underestimated during M-
368	PACE in the CTL experiment. The maximum value of IWP in CTL is smaller than 50 g m $^-$
369	2 during M-PACE, but up to 500 g m^{-2} in the measurements. The SIP_PHIL experiment
370	shows decreased LWP and increased IWP compared with CTL, reaching a better agreement
371	with the measurements. For example, IWP increases from 50 g m ⁻² in CTL to 425 g m ⁻² in
372	SIP_PHIL on 20 October, compared with $300 \sim 475$ g m ⁻² from different measurements (Fig.
373	4). The simulated LWP is overestimated during the "multilayer stratus", the second half of
374	the "single-stratus", and the "frontal cloud" periods in CTL, particularly on 20 October.
375	The SIP_PHIL experiment decreases the LWP from 550 g m ⁻² in CTL to 300 g m ⁻² on 11
376	October and from 425 g m ⁻² in CTL to 70 g m ⁻² on 20 October (Fig. 4a). The CTL_no_HM
377	experiment has similar results as the CTL experiment.
378	During the multilayer stratus period, the CTL and SIP_PHIL experiments show that the
379	cloud top is located at about 5 km with a temperature of -20 °C (Fig. 5). These cloud
380	properties are consistent with the observations (Verlinde et al., 2007) that show a minimum
381	observed cloud temperature of -17°C (Fig. 4). However, we notice a significant

- 382 overestimation of cloud amount at 6–8 km on 7 October by the model simulations in Fig. 5,
- 383 as compared to the observation in Fig. 4c.

384	During this period, IWC is increased in the SIP_PHIL experiment compared to CTL,
385	while LWC is decreased. The mean vertical profiles of simulated IWC and LWC in this
386	period are shown in Fig. 6. The simulated values of LWC and IWC are lower than
387	observations, particularly for IWC. LWC decreases from 130 mg m $^{-3}$ in CTL to 80 mg m $^{-3}$
388	in SIP_PHIL below 1 km. IWC increases from 3 mg m ⁻³ in CTL to 5 mg m ⁻³ in SIP_PHIL.
389	The time-averaged IWP increases from 11.2 g m ⁻² in CTL to 17.1 g m ⁻² in SIP_PHIL but is
390	still lower than the observed value of 55.6 g m ^{-2} (Table 2). After considering the SIP in the
391	model, for the multilayer stratus period, ICNC is increased by 1 L^{-1} (Fig. 5) at an altitude of
392	1 to 4 km. Observations of ICNC are not available during this period.
393	Between 9 and 14 October, a persistent boundary-layer mixed-phase stratus occurred
394	between 800-950 hPa, with the cloud top temperature at around -15 °C (Verlinde et al., 2007).
395	This single-layer stratus was separated from the surface based on the measurement (Fig. 4c).
396	However, modeled clouds extend to the surface in CTL (Fig. 5). This bias is alleviated in
397	SIP_PHIL on 8 and 11 October (Fig. 5). Previous studies also found that this bias partially
398	results from the overestimation of low-level moisture in the large-scale forcing data (Zhang
399	et al., 2019, 2020).
400	Observed cloud liquid is located above the cloud ice during this period, with the LWC
401	peak \sim 0.5 km above the IWC peak. Observed vertical profile of LWC shows a maximum of
402	300 mg m ⁻³ (ranging from 210 to 500 mg m ⁻³) at ~1.25 km, while observed IWC is peaked
403	at 0.75 km (Fig. 6). This characteristic is clearly captured by the SIP_PHIL experiment, with

404	the peaks of LWC and IWC located at 0.75 and 0.5 km, respectively (Fig. 6). A better relative
405	position of cloud liquid and ice in SIP_PHIL indicates a better simulation of interactions
406	between cloud physics and dynamics. This distinct feature also contributes to the longevity
407	of mixed-phase clouds in the Arctic, as discussed in Section 1. In SIP_PHIL, the maximum
408	IWC value is four times larger than that in CTL (2 versus 0.5 mg m ⁻³); accordingly,
409	temporally-averaged IWP increases from 0.9 in CTL to 2.5 g m ^{-2} in SIP_PHIL (Table 2).
410	Meanwhile, ICNC in SIP_PHIL is higher than that in CTL, and the maximum ICNC goes
411	up by 5 L^{-1} at 0.5 km on 11 October (Fig. 5). Thus, SIP adds an extra source of ice crystals
412	to the boundary-layer mixed-phase stratus clouds.

413 During the transition period, several distinct liquid layers are interrupted by the ice 414 enriched layers in the observation. Due to the coarse vertical resolution, the model may not 415 be able to capture this vertical variation accurately. Considerable variation was noticed in the observed IWC with a maximum IWC of 0.8-1.8 mg m⁻³ (Fig. 7). The CTL experiment 416 417 substantially underestimates IWC, as it produces IWC less than 0.1 mg m⁻³. The maximum 418 IWC in SIP PHIL is 1.15 mg m^{-3} , providing a better agreement with the observation. The simulated peak LWC is decreased from 80 in CTL to 65 mg m⁻³ in SIP PHIL, which is 419 closer to the observed value of 55 mg m⁻³. The temporally-averaged IWP in SIP PHIL is 420 10⁴ times larger than that in CTL, with values of 0.0001, 3.6, and 5.6 g m⁻² in CTL, SIP PHIL, 421 and observation, respectively (Table 2). The vertically-integrated ICNC is 7.66 and 4.57×10^5 422

423	L ⁻¹ in CTL and SIP_PHIL, respectively (Table 2). Considering SIP in the model increases
424	vertically-integrated ICNC by five orders of magnitude during the transition period.
425	During the frontal cloud period, stratocumulus and altostratus clouds associated with
426	the frontal system extended from the surface up to 8 km (Fig. 4). The SIP_PHIL experiment
427	shows the largest absolute increases in IWC and ICNC compared to the other periods (Fig.

