The influence of multiple groups of biological ice nucleating particles on microphysical properties of mixed-phase clouds observed during MC3E

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31 Abstract:

A new empirical parameterization (EP) for multiple groups of primary biological aerosol particles (PBAPs) is implemented in the aerosol-cloud model (AC) to investigate their roles as ice-nucleating particles (INPs). The EP describes the heterogeneous ice nucleation by (1) fungal spores, (2) bacteria, (3) pollen, (4) detritus of plants, animals, and viruses, and (5) algae. Each group includes fragments from the originally emitted particles. A high-resolution simulation of a midlatitude mesoscale squall line by AC is validated against airborne and ground observations.

Sensitivity tests are carried out by varying the initial vertical profiles of the loadings 39 of individual PBAP groups. The resulting changes in warm and ice cloud microphysical 40 parameters are investigated. The changes in warm microphysical parameters including liquid 41 42 water content, and cloud droplet number concentration are minimal (< 10%). Overall, PBAPs have little effect on ice number concentration (< 6%) in the convective region. In the 43 stratiform region, increasing the initial PBAP loadings by a factor of 1000 resulted in less 44 45 than 40% change in ice number concentrations. The total ice concentration is mostly 46 controlled by various mechanisms of secondary ice production (SIP). However, when SIP is intentionally shut down in sensitivity tests, increasing the PBAP loading by a factor of 100 47 48 has less than a 3% effect on the ice phase. Further sensitivity tests revealed that PBAPs have little effect on surface precipitation as well as on shortwave and longwave flux (< 4%) for 49 100-fold perturbation in PBAPs. 50

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52 **1. Introduction**

In most climate models, the largest source of uncertainty for estimating the total
anthropogenic forcing is associated with cloud-aerosol interactions (Pörtner et al., 2022).
Atmospheric aerosol particles can act as cloud condensation nuclei (CCN) and a few of them

act as ice-nucleating particles (INPs), thereby influencing the microphysical properties of
clouds and, depending on the cloud type (Fan et al. 2010; Chen et al 2019). The treatment of
INP in climate models can strongly affect the atmospheric radiation budget (DeMott et al.
2010). Various sources of aerosol particles, including dust/metallic, marine aerosols,
anthropogenic carbonaceous emissions, and primary biological aerosol particles (PBAPs),
contribute to the observed INPs (Kanji et al. 2017).

A significant amount of global precipitation is associated with the ice phase in cold clouds (Heymsfield and Field 2015; Mülmenstädt et al. 2015, Heymsfield et al. 2020). In particular, mixed-phase clouds are vital for the global climate (Dong and Mace 2003; Zuidema et al. 2005; Matus and L'Ecuyer 2017; Korolev et al. 2017 and references therein). In a multimodel simulation study, Tsushima et al. (2006) showed that the doubling of CO₂ concentrations caused the changes in the distribution of cloud-water in the mixed-phase clouds in a climate simulation to be significant.

69 PBAPs are solid particles of biological origin and are emitted from the Earth's surface (Després et al. 2012). They are highly active in initiating ice as INPs and include bacteria, 70 71 fungal spores, pollen, algae, lichens, archaea, viruses, and biological fragments (e.g., leaf litters, insects) and molecules (e.g., proteins, polysaccharides, lipids) (Després et al., 2012; 72 Fröhlich-Nowoisky et al., 2015; Knopf et al., 2011; Szyrmer and Zawadzki, 1997;). 73 74 Considering the onset temperature of freezing, some ice nucleation active fungi and bacteria (especially *Pseudomonas syringae* with onset freezing temperature around -3°C) are among 75 the most active INPs present in the atmosphere (Després et al. 2012; Hoose and Möhler 76 77 2012). The potential impact of PBAP INPs on cloud microphysical characteristics has been recognized for many years; however, this topic remains a subject of debate (DeMott and 78 Prenni 2010; Spracklen and Herald, 2014; Hoose et al. 2010b). Some previous modeling 79 studies have shown that on a global scale PBAPs have only a limited influence on clouds and 80

precipitation (Hoose et al. 2010; Sesartic et al. 2012, 2013; Spracklen and Heald 2014). On a
global scale, the percentage contribution of PBAPs to the immersion freezing (ice nucleation
by INP immersed in supercooled water drop) is predicted to be much smaller (0.6%) as
compared to dust (87%) and soot (12%) (Hoose et al. 2010).

Many studies have used cloud models to highlight the potential impact of PBAP INPs 85 on cloud microphysics and precipitation (e.g., Levin et al. 1987; Grützun et al. 2008; Phillips 86 et al. 2009). For example, the mesoscale aerosol-cloud model by Phillips et al. (2009) had a 87 3-D domain of about 100 km in width, and many cloud types were present in the mesoscale 88 convective system that was simulated. Their simulations revealed that the cloud cover, 89 domain radiative fluxes, and surface precipitation rate were significantly altered by boosting 90 organic aerosols representing PBAPs. According to Hummel et al. (2018) in shallow mixed-91 phase clouds (i.e., altostratus) when the cloud top temperature is below -15°C, PBAPs have 92 the potential to influence the cloud ice phase and produce ice crystals in the absence of other 93 INPs. 94

The quest for insights into the broader atmospheric role of PBAP INPs for cloud 95 microphysical properties and precipitation is hampered by the limited availability of 96 observations both of their ice nucleation activities for various species and their aerosol 97 distributions in the real atmosphere (Huang et al. 2021). More generally, there is incomplete 98 knowledge about the chemical identity of the key INPs, whether biological or otherwise 99 (Murray et al. 2012). In many global and regional models, the ice nucleation activity of 100 bioaerosols is represented either empirically or theoretically based on laboratory 101 102 measurements of specific biological species of PBAPs that are assumed as representative candidates (e.g., Pseudomonas syringae). This assumption of representativeness introduces 103 uncertainties that would be expected to impact the model results, potentially introducing a 104

bias into the estimation of the effects of bioaerosols on clouds (e.g. Sahyoun et al., 2016;
Hoose et al. 2010b; Spracklen and Herald, 2014, Huang et al. 2021 and references therein).

107 In addition to primary ice nucleation, ice formation in clouds can occur because of processes generating new particles from pre-existing ice, and these are known as Secondary 108 Ice Production (SIP) mechanisms (Korolev and Leisner, 2020; Korolev et al, 2020). SIP can 109 110 have a considerable impact on cloud micro- and macro-physical properties such as precipitation rate, glaciation time, cloud lifetime, and cloud electrification by increasing the 111 ice number concentrations by a few orders of magnitude (e.g., Blyth and Latham 1993; 112 Crawford et al., 2012; Lawson et al., 2015; Phillips et al., 2017b, 2018, 2020; Phillips and 113 Patade, 2021; Sotiropoulou et al. 2021a,b). This in turn can influence the global hydrological 114 cycle and climate (Zhao and Liu 2021). 115

116 However, in many cloud models, the representations of these SIP mechanisms are uncertain as most of the cloud models include only the Hallet-Mossop (hereafter HM; Hallett 117 and Mossop, 1974) process and neglect other SIP mechanisms (e.g. Fan et 2017; Han et al 118 2019). A few secondary ice formation processes (e.g., the HM process) have been suggested 119 to be active in the temperature range where active PBAP INPs exhibit strong ice nucleation 120 activity. The INPs of biological origin such as bacteria are highly active in the temperature 121 range of the HM process (-3 to -8°C) as compared with non-biological INPs (Möhler et al. 122 123 2008; Patade et al., 2021, henceforth PT21). At temperatures warmer than -15°C, some of the PBAPs generated by biologically active landscapes (e.g. forests, woodlands) can promote ice 124 formation and crystal growth in clouds (Morris et al., 2014). 125

