Investigating the contribution of secondary ice production to in-cloud ice crystal numbers

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Abstract In-cloud measurements of ice crystal number concentration can be orders of magnitude higher than the precloud ice nucleating particle number concentration. This disparity may be explained with secondary ice production processes. Several such processes have been proposed, but their relative importance and even the exact physics are not well known. In this work, a six-hydrometeor-class parcel model is developed to investigate the ice crystal number enhancement, both its bounds and its value for different cloud states, from rime splintering and breakup upon graupel-graupel collision. The model also includes ice aggregation and droplet coalescence, ice hydrometeor nonsphericity, and a time delay formulation for hydrometeor growth. Conditions to maximize the breakup contribution, as well as the effects of nonsphericity and turbulence, are discussed. We find that the largest enhancement of ice crystal number occurs for “intermediate” conditions, characterized by moderate updrafts and aerosol concentrations, and when ice hydrometeor nonsphericity is included.

Plain Language Summary Ice formation processes occurring in clouds can significantly influence precipitation, the hydrological cycle, and Earth’s climate. Mechanical generation of ice, called ice multiplication or secondary ice production, can have a profound impact on cloud development. Yet secondary ice production is poorly understood and rarely described in models. This work discusses development of a model to simulate in-cloud ice crystal formation, both by nucleation and by secondary production, for a broad range of thermodynamic conditions. We find that the largest enhancement of ice crystal number occurs for “intermediate” conditions, such as the moderate updrafts and aerosol loadings within maritime convective clouds. In such cases, ice crystal number can be enhanced by a factor of 104. Our results indicate the importance of including secondary ice formation in forthcoming climate model cloud schemes.

1. Introduction

In-cloud phase partitioning between liquid droplets and ice crystals determines cloud radiative forcing [e.g., Fowler and Randall, 1996]. The number of ice-nucleating particles (INP) has a strong influence on this partitioning and is an initial upper limit to ice crystal numbers in mixed-phase clouds [Choi et al., 2010; Kormurcu et al., 2014]. But ice crystal numbers significantly higher than the precloud INP numbers have also been observed at warmer subzero temperatures in field studies over the past 40 years [e.g., Mossop and Hallett, 1974; Mossop, 1985; Hobbs and Rangno, 1985; Rangno and Hobbs, 1991; Beard, 1992; Rangno and Hobbs, 2001; Crawford et al., 2012]. Older measurements are subject to shattering artifacts, as ice crystals collide with the tips of aircraft-mounted cloud probes [Heymsfield, 2007; McFarquhar et al., 2007; Baker et al., 2009]. In an effort to mitigate these artifacts, probe tips (called K-tips below) have been developed to minimize inlet surface area, airflow disturbances around this inlet, and any inefficient heating that leads to ice build-up on the probe.
Table 1. ICNC Enhancements From Observations Relevant to This Study*

<table>
<thead>
<tr>
<th>Source</th>
<th>Technique</th>
<th>Enhancement</th>
<th>Conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Crawford et al. [2012]</td>
<td>CDP, CIP-100, 2DS (CAS and APPRAISE)</td>
<td>(N_t \sim \sigma ) (100 L(^{-1}))</td>
<td>S. England; shallow convection</td>
</tr>
<tr>
<td>2. Crosier et al. [2014]</td>
<td>CDP, CIP-15, and 100 (with IATF) APPRAISE</td>
<td>(N_t ) (2 km, (-8^\circ) C) = 10–100 L(^{-1})</td>
<td>S. England; cold frontal rainband</td>
</tr>
<tr>
<td>3. Heymsfield and Willis [2014]</td>
<td>CAS (0.5–50 (\mu)m with IATF) NAMMA</td>
<td>(\bar{N}_t ) ((D_t &gt; 125\mu)m) = 108.5 L(^{-1})</td>
<td>Cape Verde; mesoscale convection</td>
</tr>
<tr>
<td>4. Lawson et al. [2015]</td>
<td>CPI, 2D-S, FFSSP, FCDP ICE-T</td>
<td>(\bar{N}_t ) = 572 L(^{-1})</td>
<td>St. Croix; developing cumuli cores</td>
</tr>
<tr>
<td>5. Lasher-Trapp et al. [2016]</td>
<td>FFSSP (2–47 (\mu)m), 2D-C (both with IATF); SID-2H (2–50 (\mu)m) ICE-T</td>
<td>(N_t^{\text{max}} ) ((D_t &gt; 100\mu)m) = 101–126 L(^{-1})</td>
<td>St. Croix; maritime cumuli</td>
</tr>
<tr>
<td>6. Taylor et al. [2016]</td>
<td>CDP, 2D-S, CIP-100 COPE</td>
<td>(\bar{N}_t ) = 137 L(^{-1})</td>
<td>SW peninsula of the UK</td>
</tr>
<tr>
<td>7. Ladino et al. [2017]</td>
<td>FSSP-100, OAP-2DC (50–1600 (\mu)m) HAIC-HIWC</td>
<td>(\bar{N}_t \approx 200 L^{-1})</td>
<td>Cayenne, French Guiana</td>
</tr>
</tbody>
</table>

*Campaigns: APPRAISE = Aerosol Properties, PEnclosureS And InfluenceS on the Earth’s climater; NAMMA = NASA African Monsoon Multidisciplinary Analyses; ICE-T = Ice in Clouds Experiment-Tropical; COPE = COnvective Precipitation Experiment; HAIC-HIWC = High Altitude Ice Crystals - High Ice Water Content project. Instrumentation: (F)CDP = (Fast) Cloud Droplet Probe; CIP = CCD Imaging Probe; 2D-S = Two-dimensional Stereo Probe; 2D-C = Two-dimensional Cloud Probe; (F)FFSSP = (Fast) Forward Scattering Spectrometer Probe; CAS = Cloud Aerosol Spectrometer; IATF = Intercalibration time filtering; OAP = Optical Array Probe; CPI = Cloud Particle Imager; K-tips = shatter-resistant cloud probe tips (Korolev et al., 2013); SID-2H = Small Ice Detector-2 High-Performance Instrumented Airborne Platform for Environmental Research.

Overcounting can also be filtered from data with interarrival time (IAT) algorithms, which assume that shattering artifacts arrive in the probe sampling volume in much quicker succession than natural ice crystals [Field et al., 2003, 2006a; Korolev and Field, 2015]. But even in data sets that use K-tips and IAT filtering, an “enhancement” of ice crystal number beyond the INP number has persisted. A list of recent measurements, employing corrective measures and relevant to this theoretical study, is given in Table 1. A variety of microphysical processes, jointly called secondary ice production, has been proposed to explain this enhancement. For example, the Hallett-Mossop rime splintering (RS) process refers to the production of ice splinters after supercooled droplets rime onto small graupel [Hallett and Mossop, 1974]. It occurs principally for cloud temperatures between −3 and −8°C [Hallett and Mossop, 1974; Mossop, 1985], cold enough that impacting drops freeze on an ice surface but warm enough that they spread out beforehand and form fragile protuberances that can splinter off [Griggs and Choularton, 1983; Mason, 1996]. A broad droplet size distribution, with large droplets of diameter greater than 24 \(\mu\)m and small ones of less than 13 \(\mu\)m, facilitates this process [Hallett and Mossop, 1974; Mossop and Hallett, 1974], and the riming graupel must have an appreciable terminal velocity of about 0.7 m \(s^{-1}\) or greater [Hallett and Mossop, 1974; Mossop, 1985]. The process is self-sustaining, as these ice splinters can depositionally grow to a rimeable size in a matter of minutes. Hallett and Mossop (1974) have observed a maximum of 360 splinters generated per milligram of rime around −5°C.