428 5). The peak of modeled IWC is located at 2.5 km, with values of 2 and 8 mg m⁻³ in CTL

429 and SIP_PHIL, respectively (Fig. 7), much lower than the observation (ranging from 8 to 40

430 mg m⁻³). IWP is 10.4, 26.1, and 96 g m⁻² in CTL, SIP_PHIL, and the observation,

431 respectively (Table 2). ICNC is increased by up to 7 L^{-1} between 2 to 4 km on 20 October

432 from CTL to SIP_PHIL (Fig. 5). The simulated LWP is decreased from 127.6 to 41.2 g m^{-2} ,

433 which is closer to the observed value of 50.2 g m⁻².

434 The relative importance of primary and secondary ice production is shown as pie 435 charts in Fig. 8, to identify the dominant ice production mechanism in different types of 436 the Arctic clouds. The primary ice production (i.e., ice nucleation) is more important in the 437 clouds with colder cloud tops, such as multilayer stratus and frontal clouds with cloud top 438 temperatures colder than -25 °C and -40 °C, respectively. The primary ice production 439 contributes 37% and 69% to the total ice production during the multilayer stratus and 440 frontal cloud periods, respectively. Primary ice production is more efficient in deep clouds 441 due to the inverse relationship between the ice nucleation rate and temperature. SIP is more 442 important than primary ice production in the boundary-layer stratus and in clouds during

443	the transition period when cloud top temperatures were at -15 °C. The fragmentation of
444	freezing raindrops contributes the most (up to 80%) to the ice production in the single-layer
445	boundary-layer stratus. The break-up from ice-ice collisions contributes 22% to the total
446	ice production in the frontal clouds, while the rime splintering contributes 22% to the
447	multilayer stratus. These two SIP mechanisms (i.e., break-up from ice-ice collisions and
448	rime splintering) account for a small fraction of the ice production in the boundary-layer
449	stratus.

450 Next, we will focus on the SIP impacts on the boundary-layer stratus related to the451 phase partitioning (section 4.2) and ICNC (section 4.3).

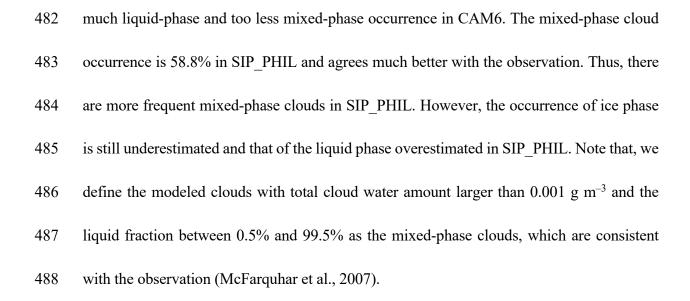
452 4.2 SIP impact on occurrence and phase partitioning of the mixed-phase 453 clouds

454 Figure 9 shows the liquid fraction (defined as LWC/(LWC+IWC)) as a function of normalized height in the single-layer boundary-layer stratus. The normalized height Z_n is 0 455 456 at cloud base and 1 at cloud top. IWC from the model includes all the ice hydrometeors to 457 compare it with observations. Fig. 9a reveals two features of the observed single-layer 458 boundary-layer clouds: (1) mixed-phase is dominant in the clouds, and (2) the liquid fraction 459 increases with cloud altitude. The liquid fraction is between 0.05 and 0.95 in most portions 460 of the clouds, indicating a mixed-phase feature in the observation. In the upper portion of the 461 clouds, observed liquid fraction is larger than 0.6 with the mean value increasing with height.

462	In the lower portion of the clouds ice mass fraction increases as a result of ice growth by
463	riming of cloud liquid and ice sedimentation from the upper levels. The CTL experiment
464	cannot reproduce the observed mixed-phase feature. A large portion of the clouds is in liquid
465	phase with the liquid fraction close to 1 in CTL, which significantly overestimates the liquid
466	fraction in the clouds. This is vastly different from previous versions of CAM. CAM5
467	showed an underestimation of the liquid fraction (Liu et al., 2011; Cesana et al., 2015; Tan
468	and Storelvmo, 2016; Zhang et al., 2019; Tan and Storelvmo, 2019), while CAM3 showed
469	a decrease of the liquid fraction with height due to its use of a temperature-dependent phase
470	partitioning (Liu et al., 2007).
4771	

471 The SIP PHIL experiment improves the model simulation of cloud phase with 472 increased ice fraction in the bottom half of the clouds by adding an extra source of ice crystals 473 from SIP (Fig. 9c). The CTL no HM experiment gives very similar results as the CTL 474 experiment (Fig. 9d). Note that the modeled liquid fraction distributes on discrete vertical 475 levels (Fig. 9b, c, d) due to the coarse model vertical resolution (with only 10 vertical levels 476 below 2 km). In contrast, observed data were detected at 10 s⁻¹ resolution during spiral 477 ascents and descents in the clouds so that the observed liquid fraction is distributed 478 continuously with height.