In the USA, about 18% of the total landmass is used as cropland, farmland, and agricultural activities (Garcia et al. 2012). These are major sources of biological particles in the atmosphere. Biogenic particles released from crops, either pre- and post-harvest, have

previously been shown to serve as INPs (in Colorado and Nebraska, Garcia et al. 2012). 129 Huffman et al. (2013) found that airborne biological particles increase significantly in 130 concentration, by an order of magnitude or more, during rainfall in a forest in the western US 131 and that bioaerosols are well correlated with INPs. Prenni et al. (2013) observed a similar 132 increase in concentrations of ground-level INPs during rain at a forested site in Colorado, 133 which was associated with increased biological particles. Convective clouds can efficiently 134 135 transport lower tropospheric aerosol particles into the upper troposphere where they can affect the cloud properties (Cui and Carslaw, 2006) 136

The current study aims to simulate realistic concentrations of multiple groups of 137 PBAP INPs, including bacterial and fungal particles, to investigate their interactions with 138 convective clouds observed during the Midlatitude Continental Convective Clouds 139 Experiment field campaign (MC3E), in the USA (Jensen et al. 2016), in the USA. In view of 140 the literature noted above about the effects of PBAP INPs, there is a need for more detailed 141 142 analyses of their role in altering cloud microphysical properties and precipitation because the realistic treatment of ice nucleation activity for major PBAP groups was not available, prior 143 to our empirical scheme (PT21). Hitherto, laboratory measurements of isolated biological 144 species (e.g., Pseudomonas syringae, Cladosporium sp) have been the basis for attempts to 145 simulate biological ice nucleation in clouds, but the representativeness of the choice of such 146 147 species has been a longstanding issue. For example, Hummel et al (2018) considered three highly ice-nucleation-active PBAP species in their model which may not represent the ice 148 nucleation activity of PBAP in the atmosphere. It is not known which biological species of 149 ice nucleation active (INA) PBAPs contribute the most to biological ice nucleation. 150 151 Consequently, there is a need for a new approach oriented toward laboratory measurements of biological INPs sampled from the atmosphere, thus optimizing the representativeness of 152 the data for studies of clouds. 153

In this paper, such an approach is followed to investigate the effect on cloud 154 properties from various major groups of PBAP. We incorporated a recent empirical 155 parameterization for various PBAP groups by PT21 into our 3D aerosol-cloud model (AC). 156 PT21 created an empirical formulation resolving the ice nucleation of each group of PBAPs 157 including 1) fungal spores and their fragments, 2) bacteria and their fragments, 3) pollen and 158 their fragments, 4) detritus of plants, animals, and viruses, and 5) algae. We also examine the 159 160 relative importance of various secondary ice processes in their role in mediating the PBAP effects on cloud microphysical properties, given the weakness of PBAP effects on cloud 161 162 microphysical properties.

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164 **2. Description of observations**

165 2.1 Selected case of a deep convective system

166 In the current study, we simulated a squall line that occurred on 20 May 2011 MC3E (Jensen et al. 2016). The MC3E campaign took place from 22 April through to 6 June 2011 167 and was centered at the Atmospheric Radiation Measurement (ARM) Southern Great Plains 168 169 (SGP) Central Facility (CF), (36.6°N, 97.5°W) in north-central Oklahoma. The surface meteorological analysis on 20 May indicated a southerly flow at the surface, which provided 170 enough moisture from the Gulf of Mexico to trigger convection. Deep convection, organized 171 in the form of a squall line, passed over the measurement site between 1030 and 1100 UTC, 172 resulting in convective precipitation. It was followed by widespread stratiform precipitation 173 that was well observed by both airborne and ground-based measurements. Vertical sounding 174 characteristics of this case are described in Supplement Information (Figure S1) based on the 175 Skew-T plot. 176

178 2.2 Aircraft Observations

The in-situ cloud microphysical observations used in this study were obtained from a 179 180 University of North Dakota Citation II aircraft. The aircraft collected observations of cloud microphysical parameters from the cloud base (1.8 km above mean sea level; hereafter MSL) 181 to a maximum altitude of 7.5 kilometers above MSL. The MC3E campaign collected 182 183 extensive airborne measurements of aerosols and cloud microphysical properties over northcentral Oklahoma. A detailed description of the scientific objectives of the MC3E program, 184 including the field experiment strategy, airborne and ground-based instrumentation, is given 185 in the paper by Jensen et al. (2016). A summary of the airborne instrumentation during 186 MC3E is provided in Supplementary Information. 187

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189 2.3 Ground-based measurements

A comprehensive instrumentation suite deployed at the ARM-SGP central facility provided continuous measurements of atmospheric gases, aerosols, clouds, and local meteorological conditions (e.g., wind, temperature, precipitation, and atmospheric profiles). A cloud condensation nuclei (CCN) counter (CCN-100) (DMT) measured the CCN number concentration at seven supersaturation values with a temporal resolution of 1 hour. Surface precipitation was measured with 16 rain gauge pairs placed within a 6-kilometer radius of the SGP CF.

During the MC3E campaign, the measurement facility deployed at CF measured the spatial variability of surface fluxes of heat, moisture, and momentum. A radiosonde array of 6 sites, covering an area of 300 km \times 300 km, was designed to capture the large-scale variability of the atmospheric state. Radiosonde observations (Vaisala RS92-SGP) were conducted with a 6-hour frequency (four times daily) at around 05:30, 11:30, 16:30, and

202 22:30 UTC, providing vertical profiles of atmospheric state variables (pressure, temperature,
203 humidity, and winds) of the environment surrounding the ARM SGP site. When aircraft
204 operations were planned based on forecasted convective conditions, the sounding frequency
205 was increased to a 3-hour frequency with the starting time at 05:30 UTC.

In addition to airborne observations, the ARM radar network was used to conduct unique radar observations during the MC3E campaign. The information about various radar assets during MC3E is given by Jensen et al. (2016). The surface precipitation used for model validation in this study is a radar-based precipitation estimate as described by Giangrande et al. (2014). Their radar-based rainfall retrievals were in good agreement with observations with an absolute bias of less than 0.5 mm for accumulations less than 20 mm.

212 The Interagency Monitoring of Protected Visual Environments (IMPROVE) network 213 stations close to the location of airborne observations provided ground-level measurements of various chemical species. These included carbonaceous compounds (black and organic 214 carbon), salt, ammonium sulfate, and dust. The details of the measurement techniques used 215 for mass mixing ratios of these compounds are summarized in Malm et al. (1994). The 216 measurements of these aerosol species from various IMPROVE sites, including Ellis 217 (36.08°N, 99.93°W), Stilwell (35.75°N, 94.66°W), and Wichita Mountains (34.73 °N, 218 98.71°W) sites in Oklahoma, were averaged to provide inputs to AC. 219

Initial mass concentrations for the aerosol species of AC (11 species) including sulfate, sea salt, dust, black carbon, soluble organic, biological and non-biological insoluble organic (five groups of PBAPs) were derived from the Goddard Chemistry Aerosol Radiation and Transport (GOCART) model (Chin et al. 2000). The prescribed mass mixing ratios of aerosol species in A are based on IMPROVE observations and are enlisted in Supplementary Information (Table S2). It should be noted that for the MC3E case considered in this study,

coincident IMPROVE measurements were not available. The mean values of the IMPROVE
 measurements conducted on May 18 and 21 are used to prescribe the mass of various aerosol
 species.