Rime splintering parameterizations generally depend on the mass of rime, enhanced by a fixed fragment number and weighted by a temperature efficiency. Others depend instead on the number of rimed droplets that are larger than a threshold size. Bylth and Latham [1997] ran a multitrajectory cloud model with such a
parameterization and introduced nucleated ice crystals at cloud top, assuming that 50 ice splinters were produced per milligram of rime. They found that ice crystal number enhancement was most sensitive to the liquid water content. Cardwell et al. [2002] implemented a new microphysics scheme with explicit hydrometeor size distributions, an RS parameterization, and embedded convection in the Hadley Centre climate model. Their RS parameterization assumed 200 splinters produced per milligram of rime, and its influence was greatest for thermals ascending fast enough to facilitate riming. Many other studies have also examined the effect of rime splintering within three-dimensional frameworks [e.g., Ovtchinnikov and Kogan, 2000; Phillips et al., 2001; Clark et al., 2005; Phillips et al., 2007; Storelvmo et al., 2008].

In some cases, however, rime splintering alone cannot explain observed ice crystal number enhancements [e.g., Chisnell and Latham, 1976; Mossop, 1985; Rangno and Hobbs, 1994]. Laboratory experiments show that about one splinter is produced per milligram of rime [e.g., Mossop et al., 1974; Bader et al., 1974], while a value on the order of 100 is needed to reproduce some observations [Chisnell and Latham, 1976]. Temperature and droplet size distributions do not always favor rime splintering [Hobbs and Rangno, 1998], and a second peak in ice crystal number enhancement around −15°C has been observed [Leisner et al., 2014; Lloyd et al., 2015]. Finally, such large number concentrations appear quite quickly: hundreds of ice crystals per liter can form within 10 to 15 min [e.g., Hobbs and Rangno, 1990; Rangno and Hobbs, 1991, 1994], a time frame too rapid to be explained by rime splintering alone. Although estimates depend on factors like updraft and liquid water content, a calculation by Mason [1996] shows that the cloud should exist for about an hour to generate observed enhancements solely from rime splintering.

Breakup upon mechanical collision of two ice hydrometeors (breakup hereafter) has been suggested as an additional mechanism in the development of such large ice number concentrations [e.g., Hobbs and Farber, 1972; Vardiman, 1978; Takahashi et al., 1995]. In particular, Takahashi et al. [1995] performed laboratory experiments in which they saw a maximum in the number of ejected ice crystals around 257 K. Few breakup parameterizations have been developed, and they have not been incorporated into models. Vardiman [1978] calculated a fragment generation rate as the product of collision frequency and a fragment number, dependent on the change in momentum between the two colliding hydrometeors. Simulations showed that the mechanism could be important under quite limited conditions but particularly for embedded convective clouds. More recently, Yano and Phillips [2011] developed a zero-dimensional, time-lag model and identified an atmospherically relevant regime of explosive ice crystal generation by breakup based on two nondimensional parameters.

In calculating potential ice crystal number enhancements, other counteracting microphysics should be considered. In particular, ice-ice aggregation, like breakup, seems to be most efficient around 257 K. This aggregation may occur through interlocking of dendritic branches, electrostatic forces, or regelation [Hosler et al., 1957; Hobbs, 1965]. Appropriate collection kernels or sticking efficiencies have been recently developed. For example, Field et al. [2006b] used sweep-out, sum, and modified-Golovin collection kernels to reproduce ice crystal size distributions that were affected by aggregation during the Cirrus Regional Study of Tropical Anvils and Cirrus Layers—Florida Area Cirrus Experiment (CRYSTAL-FACE) campaign. They obtained the most accurate reproductions using a modified-Golovin kernel. Phillips et al. [2015] parameterized sticking efficiency of ice crystals on snow or graupel as an exponential function of collisional kinetic energy and a thermal smoothness coefficient to describe surface texture effects.

In this work, we model secondary ice production via rime splintering and breakup and the mitigating effects of ice-ice aggregation in an adiabatic parcel, employing a time delay formulation for hydrometeor growth as in Yano and Phillips [2011]. The model includes both liquid and ice hydrometeors in six classes and incorporates graupel nonsphericity. The model is used to answer two questions:

1. What maximum ice crystal number enhancement can be obtained in the parameter space when secondary ice production is active?
2. For which updraft and aerosol proxy conditions (i.e., initial hydrometeor number and size and CCN spectrum parameters), representative of certain cloud states, can a maximum ice crystal number enhancement be obtained?

Thereafter, we consider the relative contributions of rime splintering and breakup to the ice crystal number concentrations and the effect of including graupel nonsphericity or turbulence.
2. Model

2.1. Hydrometeor Number Tendencies

The model tracks six interacting hydrometeor classes for small ice crystals and droplets, small and large graupel, and medium and large droplets. The simulations are generally initialized with no hydrometeors present; once these begin to form, the sizes in each class are initialized with values in Table 2 and evolved according to growth equations. For small ice crystals, a generation function, \( G_{\text{ice}} \), is defined as the summation of primary nucleation and secondary production by breakup and rime splintering:

\[
G_{\text{ice}}(t) = c_0(T(t))H(t) + \eta_T K_T N_T N_g(t) N_{\gamma}(t) + \eta_K [K_{RS,g} N_{RS,g} N_g(t) + K_{RS,c} N_{RS,c} N_c(t)] N_{\gamma}(t).
\]

where \( c_0 \) is the nucleation rate; \( H \) is the Heaviside function; \( K \) is the collection kernels defined below in equation (9); \( \eta \) is process weightings from 0 to 100%; and \( N_T, N_g, \) and \( N_{\gamma} \) are ice crystal, small graupel, and large graupel numbers, respectively. All variables are also listed in a notation section at the end. \( c_0 \) is calculated as the product of updraft, \( u_z \); an assumed lapse rate, \( \Gamma \), of 6 K km\(^{-1}\); and the derivative of the temperature-dependent fit to INP data from DeMott et al. [2010]; \( c_0 = u_z \Gamma d/dT [a_1 \exp(a_2(T - a_3))] \). The final \( f \) factor is introduced to account for limited INP at the warmer subzero temperatures where the simulation is initiated. \( N_{RS} \) is the number of splinters generated upon breakup as a three-parameter distribution of temperature based on data of Takahashi et al. [1995]: \( N_{RS} = F(T - T_{min})^2 \exp[-(T - T_{min})/\gamma] \), where \( F \) is a leading coefficient, \( T_{min} \) is a lower temperature bound beyond which no breakup occurs, and \( \gamma \) controls the decay rate of fragment numbers at warmer subzero temperatures. Below 253 K, \( N_{RS} \) is set to 10 (see Figure 2a). \( N_{K} \) is the number of splinters produced per number of large droplets, given the large droplet radius and assuming 300 splinters per milligram of rime [Hallett and Mossop, 1974].

For the liquid phase, the droplet number generation function, \( G_{\text{drop}} \), consists simply of the product of droplet activation rate and a Heaviside function:

\[
G_{\text{drop}}(t) = u_z \Gamma d_s/dT \cdot N_{CCN} \cdot k_{CCN}^{-1} H(t).
\]

where \( u_z \) is updraft velocity, \( \Gamma \) is a lapse rate assumed to be 6 K km\(^{-1}\), \( d_s/dT \) is the temperature derivative of the supersaturation correlation, \( N_{CCN} \) and \( k_{CCN} \) are the coefficient and exponent of a Twomey power-law CCN spectrum [Twomey, 1959], and \( s_w \) is liquid supersaturation. Default values for \( u_z, N_{CCN}, \) and \( k_{CCN} \) are given in Table 2.