For the cloud occurrence, 62.7% of observed clouds are mixed-phase, and only 16%
are liquid-phase during the single-layer stratus period, as shown in Table 3. The liquid phase
cloud occurrence is 73% in CTL and only 26.9% for mixed-phase clouds, indicating too



4.3 SIP impact on ice crystal number concentration

4.3.1 Vertical distribution of ice crystal number concentration

491	The vertical distribution of ICNCs in the single-layer boundary-layer stratus clouds
492	on October 9, 10, and 11 from model simulations and observations is shown in Figure 10.
493	The measured ICNCs when applied with a correction factor of $^{1\!/_{\!\!4}}$ range from 0.02 to 20 L^-
494	¹ , with an average value of 1 L^{-1} . The CTL and CTL_no_HM experiments have similar
495	results, and both underestimate the ICNCs in all the cloud layers, with a mean ICNC of
496	~0.1 L^{-1} and the maximum concentration of 1 L^{-1} . The mean ICNC is increased to ~1 L^{-1}
497	in the SIP_PHIL experiment with the maximum concentration of $30 L^{-1}$, which are in better
498	agreement with the observations compared to CTL. ICNCs are increased by more than one

order of magnitude in the lower portion of the clouds, although they are still lower thanthose observed in the upper portion of the clouds.

501 Figure 10 also shows the linear regressions of ICNCs as a function of cloud altitude 502 (black lines). ICNCs increase towards the cloud base in the observation, revealing ice 503 multiplication during the ice growth and sedimentation. The CTL experiment shows that 504 the ICNCs decrease towards the cloud base, an opposite pattern compared to the 505 observation. SIP PHIL captures the observed pattern in the vertical profile of ICNCs (Fig. 506 10c), suggesting that SIP is an important source of ice crystals near the cloud base in the 507 Arctic boundary-layer mixed-phase stratus. Furthermore, the vertical distribution of ice 508 particles is important for the longevity of the Arctic mixed-phase clouds, which features 509 lower ICNCs in the upper portion of clouds and higher ICNCs towards the cloud base.

510

4.3.2 PDF of ice crystal number concentration

Figure 11 shows the probability density function (PDF) (i.e., the frequency of occurrence) of ICNCs from model simulations and observations for the boundary-layer mixed-phase stratus period (October 9-12, 2004). Note that only particles with a diameter greater than 100 μ m are included in the observed and modeled ICNCs. The PDF distribution in SIP_PHIL shows a shift to the right, with the ICNC peak much closer to the observations than CTL. The median ICNC is 0.13 L⁻¹ in CTL, shifting to 0.27 L⁻¹ in SIP_PHIL, which is closer to the observed median value of 0.32 L⁻¹.

518	The PDF distribution in SIP_PHIL also has a broader distribution than CTL. A
519	broader distribution indicates that the maximum concentrations are higher in the
520	observation and SIP_PHIL compared to CTL. In the CTL experiment, the frequency of
521	occurrence of ICNCs is much lower (higher) than observations when their values are higher
522	(lower) than 0.1 L^{-1} . These biases in ICNCs PDF are much improved in SIP_PHIL, leading
523	to a better agreement with the observation. The frequency occurrence of ICNC at 1 L^{-1} is
524	2.12%, 10.37%, 13.77% in CTL, SIP_PHIL, and observation, respectively. Thus,
525	SIP_PHIL has an occurrence frequency of ICNC larger than 1 L ⁻¹ , which is 5 times of that
526	in CTL. We note that the agreement between modeled and observed ICNCs is improved
527	with a correction factor of $^{1}\!\!\!/_{4}$ (Figures 10 and 11) and a correction factor of $^{1}\!\!/_{2}$
528	(supplementary Figures S2 and S4) to the observed ICNCs, compared to that without a
529	correction factor (Supplementary Figures S1 and S3). This is because model simulations
530	including SIP_PHIL underestimate the observed ICNCs without the correction of the
531	shattering effect.

532 **4.3.3** Dependence of ice enhancement on cloud temperature

533 The bivariate joint PDF defined in terms of temperature and ice enhancement 534 (N_{SIP_PHIL}/N_{CTL}) during the M-PACE is shown in Fig. S5. Strong ice enhancements are 535 noticed at temperatures from -3 to -16°C, and ICNCs are increased by nearly 4 orders of 536 magnitude in SIP PHIL compared with CTL. As temperature decreases below -35°C, ice enhancement happens again, but with a reduced magnitude. For example, the largest
enhancement at -44°C is around 3.2, with a frequency of 1% to 7%.