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230 **3. Methodology**

231 3.1 Model description

The 'aerosol-cloud model' (AC) used in this study is a cloud-resolving model (CRM) with a 232 hybrid spectral bin/two-moment bulk microphysics, interactive radiation, and semi-233 prognostic aerosol schemes (Phillips et al. 2017b, 2020). The model predicts the mass and 234 235 number concentrations for five types of hydrometeors: cloud liquid, cloud ice (or "crystals"), 236 rain, graupel/hail, and snow. The mixing ratios of the total number and mass of all particles in each microphysical species are treated as model prognostic variables. AC treats all known 237 238 microphysical processes such as droplet nucleation, ice initiation through primary and 239 secondary processes, and growth processes such as deposition/sublimation of ice particles, condensation/evaporation of drops, freezing/melting, as well as coagulation by collisions 240 between various hydrometeor types. Both cloud-base and in-cloud activation of aerosols to 241 form cloud-droplets are treated explicitly, with the predicted in-cloud supersaturation 242 resolved on the model grid being used to activate aerosols aloft. Bin-resolved size 243 distributions of each aerosol species are predicted for the interstitial and immersed 244 components of each aerosol species. Extra prognostic variables track the number of aerosols 245 in each aerosol species that have been lost by INP and CCN activation. 246

247 Secondary ice formation is represented by four types of fragmentation:

breakup in ice-ice collisions (Phillips et al. 2017a, b) (most active between -10 to - 20°C);

- Hallett and Mossop (1974), rime splintering (most active between -3 to -8°C);
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active around -15°C);

fragmentation of freezing rain/drizzle by modes 1 and 2 (Phillips et al. 2018) (most

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• and sublimation breakup (Deshmukh et al. 2021) (most active between -0 to -18°C).

The empirical parameterization (EP) (Phillips et al. 2013) of heterogeneous ice nucleation 254 treats all known modes of ice formation (deposition mode, condensation-/immersion-255 freezing, inside-out and outside-in contact-freezing) in terms of dependencies on the loading, 256 size, and chemistry of multiple aerosol species. In the previous version of the EP, prior to 257 PT21, there were four species of INP aerosol. One of these was PBAP INPs. However, that 258 version of the EP did not resolve the individual types of PBAP INP, which exhibit a wide 259 260 range of ice-nucleating abilities. The current version of AC also includes the ice nucleation 261 (IN) activity of dust and black carbon. The ice nucleation parameterization of dust, as well as black carbon, is based on studies by Phillips et al. (2008) and (2013). The activation of dust 262 and black carbon INP starts at temperatures colder than -10 and -15°C. 263

There are two types of homogeneous freezing represented: that of cloud droplets near -36°C and that of solute aerosols at colder temperatures. Both schemes are described by Phillips et al. (2007, 2009). For cloud droplets, a look-up table from simulations with a spectral bin microphysics parcel model treats the fraction of all supercooled cloud droplets that evaporate without freezing near -36°C, depending on the ascent, initial droplet concentration, and supersaturation. The size dependence of the temperature of homogeneous freezing is represented.

Cloud processes and rainfall formation have been detected using different radar variables, such as specific differential phase K_{DP} . Moisseev et al. (2015), for example, noted an increase in observed K_{DP} because of aggregation. In addition, a few studies have

hypothesized evidence of SIP via K_{DP} (e.g., Sinclair et al. 2016; Kumjian and Lombardo 2017; Carlin et al. 2021). In this study, we attempted to detect secondary ice formation signatures by implementing K_{DP} estimations into AC. Based on Ryzhkov et al. (2011), K_{DP} values were estimated for various hydrometeor types, including cloud drops, raindrops, cloud ice, snow, and graupel (their equations 22, 23, 24, 26, and 29).

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3.2 Empirical formulation for PBAP INPs:

281 In a recent study, PT21 provided an empirical formulation for multiple groups of PBAP INPs based on field observations over the central Amazon. In this study, we modified 282 AC by implementing the recent empirical parameterization of PBAP INPs by PT21. The 283 284 empirical formulation by PT21 is based on observations of PBAP collected at the Amazon Tall Tower Observatory (ATTO), a research site located in the middle of the Amazon 285 rainforest in northern Brazil. The empirical formulation by PT21 for multiple groups of 286 PBAPs includes: - 1) fungal spores (FNG), 2) bacteria (BCT), 3) pollen (PLN), 4) viral 287 particles, plant/animal detritus (DTS), 5) algae (ALG) and their respective fragments are 288 289 implemented in AC. This formulation has empirically derived dependencies on the surface area of each group (except algae) and it applies to particles with diameters greater than 0.1 290 μ m. Additional details about the formulation by PT21 are given in Supplementary 291 292 Information.

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294 *3.3 Model setup*

AC was driven by initial and evolving boundary data for meteorological conditions. The large-scale advection of humidity and temperature tendencies maintained the convection.

Convection was initiated by imposing perturbations onto the initial field of vapour mixing 297 ratio. The large-scale forcing condition used for the simulation was derived using the 298 constrained variational analysis method described in Xie et al (2014). Based on this method, 299 the so-called large-scale forcing including large-scale vertical velocity and advective 300 tendencies of temperature and moisture were derived from the sounding measurements 301 network. During the MC3E campaign, the sounding network consists of five sounding 302 303 stations centered on a sixth site at the ARM SGP central facility. An area with a diameter of approximately 300 km was covered by this sounding network covers. The supplementary 304 305 Figure S2 shows the time height evolution of potential temperature and water vapor mixing ratio from large-scale forcing data. It also shows the time variation of CAPE based on 306 observations. The maximum value of CAPE 2400 JKg⁻¹ was noticed around 12 UTC on 20th 307 May. 308

The model simulations were carried out for a three-dimension domain of 80 km x 80 km with horizontal grid spacings of 2 km. In vertical, the model resolution was 0.5 km, and the model top was located at about 16 km. The lateral boundary conditions are doubly periodic on all sides of the domain. The initial time of the simulations was at 1200 UTC on 19 May 2011 and all simulations were performed for 48 hours at a time step of 10 seconds.

The GOCART model (Chin et al. 2000) was used to initialize the seven chemical 314 species associated with the EP. The data from the three IMPROVE sites mentioned above 315 (Section 2.3) was used to rescale the mass concentration profiles at all levels so that they 316 match the measurements near the surface. Table S2 in Supplementary Information lists the 317 mass mixing ratios of various aerosol species after the corrections. The corresponding 318 vertical profiles of various aerosol species including sulfate, dust, sea salt, black carbon, and 319 total organic carbon are shown in Supplementary Figure S3 (panel a-e). The corresponding 320 321 IMPROVE measurements are also shown in the same Figure. There were no direct measurements of PBAP mass during IMPROVE and therefore it was derived from the measured mass of the total organic carbon (TOC). The relative contribution of insoluble and soluble organic carbon to TOC was assumed to be 20% and 80%, respectively by assuming a water-soluble fraction of 80% for carbonaceous aerosol (Phillips et al. 2017b). AC takes into account the soluble fraction of each type of aerosol. The values of this factor are 0.15 for dust, and 0.8 for carbonaceous species. The value of this fraction for all PBAP groups is 0.1.