The number balance in each class is then the generation function at the current time as a source and the generation function at a time delay as the sink, along with aggregation, rime splintering, and coalescence losses, as in Yano and Phillips [2011]:

\[
\frac{dN_T}{dt} = G_{\text{ice}}(t) - G_{\text{ice}}(t - \tau_T) - \eta_{agg} K_{agg} N_{agg} N_{g} \tag{3}
\]

\[
\frac{dN_g}{dt} = G_{\text{ice}}(t - \tau_T) - G_{\text{ice}}(t - \tau_T - \tau_g) - \eta_{agg} K_{agg} N_{agg} N_{g} \tag{4}
\]

\[
\frac{dN_{\gamma}}{dt} = G_{\text{ice}}(t - \tau_T - \tau_g) - G_{\text{ice}}(t - \tau_T - \tau_g - \tau_{\gamma}) + \eta_{agg} K_{agg} N_{agg} N_{g} \tag{5}
\]

\[
\frac{dN_{\text{drop}}}{dt} = G_{\text{drop}}(t) - G_{\text{drop}}(t - \tau_{\text{drop}}) \tag{6}
\]

\[
\frac{dN_{\text{coal}}}{dt} = G_{\text{drop}}(t - \tau_{\text{drop}}) - G_{\text{drop}}(t - \tau_{\text{drop}} - \tau_{\text{coal}}) - \eta_{coal} K_{coal} N_{\text{coal}} N_{g} \tag{7}
\]

\[
\frac{dN_{\text{coal}}}{dt} = G_{\text{drop}}(t) - G_{\text{drop}}(t - \tau_{\text{drop}}) \tag{8}
\]

where \( \tau_T \) is the time for growth of ice crystals to small graupel, \( \tau_g \) for growth of small to large graupel, \( \tau_{\gamma} \) for large graupel to fall out, \( \tau_{\text{drop}} \) for growth of small to medium droplets, \( \tau_{\text{coal}} \) for growth of medium to large droplets, and \( \tau_{\text{coal}} \) for large droplets to fall out. The generation function at time delay represents depositional, riming, or condensational growth of hydrometeors to the next largest size class. Time delays are solved for approximately using growth equations, evolving temperature, and material properties but assuming a
Table 2. Default Simulation Values for All Parameters

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Process weightings</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\eta_{coal}$</td>
<td>100%</td>
<td></td>
</tr>
<tr>
<td>$\eta_{agg}$</td>
<td>100%</td>
<td></td>
</tr>
<tr>
<td>$\eta_{br}$</td>
<td>100%</td>
<td></td>
</tr>
<tr>
<td>$\eta_{RS}$</td>
<td>100%</td>
<td></td>
</tr>
<tr>
<td>$N_{X0}$</td>
<td>0 cm$^{-3}$</td>
<td></td>
</tr>
<tr>
<td><strong>Initial conditions</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>$r_{db}$</td>
<td>1 $\mu$m</td>
<td>Mossop [1978, 1985]</td>
</tr>
<tr>
<td>$r_{d0}$</td>
<td>12 $\mu$m</td>
<td>Mossop [1978, 1985]</td>
</tr>
<tr>
<td>$r_{R0}$</td>
<td>25 $\mu$m</td>
<td>Mossop [1978, 1985]</td>
</tr>
<tr>
<td>$r_{i0}$</td>
<td>5 $\mu$m</td>
<td>Zhang et al. [2014]</td>
</tr>
<tr>
<td>$a_{g0}$</td>
<td>50 $\mu$m</td>
<td>Reinking [1975]</td>
</tr>
<tr>
<td>$a_{GO}$</td>
<td>200 $\mu$m</td>
<td>Reinking [1975]</td>
</tr>
<tr>
<td>$T_0$</td>
<td>272 K</td>
<td></td>
</tr>
<tr>
<td>$p_0$</td>
<td>680 hPa</td>
<td></td>
</tr>
<tr>
<td>$s_{w,0}$</td>
<td>$10^{-6}$%</td>
<td></td>
</tr>
<tr>
<td><strong>Time scales</strong></td>
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<td></td>
</tr>
<tr>
<td>$\tau_d$</td>
<td>5 min</td>
<td>Approximate solution of equation (16)</td>
</tr>
<tr>
<td>$\tau_r$</td>
<td>15 min</td>
<td>Approximate solution of equation (17)</td>
</tr>
<tr>
<td>$\tau_R$</td>
<td>25 min</td>
<td>Approximate solution of equation (18)</td>
</tr>
<tr>
<td>$\tau_i$</td>
<td>7.5 min</td>
<td>Approximate solution of equation (19)</td>
</tr>
<tr>
<td>$\tau_g$</td>
<td>20 min</td>
<td>Approximate solution of equation (20)</td>
</tr>
<tr>
<td>$\tau_G$</td>
<td>17.5 min</td>
<td>Approximate solution of equation (21)</td>
</tr>
<tr>
<td><strong>Time step</strong></td>
<td>$\Delta t$</td>
<td>6 s</td>
</tr>
<tr>
<td><strong>Droplet spectrum</strong></td>
<td>$k_{CCN}$</td>
<td>0.308</td>
</tr>
<tr>
<td></td>
<td>$N_{CCN}$</td>
<td>100 cm$^{-3}$</td>
</tr>
<tr>
<td><strong>Updraft</strong></td>
<td>$u_z$</td>
<td>2 m s$^{-1}$</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Terminal velocity</strong></td>
<td>$B_0$</td>
<td>0.6</td>
</tr>
<tr>
<td></td>
<td>$\delta$</td>
<td>5.83</td>
</tr>
<tr>
<td><strong>Nucleation reduction</strong></td>
<td>$f$</td>
<td>10$^2$</td>
</tr>
</tbody>
</table>

Constant supersaturation and a constant radius for the riming droplets. This set of hydrometeor number tendencies is solved with an explicit Runge-Kutta (2,3) pair extended for delay-differential equations [Bogacki and Shampine, 1989].

In the generation functions and number balances above, gravitational collection kernels are used to describe all processes:

\[
K_{br}(t) = \pi (\xi_G a_G(t)^2 + \xi_g a_g(t)^2)(v_{t,G} - v_t) \tag{9}
\]

\[
K_{RS,G}(t) = \pi (r_{G}(t)^2 + \xi_g a_g(t)^2)(v_{t,G} - v_{t,R})RS_T
\]

\[
K_{RS,C}(t) = \pi (r_{C}(t)^2 + \xi_G a_G(t)^2)(v_{t,G} - v_{t,R})RS_T
\]

\[
K_{agg}(t) = \pi (\xi_g a_g(t)^2 + r_i^2)(v_{t,G} - v_t)
\]

\[
K_{coal}(t) = \pi (r_i^2 + r_{i0}^2)w_{t,r}.
\]

where $\xi$ is the ratio of actual cross section to that of a circumscribed circle as in Jensen and Harrington [2015], $a$ is a spheroidal major axis, $r$ is a radius, and $v_i$ is the hydrometeor terminal velocity as in Mitchell and Heymsfield [2005]. Within the coalescence kernel, we assume a coalescence efficiency of 1 and that the terminal velocity of small droplets is negligible relative to that of medium droplets. $RS_T$ is a temperature weighting for the rime splintering process, equal to 50% from 269 to 271 K and from 265 to 267 K, 100% from 267 to 269 K, and 5% from 243 to 265 K, as in Ferrier [1994].
2.1.1. Microphysical Assumptions

Figure 1 shows a schematic of the model microphysics. In the calculation of the hydrometeor number tendencies, the following simplifying assumptions are made:

1. **Breakup and rime splintering generate ice crystals in the smallest class.** All fragments from breakup are assumed to be small ice crystals because there are no laboratory measurements of the fragment size distribution. For rime splintering, experimental evidence indicates that fragments have a size on the order of 10 μm [e.g., Hallett and Mossop, 1974] (their Figure 1b) and [Heymsfield and Willis, 2014] (their Figure 2c), or even smaller [e.g., Bader et al., 1974].