539	To investigate the dominant processes that contribute to the strong enhancement
540	near -10 °C we plotted the bivariate joint PDF defined in terms of temperature and ice
541	production rate (Fig. 12). A clear relationship between ice enhancement and fragmentation
542	of freezing raindrops can be seen at temperatures from -20 to -4 °C in Fig. 12 and Fig. S5.
543	The maximum ice production from the fragmentation of freezing raindrops is 160 L^{-1} (i.e.,
544	10 ^{2.2}) at temperatures ranging from -8 to -14 °C. Even though rime splintering also
545	happens at temperatures between -8 to -3 °C with a maximum value of 20 L ⁻¹ , its ice
546	production is almost one order of magnitude lower than that from the fragmentation of
547	freezing raindrops. Between -20 to -16 °C, primary ice nucleation and fragmentation of
548	freezing raindrops coexist, with the fragmentation of freezing raindrops more efficient
549	(with a magnitude of $10 L^{-1}$) comparing to the primary ice nucleation (about $1 L^{-1}$). Primary
550	ice nucleation has the largest production of up to 250 L^{-1} at temperatures ranging from –
551	32 to -25 °C. Below -35 °C, ice-ice collision break-up frequently happens, but with a lower
552	process rate.

In summary, the strongest ice enhancement occurs in the moderately supercooled clouds with temperatures around -10° C. ICNCs are increased by up to 4 orders of magnitude mainly from the fragmentation of freezing raindrops. A weaker ice enhancement is noticed frequently in ice clouds with temperatures below -35° C, which is attributed to the ice-ice collision break-up.

558 5 Summary, conclusions and outlook

In this study, two new SIP mechanisms are implemented in a GCM model (CAM6) to investigate their impacts on the Arctic mixed-phase clouds, which were observed during the DOE ARM M-PACE field campaign. The CAM6 model with the new SIP provides a better simulation of the distinct "liquid cloud top, ice cloud base" feature of long-lived Arctic boundary-layer mixed-phase clouds.

We find that model biases of underestimation of mixed-phase cloud occurrence and overestimation of pure liquid cloud occurrence are reduced for the single-layer stratus after considering the new SIP processes. The mixed-phase cloud occurrence is 26.9%, 58.8%, and 62.7% in CTL, SIP_PHIL and the observation, respectively, while the pure liquid cloud occurrence is reduced from 73% in CTL to 40% in SIP_PHIL, in a better agreement with observed 16%.

We find that the pattern of the vertical distribution of ICNCs in the single-layer stratus is reversed after considering the new SIP processes in the model. The measured decrease of ICNCs with cloud height is captured by SIP_PHIL but not by CTL. SIP also leads to a shift of PDF of ICNCs towards a more frequent occurrence of high ICNCs and

574	a less frequent occurrence of low ICNCs. We notice a taller PDF with higher peak and a
575	broader tail in SIP_PHIL, indicating that high ICNCs occur more frequently with the
576	occurrence of extreme high ICNCs (>10 ² L ^{-1}) in SIP_PHIL, which is absent in CTL.
577	The maximum ICNC is around 1, 30, and 20 L ⁻¹ in CTL, SIP_PHIL, and
578	observation, respectively, in the single-layer stratus. During the frontal cloud period, the
579	SIP_PHIL experiment shows the largest absolute increases in IWC and ICNC by 6 mg m ^{-3}
580	and 7 $L^{-1},$ respectively. The largest ice enhancement (N_{SIP_PHIL}/N_{CTL}) is noticed during the
581	transition period with a moderately cold cloud top temperature. The column integrated ICNC
582	increases by five orders of magnitude and IWP increases by four orders of magnitude in
583	SIP_PHIL compared to CTL. When comparing the relative importance between primary
584	and secondary ice production, we notice that primary ice nucleation is more dominant in
585	the deep clouds with cloud tops reaching up to 10 km. At the same time, the fragmentation
586	of freezing raindrops contributes more to ICNCs in the boundary-layer clouds.
587	At temperatures from -4 to -20 °C, significant ice enhancement is attributed to the
588	fragmentation of freezing raindrops, with the maximum ice production of 160 L ⁻¹ at -10 °C.
589	A weaker ice enhancement due to ice-ice collision break-up is noticed in ice clouds with
590	temperatures below -35 °C but with unneglectable occurrence frequencies. Primary ice
591	nucleation has the largest production by up to 251 L^{-1} in the relatively cold-mixed phase
592	clouds with temperatures between -32 to -25 °C.

In summary, the consideration of the new SIP processes in CAM6 results in a significant improvement in the model simulated clouds during M-PACE. It underscores the critical role of SIP in cloud microphysics, which should be considered in the parameterizations of GCMs.

In this study, the parameterization of the HM process rate is based on Cotton et al. 597 598 (1986). In this parameterization the ice production rate does not have a dependence on 599 droplet size. Lacking the effect of cloud droplet spectrum in the HM process is supposed to 600 result in an overestimated splintering rate in the Arctic clouds, especially for the clouds with 601 cloud-bases close to the freezing level and with small droplets in the clouds. However, the 602 overestimation in the HM splintering rate due to lack of the cloud droplet spectrum might be 603 balanced by neglecting the raindrop splintering in the HM process in the MG microphysics. 604 In this study, we keep using the bulk approach to represent the HM process, to be the same 605 as that in the standard MG microphysics scheme. It would be interesting to examine the 606 impact of a bin approach to represent the HM process on modeled clouds, which will be a 607 topic of our future studies.