There are very few observations available in the literature showing the fraction of 328 PBAP in the insoluble organics or total aerosol particles. For example, observations by 329 Matthias-Maser et al. (2000) found that 25% of the total insoluble particles are biological. 330 PBAPs can contribute a significant fraction to the number concentrations of total aerosol 331 particles (Mattias-Maser et al., 1999). Mattias-Maser and Jaenicke (1995) showed that 332 PBAPs can amount to 20% and 30% of the total aerosol particles. The observation by 333 Jaenicke (2005) in a semi-rural location showed that cellular particles can contribute up to 334 about 50% of total particles. Based on these studies we assumed that 50% of the insoluble 335 organics were biological in origin. The total PBAP loading was prescribed partly based on 336 observations of insoluble organics. The mass fraction of each PBAP group in total PBAP 337 mass is prescribed based on the PT21 observations. The fraction of mass mixing ratio for 338 various PBAP groups is: FNG= 0.39, BCT= 0.13; PLN= 0.31; DTS= 0.17; ALG= 2.5 × 339 10^{-4} . 340

It should be noted that the observations of PBAPs over different geographical locations (including the region where we carried out the simulation) are rare, which prevents us from using the region-specific PBAP observations for the present study. Hence, PT21's default observations were used to calculate the relative contribution of various PBAP groups to insoluble organics. The parameters for the shape of PSD of each PBAP group (modal mean

diameters, standard deviation ratios, and relative numbers in various modes) are prescribed 346 based on observations from Amazon (PT21). Supplementary Figure S4 depicts the 347 corresponding size distribution of various PBAP groups in AC. To check the validity of the 348 observation from PT21 over the region considered in the current study, the model estimated 349 values of one of the major PBAP bacteria are compared with the observations as shown in 350 Supplementary Figure S5. It shows that the estimated values of bacterial number 351 352 concentration are overall in fair agreement with previous observations (e.g. Bowers et al 2009; Bauer et al. 2002; Burrows et al. 2009). The simulated bacterial (~ 10^4 m⁻³) and fungal 353 (~ 10^3 m⁻³) number concentration by AC is in good agreement with their typical 354 concentration in the atmosphere (Després et al. 2012). The resulted vertical profiles of mass 355 of the various PBAP groups are shown in Figure S3 (panel f). 356

From these prescribed loadings of aerosol species, AC predicts their size distribution 357 and hence the CCN activity spectrum. Using the initial sounding and aerosol profile, AC can 358 359 predict the in-cloud size distribution of aerosols in each species as well as in-cloud supersaturation. Figure S6 in Supplementary Information shows the predicted CCN spectrum 360 comparison with observations from the CCN counter at the surface at the SGP site. It should 361 be noted that the aerosol mass loading from IMPROVE observations showed variations of 362 20-30% for the simulated case. The uncertainties in the input aerosol mass loading can result 363 in simulated CCN concentration and are shown by the errors in the CCN concentration 364 predicted by the AC. During 19-20 May, the measured number concentration of active CCN 365 at the SGP CF ranged from 400 to 3000 cm⁻³ at 1% supersaturation (Fridlind et al. 2017). The 366 367 measurements were made on 20 May before the start of the rain in clear air. The normalized CCN number concentrations at 1% supersaturation from observations and AC are ~ 1000 cm⁻ 368 3 and ~ 940 cm⁻³, respectively. Given the wide range of observed CCN concentrations at each 369

supersaturation as well as the uncertainties in the model predicted CCN concentration, thepredicted and observed CCN activity spectra are in acceptable agreement.

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4. Results from control simulation and model validation

374 *4.1 Overview of the control simulation*

An intense north-to-south oriented squall line moved over the ARM SGP CF on May 20, 2011, from 1100 to 1400 UTC (Sec. 2.1). The new version of AC simulated this case, after implementing the empirical formulation by PT21 for multiple groups of PBAP INPs ('control' simulation) (Sec. 3). It should be noted that five ensemble runs were carried out for control simulation (See Table S3 in Supplement Information) varying the perturbing in the initial water vapor mixing ratio.

Figure 1 shows the time-height evolution of various liquid and ice cloud 381 microphysical parameters derived from the control simulation conditionally averaged over 382 cloudy regions. The maximum average cloud droplet number concentration was around 250 383 cm⁻³. The LWC was typically less than 0.5 g m⁻³. The freezing level (0°C) was around 4.1 384 km above MSL. The deep convection began around 10 UTC, followed by intense 385 precipitation around 11 UTC, and reached its peak around 12 UTC. The time-height 386 evolution of cloud ice, snow, and graupel number concentrations shows maxima shortly 387 before 12 UTC, which coincides with the time of peak precipitation. This suggests that the 388 ice phase was important in precipitation formation. 389

The time-height map of simulated radar reflectivity during 20 May, unconditionally averaged over the whole domain is shown in Figure 1g. It shows the well-defined squall line passing over the domain from 1100 to 1500 UTC. The maximum of this domain-wide

simulated reflectivity is around 40 dBZ when deep convection was happening. The
instantaneous maximum of reflectivity at any grid-point (not shown here) was about 50 dBZ.
At other times, the average reflectivity was typical of the stratiform cloud of about 15 dBZ.
The cloud top height of the squall line decreases after 1400 UTC.

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Figure 1: Time-height contours of domain averaged a) cloud water mixing ratio (QCLOUD); b) cloud droplet number concentration (CDNC); c) rainwater mixing ratio (QRAIN); d) number concentration of cloud ice (NICE); e) number concentration of snow (NSNOW); f) number concentration of graupel (NGRAUPEL). Due to a wide range of values, the log values number concentrations are plotted. The surface height is ~ 500 m. The averaging was done for cloud points with LWC > 0.001gm⁻³ or total water content (TWC)> 10⁻⁶ gm⁻³. Also shown is the time-height evolution of domain averaged (g) radar reflectivity.

407 *4.2. Model validation against coincident observations of the storm*

The extended stratiform region of the squall line while in the vicinity of the SGP CF was 408 sampled by the Citation aircraft equipped with a full suite of cloud microphysical 409 instrumentation. The aircraft started sampling the stratiform precipitation region at around 410 1300 UTC and continued the observations at sub-freezing temperatures from 1335 to 1515 411 412 UTC. Occasionally, the aircraft encountered weak convective updrafts (< 6 m/s). The aircraft actively avoided convection that was more vigorous than that. In this section, we validate 413 various microphysical and dynamical quantities from the control simulation against aircraft 414 and ground measurements. The control run includes all primary and SIP processes of ice 415 initiation. The vertical profiles shown here are an average of five ensemble runs. 416

417 Figure 2 compares the aircraft observations against predicted microphysical quantities, with both the predictions and observations identically averaged, conditionally over 418 convective (6 > /w / > 1 m/s) and stratiform regions /w / < 1 m/s). The simulated LWC 419 420 decreases exponentially with height above the cloud base. There is considerable scatter in 421 observed LWC at each level. The various degrees of dilution of sampled parts of the cloud can cause these variations in LWC at a given altitude. The maximum simulated LWC of 0.5 422 gm⁻³ was observed in the convective region at temperatures warmer than -5°C. In the 423 convective region around -5°C, the measured LWC is lower than the simulated LWC by a 424 factor of 3. For the stratiform region, simulated values of LWC are in adequate agreement 425 with observations. Overall, the means of observed LWC are in acceptable agreement with the 426 model results for convective as well as stratiform regions. 427

The vertical profiles of simulated and observed Cloud Drop Number Concentration (CDNC) (Fig. 2c and 2d) showed that CDNC was lower than 300 cm⁻³. In the convective region, the measured CDNC is 40% lower than the simulated CDNC at 15°C. However, an



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Figure 2: Comparison of the control simulations by AC with aircraft observations, for liquid water content conditionally averaged over (a) convective (6 m/s > /w/ > 1 m/s) and (b) stratiform (/w/ < 1 m/s) regions; cloud drop number concentration over (c) convective and (d) stratiform regions; average size of cloud droplets (< 20 μ m) conditionally averaged over (e) convective and (f) stratiform regions. All the vertical profiles shown here are averaged for the whole domain. The error bars were estimated based on five ensemble runs.

adequate agreement between them is found around -5°C. For the stratiform region, simulated 439 CDNC is much higher in the mixed-phase region. However, at a temperature warmer than 440 0°C the values of observed CDNC are in acceptable agreement with observations. 441 The observed and simulated mean diameter of cloud droplets varied between 6 to 16 µm (Figures 442 2e and 2f). There are few points in the convective region e.g., around -5°C, where the 443 observed cloud drop diameter is 50% lower than the simulated value. An adequate agreement 444 445 between simulated and observed cloud drop diameter was found for the stratiform region. Overall, the predictions of average CDNC and cloud droplet diameter, in both convective and 446 447 stratiform regions, show a fair agreement with observations.