2. **Only small and large graupel undergo breakup upon collision.** We assume that riming protuberances and sufficient momentum are required for a fragment-generating collision. Small ice crystals do not have elaborate enough geometry or large enough terminal velocity to shatter upon collision. Precedent for these assumptions comes from Vardiman [1978]: only for large degrees of riming and relative velocity between hydrometeors did fragments form upon breakup during experiments. The results of Takahashi et al. [1995] are also relevant only for collisions of larger ice hydrometeors: the apparatus diameter used to simulate breakup was 1.8 cm, orders of magnitude larger than any ice crystal.

3. **Rime splintering occurs on both small and large graupel.** The initial major axis of small graupel, $a_{G0}$, is 50 μm and that of large graupel, $a_{G0}$, is 200 μm. Ono [1968] has shown that riming is rare on columnar ice hydrometeors with a major axis less than 50 μm or on planar ones with a major axis of less than 150 to 200 μm.
4. Aggregation generates graupel in the largest class. Laboratory and field studies have shown that aggregates have a maximum dimension on the order of a few hundred microns. For example, the pristine ice hydrometeors in the cloud chamber study of Connolly et al. [2012] have a maximum dimension of about 100 μm (their Figure 6), while aggregates have a maximum dimension of a few hundred microns (their Figure 7). In situ CPI imagery of Field and Heymsfield [2003] shows almost no aggregation for hydrometeors collected with diameter 100 μm or less but large numbers of aggregates for those of a few 100 μm diameter.

5. Aggregation occurs between ice crystals and smaller graupel. Coalescence occurs between small and medium droplets. Because the initial diameter of the ice crystals, 2μ, is on the order of magnitude smaller than the initial major axis of small graupel, we assume that collisions between two ice crystals will be relatively less efficient than collisions between an ice crystal and a small graupel particle, even when N is about an order of magnitude larger than N. A larger relative terminal velocity between ice crystals and small graupel will also enhance this collection kernel relative to that between two ice crystals. Aggregation between ice crystals and large graupel is also assumed to be negligible. Hosler and Hallgren [1960] performed aggregation measurements for ice crystals of diameter 7 to 13 μm and noted that additional contact area beyond a critical overlap does not increase sticking efficiency. And while aggregation may be possible over a large range of sizes, collisions of similarly sized hydrometeors are most important to initial aggregate formation [Connolly et al., 2012]. Finally, the coalescence of small droplets with one another is not included, given collection efficiencies around 5% or less according to Klett and Davis [1973].

2.2. Moist Thermodynamic Tendencies

The hydrometeor number tendencies, equations (3) to (8), are coupled to the following moist thermodynamic equations to account for the changing system supersaturation:

\[
\frac{dp}{dt} = \frac{g\mu_x}{R_a T} \sigma_p \left( \frac{\Delta H_r}{(1 + q_v) c_p} \right) \frac{dq_w}{dt} + \frac{\Delta H_q}{(1 + q_v) c_p} \frac{dq_i}{dt}
\]

(10)

\[
\frac{dT}{dt} = -\frac{g\mu_x}{c_p} \left( \frac{\Delta H_r}{(1 + q_v) c_p} \right) \frac{dq_w}{dt} + \frac{\Delta H_q}{(1 + q_v) c_p} \frac{dq_i}{dt}
\]

(11)

\[
\frac{ds_w}{dt} = (1 + s_w) \left[ \frac{g\Delta H_r}{c_p R_a T^2} \left( \frac{\rho_w}{\rho_a} \right) \frac{dr}{dt} + \left( \frac{1}{q_v} + \frac{\Delta H_q^2}{c_p R_a T^2} \right) \frac{dq_w}{dt} - \left( \frac{1}{q_v} + \frac{\Delta H_q \Delta H_r}{c_p R_a T^2} \right) \frac{dq_i}{dt} \right]
\]

(12)

(21)
The hydrostatic approximation is made for pressure evolution in equation (10), and equation (11) is the adiabatic energy conservation equation. Equation (12) is the supersaturation balance, derived for a mixed-phase parcel in Korolev and Mazin [2003] (their Appendix A). Equations (13) to (15) are liquid, ice, and vapor mixing ratio evolutions, and equations (16) to (21) are the growth equation for droplet and crystal radii and graupel axes. Calculation of thermodynamic values like heat of sublimation and vaporization is detailed next.

Recently nucleated ice crystals are assumed to be spherical with a radius, \( r_i \); bulk ice density, \( \rho_i \); and unit capacitance. Graupel is assumed to be spheroidal with a horizontal axis, \( a_G \); deposition density, \( \rho_{\Delta} \); and capacitance according to McDonald [1963]. The model uses the mass distribution hypothesis of Chen and Lamb [1994] to describe the relative depositional growth of spheroidal axes. A temperature-dependent polynomial, \( \Gamma_{IG}(T) \), is fit to the inherent growth factor data in Figure 2b. In general, a single updraft velocity is prescribed, but one simulation performs a Monte Carlo sampling of a Gaussian updraft distribution to mimic the effects of turbulent velocity fluctuations. This set of thermodynamic tendencies is solved with a modified Rosenbrock formula of order 2 [Rosenbrock, 1963].

### 2.3. Thermodynamic Correlations and Parameters

For heat of sublimation, \( \Delta H_s \), a temperature-dependent correlation of Rogers and Yau [1979] is used. The correlation for the latent heat of vaporization of supercooled liquid is used from Murphy and Koop [2005].

\[
\Delta H_s \approx 10^3 \left[ 2834.1 - 0.29(T - 273.15) - 0.004(T - 273.15) \right] \quad (22)
\]

\[
\Delta H_v \approx 56579 - 42.2127T + \exp\left[0.1149(281.6 - T)\right] \quad (23)
\]

Capacitances of small and large graupel are calculated via an electrostatic analogy, according to McDonald [1963]:

\[
C = \begin{cases} \frac{\pi a^2}{2 \ln(\psi)} & \Gamma_{IG} > 1 \text{ (oblate hydrometeor)} \\ \frac{\pi a \Omega}{\ln(\psi^{-1} \exp\psi)} & \Gamma_{IG} < 1 \text{ (prolate hydrometeor)}, \end{cases} \quad (24)
\]

where

\[
\Omega = \sqrt{1 - a^2(\Gamma_{IG} - 1)} \quad \text{and} \\
\psi = \sqrt{a^2(\Gamma_{IG} - 1)}.
\]