For the ice fragmentation from ice-ice collisions, the graupel related collisions are not included in this study, because the current MG microphysical scheme does not treat graupel. To quantify the impacts of graupel on SIP, the cloud microphysical scheme with prognostic graupel (Gettelman et al., 2019) or a "Single-Ice" microphysical scheme (Morrison and Milbrandt, 2015; Zhao et al., 2017) will be needed.

613	We note that the representation of ice properties is highly simplified in the current
614	model. Firstly, ice particles in nature are featured with continuous size distributions with
615	complex shapes and a wide range of densities. In contrast, the current model artificially
616	classifies them into two categories (i.e., cloud ice and snow) with fixed densities, e.g.,
617	densities of 500 kg m ⁻³ for cloud ice and of 250 kg m ⁻³ for snow. Moreover, the shape of
618	all ice particles is assumed to be spherical. The parameters, a and b in the relationship of
619	terminal velocity and diameter (V-D, $V=aD^b$) are fixed values for cloud ice and snow.
620	These assumptions cannot represent the complexities of ice properties (e.g., size
621	distribution, density, shape, and fall speed) in the measurement. Lastly, the riming intensity
622	of ice particles changes as ice collides with supercooled liquid, leading to significant
623	changes in density and fall speed of ice. This evolution of ice properties is currently not
624	represented in the model. A promising method is to represent the ice-phase microphysics
625	with varying ice properties (Morrison and Milbrandt, 2015; Zhao et al., 2017).
626	
627	Competing interests: The authors declare that they have no conflict of interest.
628	
629	Data availability: The code for the Community Earth System Model version 2 (CESM) is
630	freely available at http://www.cesm.ucar.edu/models/cesm2 (last access: 3 March 2021,
631	Danabasoglu et al., 2020). The model datasets and secondary ice production code are
632	archived at the NCAR Cheyenne supercomputer and are available upon request. The

633	observation data of M-PACE campaign is obtained from the Atmospheric Radiation
634	Measurement (ARM) user facility, a U.S. Department of Energy Office of Science,
635	available at https://www.arm.gov/research/campaigns/nsa2004arcticcld (last access: 3
636	March 2021; McFarquhar et al., 2007; Verlinde et al., 2007).
637	
638	Author contributions: XZ and XL conceptualized the analysis and wrote the manuscript
639	with input from the co-authors. XZ modified the code, carried out the simulations, and
640	performed the analysis. VP and SP provided the model code for the secondary ice
641	production. VP and SP also provided scientific suggestions to the manuscript. XL was
642	involved with obtaining the project grant and supervised the study. All authors were
643	involved in helpful discussions and contributed to the manuscript.
644	
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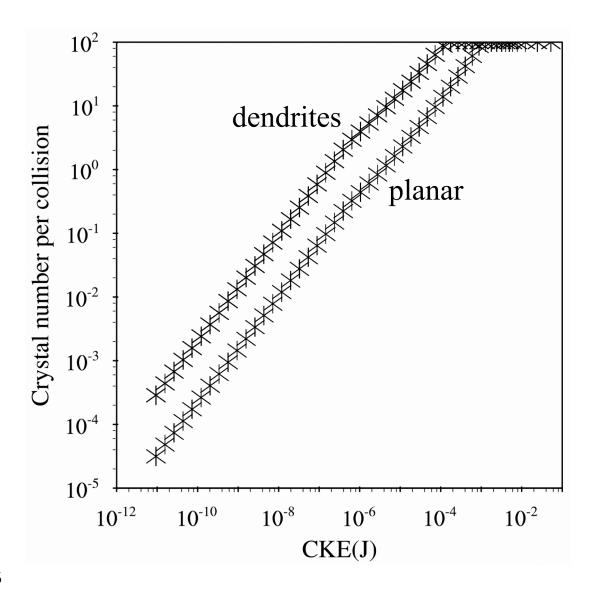
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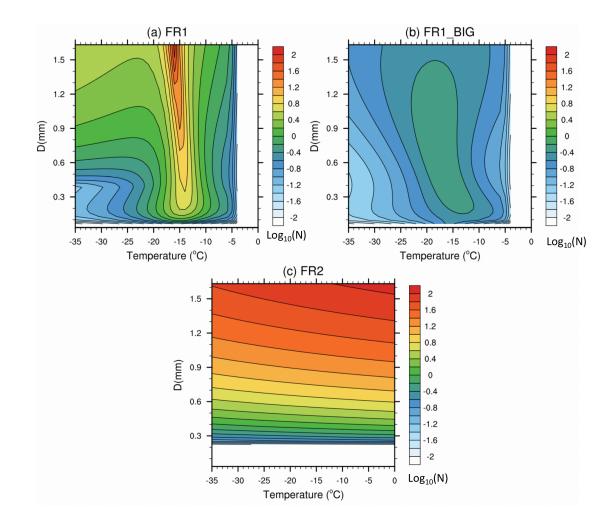
897 Figure 1. The number of fragments per collision as a function of initial collisional kinetic

898 energy (CKE). The ice habit is assumed to be dendrites when the temperature (T) is

between -12° C and -17° C and is assumed to be spatial planar when -40° C <T $<-17^{\circ}$ C