The ice particle number concentration from observations and the control simulation is 448 also compared as shown in Figures 3a and 3b for convective and stratiform regions, 449 respectively. It should be noted that the observed number concentration of ice particle 450 particles smaller than 200 µm is prone to shattering, even with the use of the shattering 451 452 correction algorithm. This can introduce a significant bias in the observed ice number 453 concentration (Korolev et al., 1991). To avoid these biases, we have compared the number concentration of ice particles with a diameter greater than 200 µm from both observation and 454 model (denoted by 'NT200'). However, in the rest of the manuscript (in sensitivity studies), 455 the number concentration from the model included ice particles of all size ranges. 456

457 Observations show that the concentration of ice particles gradually increases as the 458 temperature decreases, as expected. The maximum ice number concentration from the aircraft 459 observations (with $D > 200 \,\mu$ m) is ~ 0.06 cm⁻³ around -15 °C. Good agreement to within 50% 460 at most levels, was found between the model simulated NT200 and that observed for the 461 convective region.



Figure 3: Comparison of the control simulations by AC with aircraft observations, for ice 463 number concentration of all particles > 0.2 (NT200) mm in the maximum dimension of all 464 microphysical species (cloud ice, graupel/hail, snow), averaged over (a) convective (6 m/s > 465 466 |w| > 1 m/s) and (b) stratiform (|w| < 1 m/s) regions. (c) The vertical profile of simulated radar reflectivity conditionally averaged over all regions of significant reflectivity (> -20 467 dBZ) at each level is compared with observations from ground-based radars. The temperature 468 corresponding to each altitude is mentioned on the right axes; (d) predicted precipitation rate 469 (mm/hr) compared with ground observations at the SGP CF. All the vertical profiles shown 470 here are averaged for the whole domain. The error bars were estimated based on five 471 ensemble runs. 472

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In the stratiform region, at most levels, model values of NT200 have the same order of magnitude as observations. However, between about the -10 and -16°C levels, the stratiform NT200 values are about half an order of magnitude lower than the observations. In similar simulations of the 20 May case, Fan et al. (2015) and Fridlind et al. (2017) also showed underestimation of simulated ice number concentrations. Compared to observations,

their simulations showed half an order of magnitude bias in ice crystal number concentration. 479 Comparatively, for the convective region, our model predicted ice number concentrations 480 were in better agreement with observations. As mentioned in section 2.2, imaging probe data 481 is prone to shattering, and various corrections were used to rectify it. However, there are 482 currently no ways to determine how many undetected artifacts remain after shattering 483 corrections have been applied (Baumgardner et al. 2022). Such uncertainties in measured ice 484 485 number concentration could result in such bias in observed and simulated ice number concentrations. In summary, though the AC model is not totally perfect, it did a fair job in 486 487 simulating observed ice number concentrations.

In Figure 3c, the radar reflectivity from vertically pointing Ka-band ARM zenith radar 488 is compared with the mean profile from model simulations. This figure illustrates that 489 simulated reflectivity profiles below roughly 3 km and above 8 km MSL altitudes are in good 490 agreement with observations. Between 3 and 8 km MSL (temperatures of 2 and -30°C), the 491 492 bias in reflectivity from model simulations and observations is about 10 dBZ. Thus, the simulated reflectivity is substantially higher than observed, particularly at levels where the 493 aircraft sampled the clouds. Fridlind et al. (2017), as well as Fan et al. (2015), noticed similar 494 overestimations of reflectivity within stratiform outflow of the squall line case on 20 May. 495 They attributed the reflectivity biases to significantly larger ice particles in the simulations 496 than observed. 497

Figure 3d compares the time series of precipitation rate from the control simulation with the radar-based precipitation estimates. In both, control simulation and observations, a maximum precipitation rate of about 5 mm/hr was noticed, with an error in the prediction of less than 5%. In comparison to observations, the simulated squall line arrives 1-2 hours later. The lack of resolution of the 3D turbulence in the planetary Boundary Layer and uncertainties associated with the 3D structure of initial and boundary conditions can all have an
independent impact on the simulated rainfall structure, resulting in a delayed peak.
Nonetheless, AC has done a fair job in simulating the peak in the predicted precipitation rate.

506

507 *4.3 Analysis of simulation with ice particle budgets and tagging tracers*

508 The activated PBAP INPs from the control run are shown in Figure 4 for the convective and stratiform regions. In addition to the PBAP INPs, Figure 4 also shows the activated INPs 509 510 from dust and black carbon. It should be noted that these concentrations shown here are based on advective tagging tracers that follow the diffusion, ascent, and descent inside cloud 511 motions. Overall, bacterial, and fungal particles dominate the biological INP concentration in 512 the simulated cloud. For example, at -20°C the activated INPs from bacteria and fungi are 513 higher than the other three groups of PBAP INPs (detritus, pollen, algal) by two orders of 514 515 magnitude in both convective as well as stratiform regions. At that level in convective regions, the average concentration of simulated active PBAP INPs is about 3×10^{-6} cm⁻³, 516 which is two orders of magnitude less than the maximum total for all active INPs (about 517 3×10^{-4} cm⁻³) in the whole simulation. Overall, the predicted total INP concentration is 518 dominated by black carbon and dust. At -10°C, the Activated INPs from dust and black 519 carbon differ by an order of magnitude from the total PBAP INPs in convection. 520





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Figure 4: The activated number concentration INPs from various PBAP groups along with dust (DUST) and black carbon (BC) and total INPs at various temperatures for (a) convective and (b) stratiform regions. All the vertical profiles shown here are averaged for the whole domain.