The temperature-dependent correlation of thermal conductivity and saturation vapor pressure over water and ice are used from Kannuluik and Carman [1951] and Murphy and Koop [2005], respectively:

\[
k_w = 0.024 \left[ 1 + 0.00317(T - 273.15) - 2.1 \times 10^6(T - 273.15)^2 \right] \quad (25)
\]
Viscosity comes from the Sutherland model, and diffusivity from the temperature- and pressure-dependent correlation given in Seinfeld and Pandis [2006]:

\[
\mu = 18.27 \times 10^{-6} \frac{411.15}{T + 120} \left( \frac{T}{291.15} \right)^{3/2} T^{1.94}
\]

Hydrometeor terminal velocities are calculated according to Mitchell and Heymsfield [2005]:

\[
Re = \frac{\rho_w L_c v}{\mu} = \frac{\delta^2}{4} \left[ 1 + 4X^{1/2} + \frac{\rho_w}{\rho_i} \right] - 1
\]

where the characteristic length \(L_c\) in the Reynolds number is taken to be the hydrometeor radius or axis; \(\delta\) and \(B_0\) are parameters accounting approximately for surface roughness; and \(X\) is the Davies number. Finally, the temperature dependence of densities and heat capacities is neglected: \(\rho_w\) as 1.395 kg m\(^{-3}\); \(\rho_i\) as 919 kg m\(^{-3}\); \(c_p\) as 1850 J kg\(^{-1}\) K\(^{-1}\); and \(\mu\) as 998 kg m\(^{-1}\) s\(^{-1}\).

The inherent growth factor and fragment generation function upon breakup versus temperature are shown in Figure 2.

3. Simulations

Twelve simulations are run, as detailed in Tables 3 and 4. The first set — Cases 1, 2, and 3 — uses process weightings before the breakup and rime splintering tendencies in \(G_{\text{tend}}\) and before the aggregation and coalescence tendencies in equations (3) to (8). These process weightings are denoted, respectively, \(\eta_{\text{br}}, \eta_{\text{br}}, \eta_{\text{agg}},\) and \(\eta_{\text{coal}}\). The processes are turned off (\(\eta_{\text{br}} = 0\%) or set at moderate (\(\eta_{\text{br}} = 50\%) or high (\(\eta_{\text{br}} = 100\%) values to estimate a range of values for \(N_{\text{tend}}\), the total number of ice phase hydrometeors. The weightings act as a mathematical tool to investigate how hydrometeor numbers evolve when conditions favor different processes.

In Case 1, secondary production via rime splintering and breakup is turned off, while Case 3 promotes secondary production with high weightings. Case 2 is intermediate with moderate weightings for all processes. These first three cases are run with the default values listed in Table 2. They are initiated for an unsaturated parcel just below the freezing temperature at a mixed-phase pressure level, ascending at a modest convective updraft.

Cases 1 to 3 are then rerun, assuming sphericity for all ice hydrometeors, and denoted Case 1S, 2S, and 3S, respectively. The description of nonsphericity is removed by replacing deposition density with bulk ice density and graupel major axes by radii. The inherent growth factor and graupel capacitances are set to unity, and the graupel terminal velocities and collection kernels are calculated as for the other spherical hydrometeors. All other parameters and weightings remain the same as in Cases 1, 2, and 3. Another variant of these cases is run as Cases 2T and 3T, in which a small ensemble of simulations is run with updrafts set at each time step by Monte-Carlo sampling from a normal distribution.

Cases 4A and 4B investigate the maximum contribution of breakup to secondary production. Case 4A adjusts process weightings and parameters to favor breakup: rime splintering and coalescence are turned off, the updraft is decreased, the nucleation rate is increased, and the simulation is run with some small and large graupel initially. Case 4B adjusts growth times to favor breakup: rime splintering is allowed to occur again with low weighting, and the initial temperature is lowered to 265 K, elongating the characteristic times for the liquid phase. Cases 4AT and 4BT use the same setup as Cases 4A and 4B but with an ensemble of Monte-Carlo sampled updrafts as in Cases 2T and 3T.

Finally, three points are defined in the parameter space with conditions similar to particular cloud states. A continental convective (CC) case has a stronger updraft and steeper CCN spectrum [Pruppacher and Klett, 1997]. Continental aerosol loading can be quite high, so the initial droplet radius is smaller than the default value, and the characteristic times for all hydrometeors are shorter because a stronger updraft yields faster
Table 3. Simulations With Process Weightings or Parameters Adjusted From the Default Values in Table 2

<table>
<thead>
<tr>
<th>Weightings</th>
<th>Sphericity</th>
<th>Breakup contribution</th>
<th>Updraft</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case 1</td>
<td>Case 1S</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Suppress secondary production</td>
<td>Suppress secondary production</td>
<td>Assume sphericity</td>
<td></td>
</tr>
<tr>
<td>$\eta_{RS}, \eta_{br} = 0%$</td>
<td>$\rho_{D} = \rho_{i}, \Gamma_{IG}(T) = 1$</td>
<td>$a_{g,G} = r_{g,G}, c_{g,G} = 1$</td>
<td></td>
</tr>
<tr>
<td>Case 2</td>
<td>Case 2S</td>
<td></td>
<td></td>
</tr>
<tr>
<td>All processes equal</td>
<td>All processes equal</td>
<td>Assume sphericity</td>
<td></td>
</tr>
<tr>
<td>$\eta_{cool}, \eta_{agg}, \eta_{RS}, \eta_{br} = 50%$</td>
<td>$\rho_{D} = \rho_{i}, \Gamma_{IG}(T) = 1$</td>
<td>$a_{g,G} = r_{g,G}, c_{g,G} = 1$</td>
<td></td>
</tr>
<tr>
<td>Case 3</td>
<td>Case 3S</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Promote secondary production</td>
<td>Promote secondary production</td>
<td>Assume sphericity</td>
<td></td>
</tr>
<tr>
<td>$\eta_{agg}, \eta_{cool} = 0%$</td>
<td>$\rho_{D} = \rho_{i}, \Gamma_{IG}(T) = 1$</td>
<td>$a_{g,G} = r_{g,G}, c_{g,G} = 1$</td>
<td></td>
</tr>
<tr>
<td>Case 4A</td>
<td>Case 4AT</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Promote breakup</td>
<td>Adjust process weightings</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\eta_{cool}, \eta_{RS} = 0%$</td>
<td>$u_{2} = \mathcal{N}(\mu = 0.75, \sigma = 0.25 \text{ m s}^{-1})$</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Case 4B</td>
<td>Case 4BT</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Promote breakup</td>
<td>Adjust characteristic times</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\eta_{cool} = 0%, \eta_{RS} = 1%$</td>
<td>$u_{2} = \mathcal{N}(\mu = 0.75, \sigma = 0.25 \text{ m s}^{-1})$</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

*All $\tau$ values are given in minutes. $\mathcal{N}$ indicates a normal distribution.