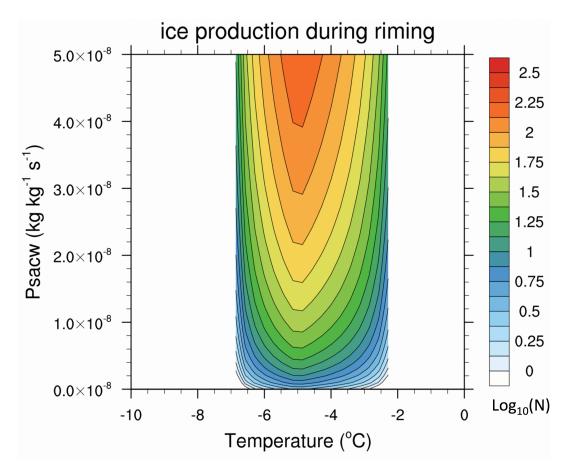
900 and $-12^{\circ}C < T < -9^{\circ}C$, following Phillips et al. (2017).

- 901
- 902



904 Figure 2. The number of fragments per frozen drop (shown as $log_{10}N$) as a function of

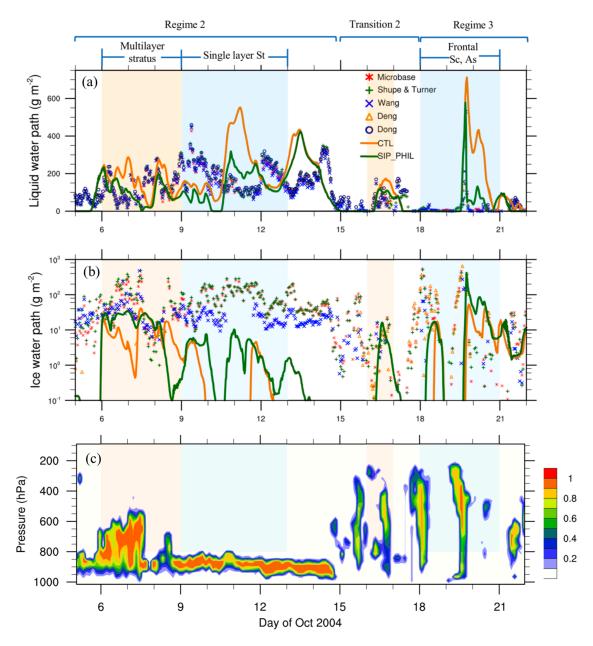
- 905 temperature and particle diameter, from (a) mode 1 of the rain freezing fragmentation
- 906 (FR1), (b) mode 1 of the rain freezing fragmentation but for the big fragments
- 907 (FR1_BIG), and (c) mode 2 of the rain freezing fragmentation (FR2).



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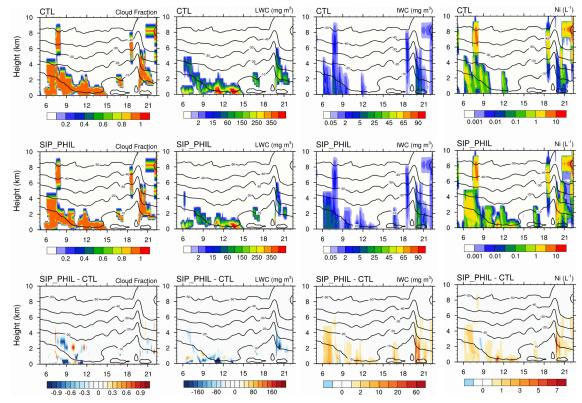
909 Figure 3. The rime splintering rate (shown as $log_{10}N$) as a function of temperature and

910 riming rate.





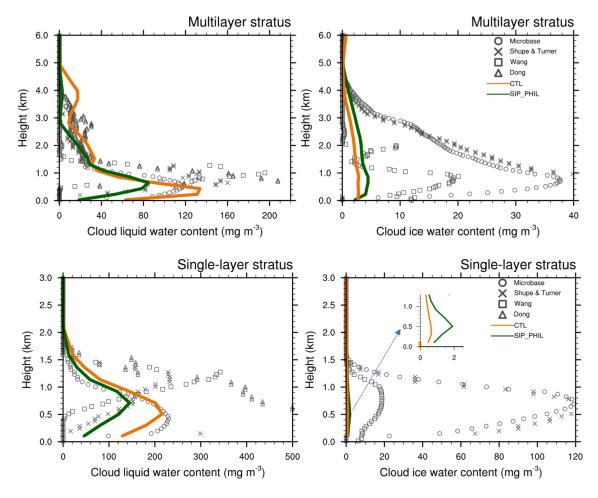
912 Figure 4. Temporal evolution of (a) LWP, (b) IWP from remote sensing retrievals shown 913 as different markers, CTL experiment (orange solid line) and SIP_PHIL experiment (dark 914 green solid line), and (c) observed time-pressure cross section of the cloud fraction. The 915 shadings show the multilayer stratus, single-layer stratus, transition, and frontal periods.



917

918 Figure 5. Time-height cross section of cloud fraction (first column), LWC (second

- 919 column), IWC (third column) and ice crystal number concentration (fourth column) from
- 920 CTL (first row), SIP_PHIL (second row) and the differences between SIP_PHIL and
- 921 CTL (SIP_PHIL minus CTL, third row).