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The formation of ice in a cloud is a result of several primary and secondary processes. 528 529 It is important to understand the relative importance of these processes in precipitation formation. To that end, Figure 5a shows the ice particles initiated from various sources 530 throughout the 3D domain of the entire simulation. The primary homogeneous (PRIM_HOM) 531 dominates the total ice budget. Among all SIP mechanisms, breakup caused by collisions 532 between various ice particles is the most important in determining total ice number 533 concentration. The ice production by sublimation breakup of graupel is slightly lower than 534 PRIM_HOM. However, the contribution of ice production via sublimation breakup of 535 dendritic ice crystals is negligible. 536

537



Figure 5: (a) Ice crystal budget for simulated MC3E case. The number of ice crystals 540 produced by various mechanisms (as shown in the legend box) per 10^{15} particles is shown. 541 Also shown is the relative contribution of various SIP mechanisms such as sublimation 542 breakup, raindrop freezing, ice-ice collision breakup, and the Hallett-Mossop process to the 543 total ice number concentration as a function of temperature, averaged conditionally over only 544 (b) convective and (c) stratiform regions. The relative contribution was calculated based on 545 advective tagging tracers for the given process. The convective and stratiform regions were 546 identified based on criteria |w| > 1 and |w| < 1, respectively. 547

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Figure 5b and 5c depict the relative importance of ice concentration from various SIP mechanisms, as well as active INPs in determining total ice number as a function of temperature for convective and stratiform regions. Each source of ice displayed is tracked with advective "tagging tracers" throughout the simulation. Overall, at temperatures warmer than -15°C, the contribution to the total ice number concentration from various SIP is 2-3

orders of magnitude higher than the concentration of active INPs, highlighting the importance 555 of SIP mechanisms in ice formation. At -25°C, breakup in ice-ice collisions contributes 556 around 75% and 20% of the total ice concentration in the convective and stratiform regions, 557 respectively. At the same temperature, in both convective and stratiform regions, sublimation 558 breakup and raindrop freezing contribute about 8% and 0.8 %, respectively. It can be 559 observed that in the convective regions at temperatures warmer than -30°C, SIP mechanisms 560 561 are important in determining the total ice concentrations, whereas at colder temperatures homogeneous nucleation is dominant. In the stratiform region, this crossover occurs at a 562 563 much warmer temperature around -18°C. At temperatures colder than this homogeneous nucleation is a major contributor to the total ice whereas at warmer temperatures SIP 564 mechanisms prevail. Overall, the contribution of active INP to the total ice is lower than 3%. 565

Secondary ice formation via the HM process of rime-splintering contributes 566 significantly to ice production at temperatures warmer than about -15 °C (Fig. 5b and 5c), 567 enhancing the ice concentration beyond the primary ice. In the convective region, the 568 contribution of the HM process in total ice can reach as high as 40% around -5°C. The 569 simulated cloud droplet diameter is mostly smaller than 15 µm. It is smaller than the cloud 570 droplet size required for the HM process to occur. In AC, the rate of the rime-splintering 571 mechanism depends on the concentration of droplets > 24 μ m. It should be noted that in the 572 573 AC model HM process is treated with a factor multiplying the fragment emission which depends on the cloud droplet size. This factor is zero for cloud diameter below 16 µm and 574 unity above 24 µm with linearly interpolated in between. 575

576

577 5. Results from sensitivity tests about the influence of PBAP

To quantify the effect of multiple types of PBAPs on cloud properties, sensitivity tests were performed by modifying the control simulation and comparing the perturbed simulations with it. Description of various sensitivity tests carried out in the current study are enlisted in Supplementary Information (Table S3). The corresponding figures for each simulation are also mentioned.

583 Simulations were performed by eliminating all PBAPs from the control ('no-PBAP' case) and by multiplying their initial loadings at all levels by factors of 10 and 100 ('high-584 PBAP' and 'very high-PBAP' cases) respectively. Comparison with the control simulation 585 reveals the overall effect from both the CCN and IN activities of all bioaerosols combined. 586 These factors are justified by considering the variations in PBAP concentrations in the range 587 of about 0.1 to 30 L⁻¹ over North American forests (Huffman et al. 2013). An additional 588 simulation was conducted with a 1000-fold increase in initial PBAP loading ('ultra high-589 PBAP') to investigate if these unrealistically high concentrations of PBAPs could affect the 590 591 ice phase in a purely hypothetical scenario. Five ensemble runs were carried out for all major simulations involving perturbations in PBAP loading. The ensemble runs were carried out by 592 varying the perturbation in initial conditions (water vapor mixing ratio). 593

Additional simulations were performed by removing treatment of biological IN activity in the EP (*'no-PBAP INP'* case) relative to the control run. A comparison of both additional simulations against the corresponding simulations with the full change in the PBAP loadings (no-PBAP and high-PBAP cases) reveals the separate roles of the INP and CCN activities for the changes in biological material. Apart from these changes in PBAPs, the perturbed simulations are identical to the control run.

Figure 6 reveals the effects of all bioaerosols on cloud properties in the convective region (/w/ > 1 m/s). Overall, changes in cloud microphysical properties including liquid water content, cloud droplet size, cloud drop number concentration, ice number concentration



Figure 6: The temperature dependence of the (a) liquid water content, the (b) cloud droplet number, (c) the cloud droplet diameter, and the (d) total ice number concentration for 'control' simulation and various sensitivity runs involving a change in total PBAP number concentrations for in the convective region. The averaging conditions are mentioned at the top of each figure. The ice number concentration from the ultra high-PBAP is also shown in panel d. All the vertical profiles shown here are averaged for the whole domain.

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are less sensitive to the changes in PBAPs for the convective part of the simulated clouds and are not statistically significant. The LWC, cloud droplet number, and cloud drop diameter in the perturbed simulations do not differ much (< 3%) from the control run. For the whole storm, considerable changes in the spatial distribution of total ice number concentration are observed due to changes in PBAPs (see Supplementary Figure S7). However, vertical profiles showed very small changes in the ice number concentrations. In the convective region, changes in ice crystal number concentration due to changes in PBAPs are negligible (< 6%).
This includes the extreme changes in bioaerosol loading (ultra high-PBAP case).



Figure 7: The temperature dependence of (a) the liquid water content, (b) the cloud droplet number, (c) the cloud droplet diameter, and the (d) total ice number concentration for 'control' simulation and various sensitivity runs involving a change in total PBAP number concentrations for in the stratiform region. Also shown is the temperature dependence of (e) ice concentration from homogeneous freezing. The averaging conditions are mentioned at the top of each figure. The total ice number concentration and ice number from homogeneous

freezing from ultra high-PBAP are also shown in panels d and e. All the vertical profilesshown here are averaged for the whole domain. The error bars are based on ensemble runs.

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Figure 7 shows the corresponding effects in the stratiform region (/w) < 1 m/s) from 629 all bioaerosols. The changes in warm microphysical properties because of changes in PBAP 630 loadings are smaller than 10%. In this part of the cloud, the ice-microphysical parameters are 631 632 comparatively more sensitive to the changes in PBAP than in the convective region. The ultra high-pbap case predicted ~40% lower ice number concentration than the control run. 633 However, these changes in ice number concentration are not significant as the error bars 634 associated with ensemble members overlap. For the stratiform region, all other simulations 635 considered here showed < 10% change in ice number concentrations compared to the control 636 run. These changes in ice number concentration due to PBAPs are mostly controlled through 637 their effect on homogeneous freezing above the -36°C level as shown in Figure 7e by tagging 638 tracer for homogeneous nucleation. These ice particles can then advect to lower levels 639 640 affecting ice number concentrations in the mixed-phase region.