Table 4. Parameters and Characteristic Times That Vary From Default Values in Table 2 for Simulations in Section 4.5

<table>
<thead>
<tr>
<th>Cloud states</th>
<th>Case CC</th>
<th>Case MC</th>
<th>Case AS</th>
</tr>
</thead>
<tbody>
<tr>
<td>Continental convective</td>
<td>$r_{d0} = 0.5 \mu m, u_{2} = 3 \text{ m s}^{-1}$</td>
<td>$r_{d0} = 7.5 \mu m, u_{2} = 2 \text{ m s}^{-1}$</td>
<td>$r_{d0} = 5 \mu m, r_{d0} = 10 \mu m, u_{2} = 1 \text{ m s}^{-1}$</td>
</tr>
<tr>
<td>$N_{CCN} = 300 \text{ cm}^{-3}, k_{CCN} = 0.9$</td>
<td>$N_{CCN} = 125 \text{ cm}^{-3}, k_{CCN} = 0.3$</td>
<td>$N_{CCN} = 10 \text{ cm}^{-3}, k_{CCN} = 0.5$</td>
<td></td>
</tr>
<tr>
<td>$(\tau_{d}, \tau_{r}, \tau_{g}) = (1.5, 4.5, 9)$</td>
<td>$(\tau_{d}, \tau_{r}, \tau_{g}) = (2, 6, 12)$</td>
<td>$(\tau_{d}, \tau_{r}, \tau_{g}) = (4.5, 18, 25)$</td>
<td></td>
</tr>
<tr>
<td>$(\tau_{i}, \tau_{g}, \tau_{0}) = (7.5, 12.5, 12.5)$</td>
<td>$(\tau_{i}, \tau_{g}, \tau_{0}) = (7.5, 12.5, 12.5)$</td>
<td>$(\tau_{i}, \tau_{g}, \tau_{0}) = (7.5, 22.5, 12.5)$</td>
<td></td>
</tr>
</tbody>
</table>

*All $\tau$ values are given in minutes.
growth rates. For an Arctic stratocumulus (AS) case, updraft is lowered and a more gradual CCN spectrum is chosen. Aerosol loading is generally lower, so the run is initialized with larger droplet and ice crystal radii. Characteristic times are adjusted to account for the different initial sizes, lower updraft, and colder initial temperature. Finally, a maritime convective (MC) case lies between the CC and AS cases, as Case 2 did above: updraft, CCN spectrum, and initial hydrometeor size are all intermediate. Characteristic times are adjusted for the updraft and initial hydrometeor sizes.

4. Results

4.1. Process Weightings

The ice hydrometeor number evolution for Cases 1, 2, and 3 is shown below in Figure 3. The simulations run until the parcel becomes subsaturated with respect to liquid, a duration that is longest for Case 1 when secondary ice production is turned off. Only when a large number of small ice crystals is formed, is depositional growth rapid enough to deplete supersaturation. When the secondary production is turned on, formation of large hydrometeors also plays a role in the simulation duration: once large droplets or graupel form, they feed into the rime splintering or breakup tendencies, respectively, and form the supersaturation-consuming ice crystals. When aggregation is suppressed in Case 3, small graupel is not efficiently consumed, and their growth speeds up the subsaturation in Case 3 relative to Case 2. The formation of large droplets and graupel around 20 and 28 min, respectively, is also reflected in the timing of the sudden enhancements in \( N_{\text{ice}} \).

Of greater interest is the total ice hydrometeor number, \( N_{\text{ice}} \), before subsaturation. The fewest ice hydrometeors form in Case 1: \( N_{\text{ice}} \) reaches a maximum of only 0.113 \( L^{-1} \) over 56.9 min. In Case 2, the maximum \( N_{\text{ice}} \) is 2 orders of magnitude higher at 31.9 \( L^{-1} \) over about half that time. If the Case 1 and 2 \( N_{\text{ice}} \) values at the same time point around 30 min are compared, the ice crystal number enhancement is instead 4 orders of magnitude (0.0062 \( L^{-1} \) relative to 31.9 \( L^{-1} \)). Finally, \( N_{\text{ice}} \) reaches a maximum of 24.6 \( L^{-1} \) in Case 3. Although secondary ice production weightings are high, these tendencies require large droplets or graupel. When droplet coalescence and aggregation are suppressed, these large hydrometeors are only slowly formed by growth from smaller hydrometeor classes.

Along with the static metrics of simulation duration and maximum \( N_{\text{ice}} \), the overall simulation structure indicates which processes are most influential. Droplets activate rapidly early on up to an \( N_g \) of a couple hundred per cm\(^3\) for all cases, until supersaturation peaks and begins to drop off. After about 5 min, the small droplets have had enough time to grow by condensation to medium droplets. Large droplets form thereafter, most quickly in Case 1 when droplet coalescence is promoted.

In the ice phase, new ice crystals nucleate throughout the simulation, and small graupel begins to form after 7.5 min. Formation of small graupel stunts the increase in \( N_i \); their growth consumes supersaturation and the aggregation sink activates. Formation of large graupel, on the other hand, boosts the increase in \( N_i \), because generation from rime splintering and breakup initiates. Part of the reason that \( N_{\text{ice, max}} \) is lower in Case 3 than in Case 2 is that the large graupel number stays very low in the former. In Case 2, \( N_g \) can reach higher values due to active aggregation. A final point is that small graupel act as “limiting hydrometeor class,” when ice crystal number enhancement is largest in Case 2. Small graupel feed the breakup tendency both directly through the collision itself and indirectly through aggregation to generate large graupel. Small graupel also rime-splinter, and these numerous sinks mean they are quickly consumed and can limit secondary production.

Referring to the measurements in Table 1, these values and evolution are comparable to those from Crawford et al. [2012], Heymsfield and Willis [2014], and Lasher-Trapp et al. [2016] with modest updraft, warmer in-cloud temperatures, and \((N_{\text{high}}, N_{\text{max}}) \sim O (0.01 \text{ or lower} \text{ } L^{-1}, \text{ } 100 \text{ } L^{-1})\). These studies explained the observed enhancement solely with rime splintering, and indeed for our simulations, the contribution of rime splintering is greater than 90% for both Cases 2 and 3. Given the large cloud depth in Lasher-Trapp et al. [2016], these cases replicate some aspects of the “microphysical progression” in that study as well: \( N_g \) greater than hundreds per liter that feeds into coalescence, \( N_g \) on the order of 0.1 \( L^{-1} \) around \(-5^\circ C\), and highest \( N_i \) and continued presence of \( N_g \) as the parcel decays.

4.2. Impact of Ice Hydrometeor Sphericity

The next set of simulations is run to investigate the contribution of graupel nonsphericity to ice crystal number enhancement. The parcel model is particularly insightful in this case because larger-scale weather and climate models cannot generally afford the computational expense of evaluating temperature-dependent inherent growth factors, deposition densities, and capacitances. Nonsphericity will affect the ice hydrometeor
The ice hydrometeor number evolution for these cases is shown in Figure 4. All simulations in this set remain supersaturated longer than the corresponding ones in the first set. Especially in Cases 2 and 3, the sphericity assumption extends the simulation duration by a factor of 2.1 or half an hour. Evolution of the liquid phase numbers (not shown) is almost identical to that in the first set; the only modification is to the rate of consumption of $N_g$ due to changes in the rime splintering collection kernel. A lower number of ice crystals and slower graupel growth rate allow the parcel to remain supersaturated for longer, elongating the simulation duration.
Despite their longer durations, Cases 15, 25, and 35 produce much lower values of $N_{\text{ice,max}}$ than the first three cases. $N_{\text{ice,max}}$ decreases by a factor of 3 between Case 2 and Case 25 from 31.9 $\text{L}^{-1}$ down to 10.8 $\text{L}^{-1}$. Between Cases 3 and 35, it decreases by a factor of 2.6 from 24.6 $\text{L}^{-1}$ down to 9.4 $\text{L}^{-1}$. These decreases can be attributed to smaller collisional cross sections in the rime splintering and breakup collection kernels. The graupel develop less extreme dimensions when we assume sphericity. Nonsphericity causes preferential deposition in regions of small radius of curvature, according to Fick’s law of diffusion, which in turn generates more extreme dimensions [Chen and Lamb, 1994; Sullivan and Harrington, 2011]. As a sidenote, the trend is reversed between Cases 1 and 15: $N_{\text{ice,max}}$ is slightly larger in Case 15 because a smaller collisional cross section lowers the aggregation sink rather than any secondary production sources.