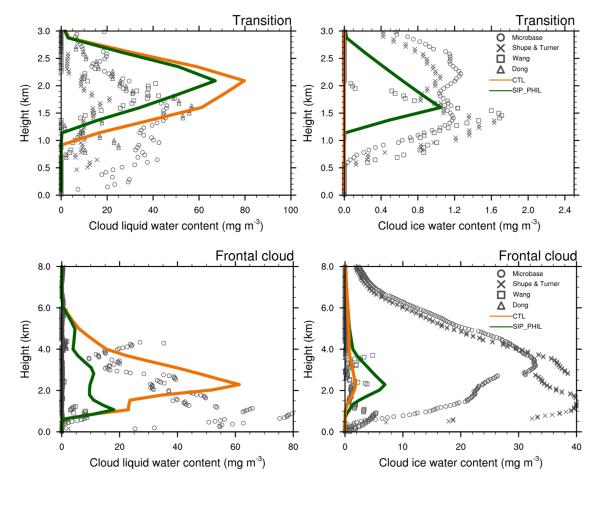


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924 Figure 6. Vertical profiles of IWC and LWC during multilayer stratus and single-layer

925 stratus periods from remote sensing retrievals shown as different markers, CTL

926 experiment (orange solid line) and SIP_PHIL experiment (dark green solid line).

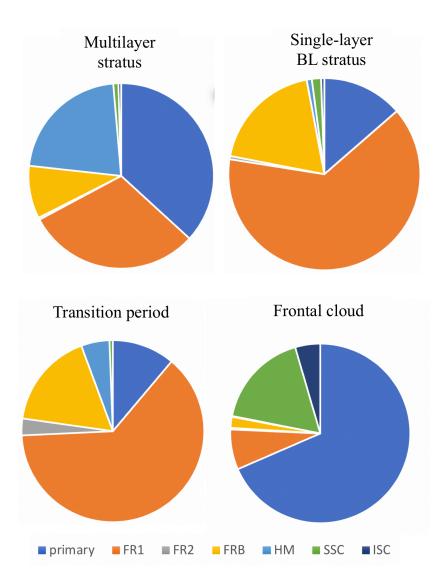


929 Figure 7. Vertical profiles of IWC and LWC during the transition and frontal cloud

930 periods, from remote sensing retrievals shown as different markers, CTL experiment

931 (orange solid line) and SIP_PHIL experiment (dark green solid line).

927





933 Figure 8. Pie charts showing the relative contributions to total ice production from

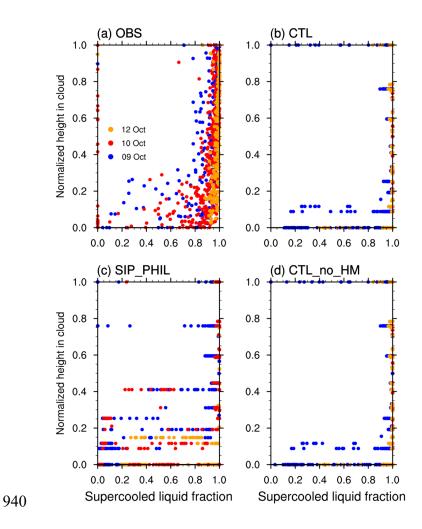
934 primary production (i.e., ice nucleation), rime splintering (HM), fragmentation of frozen

rain (including the small fragments in the first mode (FR1), big fragments in the first

936 mode (FRB), and the second mode (FR2)), breakup from ice-ice collisions (including

937 snow and cloud ice collision (ISC) and snow and snow collision (SSC)) during the four

938 M-PACE periods, the vertically integrated process rates are used in the plot.

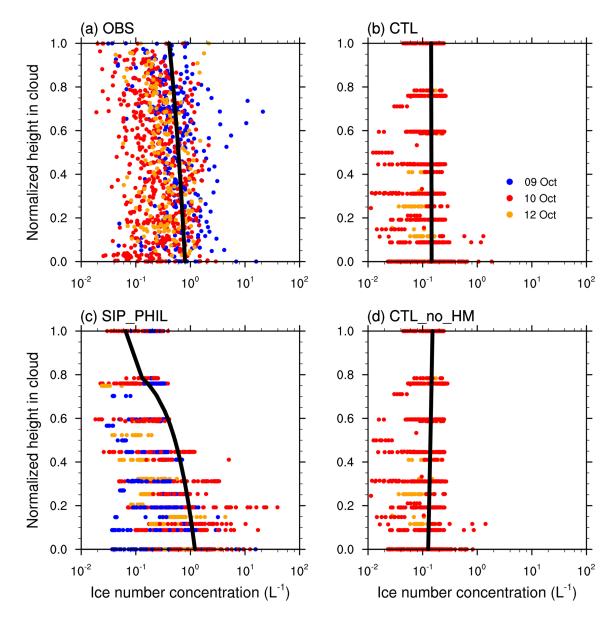


941 Figure 9. Liquid fraction as a function of normalized cloud height from cloud base. The

942 normalized cloud altitude Z_n is defined as: $Z_n = \frac{z-Z_b}{Z_t-Z_b}$, in which z is the altitude, Z_b is