Figure 8 shows the number of ice particles generated by homogeneous nucleation, 641 various mechanisms of primary nucleation (8a), and secondary ice production (8b) per 10^{15} 642 ice particles for the entire storm. Homogeneous freezing dominates the ice production among 643 the three broad types of ice formation mechanisms (heterogeneous and homogeneous ice 644 645 nucleation, SIP). The maximum changes in ice nucleated through the primary ice mechanism are noticed for the very high-PBAP case and can be attributed to the 100-fold increase in all 646 PBAP loading. The very high-PBAP simulation predicted a 15% lower number of 647 homogeneously nucleated ice than the control run. The very high-PBAP cases predicted 648 about 80% more primary ice crystals formed at temperatures warmer than -30°C. At 649 temperatures colder than -30°C, this case predicted 20% more primary ice crystals than the 650 control run. The very high pbap case showed an increase in primary heterogeneous ice and a 651

decrease in primary homogenous ice. Since the contribution of primary homogenous ice 652 nucleation is much higher in determining the total ice number concentration when compared 653 with primary homogeneous nucleation, the overall effect of the very high pbap case is a 654 decrease in total ice number concentration as shown in Figure 7 and Table S4 in the 655 Supplementary Information. However, at temperatures warmer than -35°C, ice number 656 concetration in the very high pbap case was comparable with the control run (Table S5). 657

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Figure 8: The number of ice crystals produced during the whole storm by (a) primary ice 661 nucleation mechanisms and homogeneous freezing as well as (b) various SIP mechanisms (as 662 shown in the legend box) per 10¹⁵ particles is shown for various sensitivity runs. The ice 663 enhancement ratio for the convective and stratiform regions is shown in 8c and 8d. 664

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Figure 8b shows that among SIP mechanisms, the contributions of ice-ice collision 667 breakup and sublimation breakup are higher by an order of magnitude than the HM process 668 and raindrop fragmentation. However, the budget analysis (not shown in the plot) showed 669 that about 75% of the fragments associated with sublimation breakup are prone to 670 evaporation, making ice-ice collision breakup a major SIP mechanism. The estimated ice 671 enhancement ratio, which is a ratio between the number concentrations of total ice (excluding 672 673 homogeneous nucleation) and primary ice, is shown in Figures 8c and 8d for convective and stratiform regions respectively. Overall, the ice enhancement ratio varied between 10 to 10^4 674 which indicates the importance of SIP mechanisms. The budget analysis shows that overall, 675 the perturbations in bioaerosols resulted in very small changes (with maximum change < 676 40%) in ice generated by SIP mechanisms. 677

The role of PBAPs in altering radar reflectivity and surface precipitation was limited and described briefly in Supplementary material (Figure S8). The overall effect of PBAPs on accumulated surface precipitation was minimal (< 4%) (Figure S8 and Table S4). In addition, the changes in PBAPs do not show a significant impact on shortwave and longwave radiation fluxes as well cloud fractions as discussed in the Supplementary information (Figure S9).

683

684 6. Results from sensitivity tests about secondary ice production

Various sensitivity experiments were conducted to evaluate the role of SIP mechanisms in determining micro- and macrophysical parameters of the clouds (See Table S3). SIP through sublimation breakup and breakup in ice-ice collisions were switched off in the 'nosublimation breakup' and 'no-collisional ice-ice breakup' simulations, respectively. In the 'nosecondary' case, no SIP mechanisms were active. The results from these sensitivity experiments are shown in Figure 9 for the convective as well as the stratiform region of the simulated cloud. Overall, in the convective region, the no-secondary and no-collisional ice-ice breakup cases predicted 5 and 12% higher LWC respectively, than the control run (See Table S4 in Supplementary Information). In the stratiform region, these cases predicted ~25% higher LWC than the control run. Lower ice number concentrations due to the absence of SIP mechanisms may reduce the rate of conversion of liquid to ice via mixed-phase processes, resulting in a higher LWC.

In the convective part, the absence of any SIP increased ice number concentration by 697 half an order of magnitude at temperatures warmer than -25°C. Comparing the no-SIP and 698 control cases, the effect of the inclusion of SIP mechanisms is to increase the average ice 699 concentration by up to half an order of magnitude at temperatures warmer than -15°C in the 700 701 stratiform region. For the stratiform region, at temperatures colder than this, the absence of SIP mechanisms resulted in higher ice number concentrations by a similar magnitude. These 702 703 changes at the colder levels are associated with homogeneous droplet freezing. The changes in ice number concentration in the no-collisional ice-ice breakup case are comparable with 704 the no-secondary case. Compared to break up in ice-ice collisions, sublimation breakup has a 705 706 lower impact (< 40%) on the total ice number concentration in both convective and stratiform 707 regions.

The changes in simulated radar reflectivity, total specific differential phase, and surface precipitation rate with SIP mechanisms are shown in Figures 9e, 9f, and 9g, respectively for the whole storm. Overall, the simulated radar reflectivity was 1 dBZ lower in the no-SIP and no-collisional ice-ice breakup case than in the control run and can be attributed to the overall increase in ice number concentration in the control run.



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714 Figure 9: Temperature dependence of the liquid water content in (a) the convective and (b) the stratiform region for 'control' simulation and various sensitivity runs involving SIPs. The 715 ice number concentration is also shown for the (c) convective and (d) stratiform regions. 716 717 The averaging conditions are mentioned at the top of each figure. The vertical profiles of (e) radar reflectivity, and (f) total specific differential phase are also shown for the same 718 simulations. (g) The temporal evolution of the total surface precipitation rate averaged over 719 720 the domain is also shown. All the vertical profiles shown here are averaged for the whole 721 domain.

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The no-sublimation case predicted slightly higher reflectivity than the control run. The absence of all SIPs resulted in about a 100% decrease in the $K_{\rm DP}$ at a temperature colder than -40°C. Between -10°C to -30°C, the absence of no-collisional breakup and no-secondary resulted in higher $K_{\rm DP}$ (half an order of magnitude) values than the control run. The absence of all SIP mechanisms results in a higher surface precipitation rate (75%) during the peak rainfall hour, which occurs around 11.30 UTC compared to the control run. In the previous study, Phillips et al. (2017b) have shown that SIP through ice-ice collision breakup can reduce accumulated surface precipitation in the simulated storm by 20%-40%. They
attributed it to the increase in snow particles competing for available liquid and the reduction
in their growth by riming. It resulted in smaller ice particles and a reduction in surface
precipitation.

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735 7. Results about the influence of PBAPs in the absence of SIP mechanisms

To investigate the role of PBAPs in altering cloud microphysical properties through SIP mechanisms, an additional simulation was performed by eliminating all secondary ice processes from the control run and multiplying the initial loading of all PBAP groups by a factor of 100 (the '*very high-PBAP with no SIP' case*). The results of this simulation are then compared to the no-SIP case as shown in Figure 10.

In the absence of any SIP mechanisms, the 100-fold increase in bioaerosols resulted in 741 742 minimal effect on ice number concentration. Overall, without SIP the increase in bioaerosol loading by 100-fold resulted in less than a 5% change in ice number concentration. This 743 indicates that the ice produced by various SIP mechanisms does not alter the effect of 744 745 bioaerosols on-ice number concentration in the simulated clouds. The changes in simulated radar reflectivity due to a 100-fold increase in bioaerosols are negligible (< 0.5%) (Figure 746 10c). The difference in predicted surface precipitation rate and accumulated precipitation 747 between very high-PBAP with no-secondary and no-secondary cases was lower than 3%. 748



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Figure 10: The temperature dependence of ice number concentration for the control, very high-PBAP with no SIP and no-SIP simulations averaged for (a) convective and (b) stratiform regions. The (c) vertical profile of radar reflectivity and the temporal evolution of (d) surface precipitation rate are shown for the entire simulation. All the vertical profiles shown here are averaged for the whole domain. All the vertical profiles shown here are averaged for the whole domain.

756

757 8. Discussion

Five PBAP groups have been implemented in the mesoscale AC model to predict their ice nucleation activity based on the empirical formulation by PT21. The simulated concentrations of major PBAPs including fungi and bacteria are of the same order of magnitude as results from previous modeling studies (Després et al., 2012; Hoose et al., 2010b). Still, the relative abundance of PBAP groups over the simulated region is unknown due to the lack of measurements. The AC model was run with higher resolution (2 X 2 km) compared to previous studies on a global scale (Hoose et al., 2010b), to investigate the potential impact of variations in PBAP concentration on the properties of simulated squall line events moreclearly.