We can also estimate that the difference in graupel dimension between the spherical and nonspherical case will be more pronounced, just as secondary production begins: simulations are initiated from 272 K with an updraft of 2 m s$^{-1}$, and assuming an adiabatic lapse rate of 6 K km$^{-1}$, the parcel temperature reaches 258 K when large graupel begin to form around 20 min. At this temperature, $\Gamma_{\text{ic}}$ is particularly low because more dendritic shapes are favored. And at this time in Case 2, the most dramatic increase in $N_i$ begins.

Hydrometeor terminal velocity should also be considered, along with collisional cross section. In cases with sphericity, the less extreme dimension and higher density increase hydrometeor terminal velocity. Faster terminal velocities increase the rime splintering and breakup collection kernels. This effect, however, depends linearly on dimension and that of collisional section is second order, so that the latter dominates.

Returning to the observations in Table 1, Heymsfield and Willis [2014] note that $N_i$ is highest, on the order of $100 \text{L}^{-1}$, when the concentrations of needle and columnar ice from the CIP are highest. Indeed, they specifically categorize secondary ice particles as those with these highly nonspherical geometries. Sample images from the 2D-S probe in Crosier et al. [2014] show dendritic and capped column geometries (their Figure 9). They understand the latter to be the product of rime splintering that is then transported to higher altitudes and colder temperatures that favor plate-like geometry. And CIP-100 images from Taylor et al. [2016] show the most nonspherical geometries in mixed-phase regions (Regions II and V in their Figure 5).

We can also consider the effect of sphericity on simulated ice production rates relative to those in the studies from Table 1. For example, if we assume the ice production rate from Taylor et al. [2016], calculated according to Harris-Hobbs and Cooper [1987], of 0.14 $\text{L}^{-1} \text{s}^{-1}$, the parcel should produce $N_{\text{ice}}$ of about 120 $\text{L}^{-1}$ over 15 min. This generation is faster by a factor of 4 than that occurring from 20 to 32 min of Cases 2 and 3 but a factor of 35 faster than the generation from 20 to 65 min in Cases 25 and 35. Or if we use the ice production rate of 50 $\text{s}^{-1}$ at 1.8 m s$^{-1}$ and about $-4^\circ$ C from Mossop [1976] and presented in Heymsfield and Willis [2014], the parcel should produce an $N_{\text{ice}}$ of about 45 $\text{L}^{-1}$ over 15 min. Cases 2 and 3 produce about this concentration in this time frame, while Cases 25 and 35 require 3 times as long to produce 3 times less ice.

To bring simulated and measured production rates into even better agreement, the description of ice hydrometeor nonsphericity could be made more sophisticated. Ventilation effects on depositional growth of falling hydrometeors could be incorporated [Chen and Lamb, 1994], accelerating growth rates and yielding more extreme graupel geometries (see Discussion below). Ice hydrometeors also do not necessarily maintain a fixed orientation as they fall through turbulent air [Zikmund and Vali, 1972; Cho et al., 1981]. Including hydrometeor oscillations or tumbling would tend to decrease the effective hydrometeor dimension and collection kernels.

### 4.3. Breakup Contribution

The simulated $N_{\text{ice}}$ values can be broken down into process contributions to indicate which microphysical processes $X$ are contributing most to the generation of new ice crystals. A process contribution, $p_X$, is defined as the percentage of small ice crystals generated by process $X$:

$$p_X = \frac{\left(\frac{dN_i}{dt}\right)_X}{\sum_{i \in X} \left(\frac{dN_i}{dt}\right)}.$$  

Nucleation, rime splintering, and breakup tendencies go into this calculation; as a sink, aggregation is not included. $p_{br}$ for Cases 1, 15, 2, 25, 3, and 35 is around 5% or less. This low contribution of breakup is due in part to the consideration of water saturated conditions only. To investigate whether $p_{br}$ could be more significant, Cases 4A and 4B are run with conditions favorable to breakup. In Case 4A, no rime splintering or coalescence
occurs and the updraft is smaller. The parcel is initiated with $N_g$ and $Ng$ of 10 m$^{-3}$ each, and a higher nucleation rate is assumed. These conditions could represent the larger stratiform deck adjacent to or surrounding convective cores. These regions are generally characterized by modest ascent and cold temperatures. Lofting or formation of large droplets is unlikely, but graupel may be present due to advection or detrainment from the core. Taylor et al. [2016] note that both the preexisting ice and millimeter-sized graupel requisite for breakup were present in their observations of cumulus clouds off the South West Peninsula. They measured $N_{ice}$ on the order of hundreds per liter at temperatures below $-8^\circ$ C and recorded the highest $N_{ice}$ of 350 L$^{-1}$ in a mature, mostly quiescent stratiform region (Table 1).

The ice hydrometeor numbers are shown for all classes in Figure 5a. A limited number of small hydrometeors and modest updraft lead to much longer-lived liquid supersaturation than in the first six cases: the simulation lasts 113.9 min, 50% longer than any of the previous cases. $N_{ice,max}$ reaches 0.93 L$^{-1}$ in this time, about an order of magnitude less than Case 2, but still an eightfold ice crystal number enhancement from Case 1. The slower growth in $N_i$ can be explained via an order of magnitude analysis: $N_{g} \sim O(10^2$ cm$^{-3}$), $N_{g} \sim O(10^{-5}$ cm$^{-3}$), $N_{rs} \sim O(10^2)$, and $N_{br} \sim O(10^{-2})$, so that the breakup tendency is $10^2$ to $10^3$ times weaker than the rime-spintering one. If even larger initial graupel numbers are feasible, $N_{g0} \sim N_{g0} \sim O(10^{-3}$ cm$^{-3}$ or 1 L$^{-1}$), then the rime splintering and breakup tendencies approach the same order, and a larger ice crystal number enhancement may be generated by breakup over a shorter time frame.

We also look at the contribution of nucleation versus breakup for this case in Figure 5c. $p_{nuc}$ is reduced from 100% initially by the presence of preexisting graupel. Both $N_{br}$ and $c_0$ increase as the parcel temperature gets colder, but the $N_{br}$ temperature dependence is stronger and $p_{br}$ steadily grows at the expense of $p_{nuc}$ until it reaches a maximum of 38.2% after 27.9 min. Thereafter, $N_{br}$ decreases with decreasing temperature, while $c_0$ continues to increase, and $p_{nuc}$ dominates the remaining ice production.

In Case 4B, we adjust characteristic times rather than microphysical properties to favor breakup. The simulation is initiated from a colder temperature of 265 K, and the liquid phase growth times are elongated due to decreased water vapor diffusivity and slower condensational growth. The ice phase growth times are held constant, assuming that growth by riming dominates. In line with this assumption, rime splintering is allowed...
to occur again with a low weighting. These conditions represent a cloud close to glaciation with minimal supercooled liquid fraction.

The ice phase number evolution is shown in Figure 5b. As in Case 4A, an initial tradeoff occurs between increasing \( N_{\text{br}} \) and \( c_0 \), but the \( N_{\text{br}} \) temperature dependence dominates and \( p_{\text{br}} \) reaches a maximum of 54.9% after 25.5 min. Subsaturation occurs after 91.6 min with \( N_{\text{ice, max}} \) having reached 0.997 L\(^{-1}\). With a reduced weighting for rime splintering, \( p_{\text{RS}} \) does not rise much about 5%.