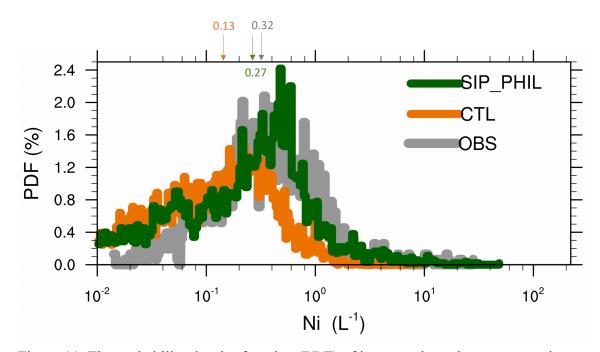
943 the altitude of cloud base, and Z_t is the altitude of cloud top, from (a) observation, (b)

944 CTL, (c) SIP PHIL, and (d) CTL no HM.



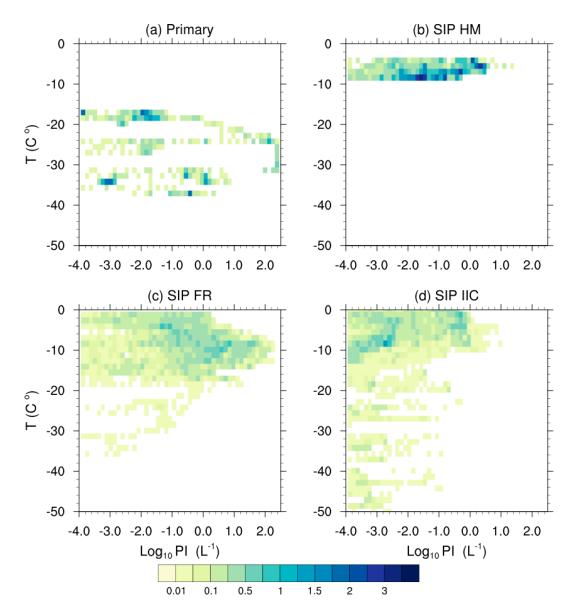
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Figure 10. Ice number concentrations as a function of normalized cloud height from cloud base from (a) observation, (b) CTL, (c) SIP_PHIL, and (d) CTL_no_HM. Black solid lines show the linear regression between ice number concentration and height. Only ice particles with diameters larger than 100 μ m from observations and model simulations are included in the comparison. A correction factor of ¹/₄ is applied to the observed ice number concentrations shown in (a) based on Jackson and McFarquhar (2014) and Jackson et al. (2014).



955

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 μ m from observations and model simulations are included in the comparison. A correction factor of ¹/₄ is applied to the observed ice number concentrations based on Jackson and McFarquhar (2014) and Jackson et al. (2014).



967

Figure 12. Bivariate joint probability density function of ice production defined in terms of temperature and ice production, (a) primary ice production; (b) ice production from riming splintering; (c) ice production from rain fragmentation; (d) ice production from ice-ice collision. The ice production (PI, with unit of # L⁻¹) is calculated as ice production rate (L⁻¹s⁻¹) multiplied by model time step (20 mins), shown in Log₁₀.

973

		Type of Secondary ice production References	
	CTL	Rime splintering	[Cotton et al., 1986]
		Rime splintering	[Cotton et al., 1986]
	SIP_PHIL	Ice-ice collision fragmentation	[Phillips et al., 2017]
		Rain freezing fragmentation	[Phillips et al., 2018]
	CTL_no_HM	Same as CTL, but no HM process	
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978			
979			
980			

982	Table 2. The temporally-averaged IWP, LWP (unit: g m ⁻²), and vertically-integrated ice
983	crystal number concentration (unit: m ⁻²) during the four periods from observation, and CTL,

- 984 CTL_no_HM, and SIP_PHIL experiments.

		Multilayer	Single-layer	Transition Frontal	Frontal
		stratus	stratus	Transition	cloud
IWP	OBS	55.6	74.7	5.6	97.0
	CTL	11.2	0.9	0.0001	10.4
	CTL_no_HM	11.1	0.9	0.0001	8.2
	SIP_PHIL	17.1	2.5	3.6	26.1
LWP	OBS	134.4	190.2	58.3	50.2
	CTL	165.1	217.6	88.4	127.6
	CTL_no_HM	166.0	218.0	88.4	129.8
	SIP_PHIL	102.8	131.0	62.1	41.2
ICNC	CTL	5.77×10^{6}	3.22×10^{5}	7.66	2.26×10^{6}
	CTL_no_HM	5.70×10^{6}	3.17×10^{5}	0.77	1.57×10^{6}
	SIP_PHIL	7.09×10^{6}	1.30×10^{6}	4.57×10^{5}	4.67×10^{6}

Table 3. Percentage of occurrence of liquid, mixed-phase, and ice clouds during single
layer mixed-phase clouds from observation, and CTL, CTL_no_HM, and SIP_PHIL
experiments.

	Liquid	Mixed-phase	Ice
OBS (%)	16.0	62.7	22.3
CTL (%)	73.0	26.9	0.1
CTL_no_HM (%)	73.0	26.9	0.1
SIP_PHIL (%)	40.8	58.0	1.2