Yet the control simulation is not perfectly accurate in all respects. In the stratiform region between -10 and -16°C, the predicted ice number concentration was lower than observed by aircraft by half an order of magnitude and in fair agreement at temperatures warmer than -10°C. This uncertainty factor is similar to the uncertainty in the measurements due to various biases (e.g., Field et al. 2006. Nevertheless, all other simulated cloud microphysical parameters, radar reflectivity, and surface precipitation rate were in acceptable agreement with aircraft and ground-based observations.

In the control simulation, the average ice concentration above the -30°C and -18°C levels is dominated by downwelling of homogeneously nucleated ice from above the mixedphase region in convective and stratiform clouds respectively. Below both levels, SIP prevails. Both processes of ice initiation (homogeneous freezing and SIP) have only weak sensitivity to PBAPs, hence the weakness of the impact on simulated cloud glaciation.

779 Based on the sensitivity experiments, it can be concluded that PBAP INPs have only a 780 limited effect on the average state of the ice phase of the simulated clouds of this mesoscale 781 convective system. Most of the changes in ice number concentration associated with changes in PBAPs are controlled by their effects on homogeneous nucleation and SIPs. The lower 782 783 dependence of simulated ice number concentration on changes in PBAPs is consistent with the findings of Hummel et al. (2018). Based on ensemble simulations of the regional 784 atmospheric model for Europe, they showed that the changes in average ice crystal 785 786 concentration by biological INPs are very small and are not statistically significant, implying 787 that PBAPs play only a minor role in altering the cloud ice phase. The limited effect of

PBAPs on cloud properties on a global scale has been highlighted in previous studies (Hoose
et al., 2010b; Sesartic et al., 2012, 2013; Spracklen and Heald, 2014).

790 The weakness of the simulated impact from realistic PBAP fluctuations is explicable mostly in terms of the low contribution from biological ice nucleation compared to non-791 biological INPs to overall ice initiation. In terms of ice nucleation efficiency and onset 792 793 temperatures, each PBAP group has different ice nucleation properties. Based on vertical profiles of active INPs (Figures 4), the overall contribution of activated INPs from all PBAP 794 groups to the total active INPs was ~1%. At -15°C, temperature, the active INPs from dust 795 and black carbon was one order higher than PBAP INPs. At -30°C, the predicted INPs from 796 dust and black carbon were higher by one and two orders of magnitude, respectively, than 797 PBAP INPs. The dust and black carbon INPs activated at these temperatures can be advected 798 down to the levels where PBAP INPs are most important. Overall, this resulted in low 799 800 sensitivity of the average ice phase to the changes in bioaerosol loading.

801 The ice production in the simulated cloud system at levels in the mixed-phase region (0 to -36°C) is largely controlled by various SIP mechanisms of which the most important is 802 the breakup in ice-ice collisions. Some of these processes are active at temperatures warmer 803 than -15°C (e.g., the HM process) where PBAP INP are important and expected to enhance 804 805 the biological ice nucleation. However, our results showed that the ice production associated 806 with SIP mechanisms is less sensitive to the initial PBAP loading because SIP causes positive feedback of ice multiplication with ice fragments growing to become precipitation-size 807 particles that then fragment again. 808

In our study, a 100-fold increase in PBAPs leads to a < 4% change in surface precipitation. Using mesoscale model simulations, Phillips et al. (2009) reported a 10% increase in accumulated surface precipitation associated with deep convective clouds due to a

100-fold increase in biological particles. Phillips et al. (2009) also noted an effect (up to 4%)
on surface shortwave and TOA longwave radiation flux because of changes in PBAP number
concentration. In our study, the changes in PBAP loading caused smaller changes in
simulated shortwave and longwave fluxes (< 3%). Sesartic et al. (2012, 2013) showed that
including fungi and bacteria in the global climate model leads to minor changes (< 0.5%) in
the ice water path, total cloud cover, and total precipitation.

It should be noted that the sensitivity experiments carried out in the current study are limited to the small domain (80 X 80 km domain) representing a limited area of the global ecosystem. Also, the model top was located at 16 km, and it may not represent the whole atmosphere. The results presented here are based on a mesoscale model and may not represent the global impact of PBAPs on clouds.

823

824 **9.** Conclusions

A framework describing the ice nucleation activity of five major groups of PBAPs including 825 fungal spores, bacteria, pollen, viral particles, plant/animal detritus, algae, and their 826 827 respective fragments was provided by PT21. The ice nucleation activity of these major PBAP groups in the EP was based on samples from the real atmosphere. The present study 828 implements this EP in AC and investigates the role of these five PBAP groups as INPs in 829 deep convective clouds. The high-resolution (2 km horizontally) simulations over a 830 mesoscale 3D domain (80 km wide) using AC elucidate the impact of these PBAP groups on 831 the cloud properties. A series of sensitivity experiments were conducted to test the impact of 832 PBAP groups on cloud properties. 833

A mid-latitude squall line that occurred on 20 May 2011 during MC3E over the US 835 Southern Great Plains is simulated with the model. The simulated number concentration of 836 ice particles showed good agreement (to within about 50%) with aircraft observations for the 837 convective clouds within the mesoscale system. In the stratiform region between -10 and -838 16°C, the model predicted ice number concentration was lower than the aircraft observation 839 by half an order of magnitude and in fair agreement at temperatures warmer than -10°C. 840 841 Various sensitivity experiments were carried out by perturbing the initial PBAP loading and by altering various SIP mechanisms. 842

Each PBAP group has diverse properties including its shape, size, and abundance in 843 the atmosphere. A small fraction of PBAPs is found to be ice nucleation active and can 844 therefore act as PBAP INPs. The relative contribution of each PBAP within the total PBAPs 845 may vary from one ecosystem to another. In the current study, their relative contribution is 846 based on previous observations from Amazonia and can be considered as the main limitations 847 of this study. However, the simulated number concentrations of major PBAPs including 848 fungi, and bacteria look reasonable and are close to their typical abundance in the 849 atmosphere. 850

Any perturbation in the PBAP concentration by factors upto 1000 assumed in the 851 current study (resulted in maximum changes in ice number concentration by < 6% convective 852 853 region and by < 40% in the stratiform region with respect to the control run. The simulations showed that simulated ice particle number concentration is much higher than the number 854 concentrations of PBAP INPs. Even at temperatures warmer than -15°C, where PBAP INPs 855 856 are thought to be the most important INP, ice crystals originated from primary heterogeneous nucleation of dust and black carbon from higher levels of the cloud frequently perturb the 857 lower levels due to sedimentation. The major ice formation comes from SIP mechanisms and 858

homogeneous nucleation, both are less sensitive to the changes in PBAPs. Therefore, PBAPINPs do not show a significant impact on the average ice phase of the simulated storm.

PBAPs have minimal effect on the warm microphysical properties of simulated
clouds. The effect on liquid water content and cloud droplet number concentration was lower
than 10% in both convective and stratiform regions. Since both ice and warm microphysical
processes are less sensitive to PBAPs, surface precipitation is not affected significantly by
changes in PBAPs. A 100-fold increase in all PBAPs resulted in less than a 5% change in
surface precipitation.

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Code and data availability: Data and the code for the empirical formulation of PBAPs are
available on request by contacting the corresponding author.

870 *Competing interests*: The authors declare no conflict of interest

Author Contributions: VJTP designed and monitored this study. SP conducted model
simulation, most of the data analysis, and wrote the initial manuscript. All authors contributed
to the scientific discussion and model development.

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