From Cases 4A and 4B, the promotion of purely ice-phase microphysics (i.e., aggregation and breakup) relative to mixed- or liquid-phase microphysics (i.e., rime splintering or droplet coalescence) can increase \( p_{\text{br}} \) to significant values. But the more influential factor is when large hydrometeors in either phase first form. If large droplets form well in advance of large graupel, rime splintering will be favored as in Cases 1, 2, and 3. If large graupel are able to form simultaneously or even earlier than large droplets, breakup contributes to secondary production. Early formation of large ice hydrometeors is feasible for colder in-cloud temperatures at which condensational growth rates, but not necessarily riming rates, have slowed. Graupel could also form outside of the cloud and be advected or detrained in. Independent of how graupel reaches the parcel, a modest nucleation rate is favorable so that large \( N_i \) does not deplete supersaturation too quickly: the time to produce a meaningful ice crystal number enhancement from breakup is relatively long.

As a final point, the necessary impact for two hydrometeors to shatter upon collision is still highly uncertain. \( N_{\text{br}} \) is probably a function of relative terminal velocity, along with temperature. Understanding this dependence will be important in order to identify the actual cloud states for which \( p_{\text{br}} \) may be important.

4.4. Updraft Velocity

Having considered how microphysical parameters affect secondary production, a set of simulations is run to understand the effect of dynamical parameters. The constant updraft from the first eight simulations is replaced by a Gaussian updraft distribution with mean, as in the corresponding nonturbulent case, and standard deviation as in Table 3. At each time point, a new updraft is Monte-Carlo sampled from the distribution. A 10-run ensemble is done for each of these cases to investigate variability in \( N_{\text{ice, max}} \) and \( t_{\text{dur}} \).

Figure 6 shows \( N_{\text{ice}} \) and \( p_{\text{br}} \) for this ensemble of Case 2, 3, 4A, and 4B reruns, denoted 2T, 3T, 4AT, and 4BT, respectively. The inclusion of the updraft distribution in Case 2T and 3T does not drastically affect the \( N_{\text{ice}} \) structure; primary nucleation dominates initially, followed by an exponential growth in \( N_{\text{ice}} \) after large droplet formation around 20 min. In Case 4AT and 4BT, both nucleation and breakup contribute initially with average updraft strength modulating the slope of the increase in \( N_{\text{ice}} \) and the simulation duration significantly. This modulation is due to the temperature dependences of \( c_0 \) and \( N_{\text{br}} \), noted above. When updraft is lower, it limits the nucleation rate or fragment number upon breakup through temperature. Fewer ice crystals yield fewer graupel, less secondary production, and later water subsaturation. We call this cycle a “dynamically limited case”.

The structure and duration not only vary between the simulations with fixed and variable updraft but also between the ensemble runs themselves. Figure 6 displays the standard deviation in maximum ice hydrometeor number, \( \sigma_{N_{\text{ice}}} \), for each simulation: 5.59, 4.995, 0.297, and 0.338 L\(^{-1}\) for Case 2T, 3T, 4AT, and 4BT, respectively. If \( \sigma_{N_{\text{ice}}} \) is normalized by the \( N_{\text{ice, max}} \) value, however, these values become 0.17, 0.20, 0.32, and 0.34. The cases favoring breakup have larger normalized variability than the cases with different weightings due to the dynamic modulation described above.

We also consider the maximum contribution of breakup for Case 4AT and 4BT in Figures 6e and 6f. Runs with the highest \( N_{\text{ice, max}} \) and shortest duration have lower \( p_{\text{br}} \) relative to the ensemble mean. Here again, there are “dynamically limited cases”: lower updraft decreases nucleation rate and increases and delays \( p_{\text{br, max}} \). Slower ascent increases and delays \( p_{\text{br, max}} \) because fewer small crystals are nucleated and the parcel attains colder temperatures where \( N_{\text{br}} \) is large before \( p_{\text{true}} \) outweighs \( p_{\text{br}} \).

More generally, for models which include droplet evaporation, a mixed-phase cloud state may be sustained by a fluctuating updraft under which droplets evaporate and reactivate periodically [Korolev and Isaac, 2003]. Theoretically, this “steady state” is not attainable for a uniformly ascending parcel, and glaciation will occur more quickly. Case 4AT and 4BT exhibit similar behavior, as they can be lengthened by 20% relative to their uniformly ascending equivalents. These simulations are halted with water subsaturation not glaciation, but water subsaturation is a prerequisite for the Bergeron process and eventual glaciation [e.g., Sednev et al., 2009]. Looking back at Table 1, Heymsfield and Willis [2014] found that the highest \( N_{\text{ice}} \) was formed at modest updrafts
Figure 6. Total ice hydrometeor number, \(N_{\text{ice}}\), evolution for (a) Case 2T, (b) Case 3T, (c) Case 4AT, and (d) Case 4BT. For each case, an ensemble of 10 runs is done with each run shown in a different color. (e and f) The contribution of breakup to \(N_{\text{ice}}\) for Case 4AT and 4BT.

for which hydrometeors could “loiter within the secondary production zone.” This behavior is replicated in our simulations; however, the ensembles in Figures 6a and 6b do not replicate the large observed variability in ice counts or production rate with updraft fluctuations ∼ \(\Theta (0.5 \text{ m s}^{-1})\) (their Figures 1 and 12d and [Mossop, 1978]).

4.5. Cloud States

The process weightings of Cases 1, 2, and 3 are intended as a mathematical tool to test the parameter space. To ensure atmospheric relevance, a final three simulations are run to characterize different cloud states with parameters in Table 4. A continental convective case is characterized by stronger updrafts, a steeper droplet spectrum, more and smaller droplets initially, and longer characteristic growth times for the liquid phase. The latter three conditions are proxies for higher continental aerosol loading. \(N_{\text{ice}}\) in this case is the smallest of all cloud states, reaching only 0.069 L\(^{-1}\) in 30.3 min. Although the updraft is strong, rapid growth of many, small droplets depletes supersaturation before large hydrometeors in either phase can form and feed into secondary ice production.

The maritime convective case, characterized by intermediate updrafts, is shown in Figure 7b. The initial droplet number, droplet spectrum, and ice crystal radius are all intermediate between that of the continental convective and Arctic stratocumulus cases. \(N_{\text{ice,max}}\) is largest in this case with a value of 7.29 L\(^{-1}\) in 42.1 min.
Figure 7. Ice phase hydrometeor number evolution for the (a) continental convective, (b) maritime convective, and (c) Arctic stratocumulus cloud states. \( N_i \) denotes ice crystals, \( N_g \) small graupel, and \( N_{LG} \) large graupel. The overall hydrometeor number in the ice phase, \( N_{ice} \), is shaded in gray, and the maximum ice number, \( N_{ice,max} \), and simulation duration, \( t_{dur} \), are given for each case.

Here supersaturation generation and consumption balance to allow the greatest ice crystal number enhancement. The initial hydrometeor number and size are large enough that the rime splintering tendency becomes significant, while updraft is strong enough to sustain this secondary production over a matter of minutes.

Finally, the Arctic stratocumulus case is shown in Figure 7c. Here the updraft and initial droplet number are set to much lower values to represent stratiform clouds with limited aerosol loading. The initial droplet and ice crystal radii are assumed to be larger and the characteristic times for the ice phase are shorter. Here \( N_{ice} \) has a maximum of 0.079 L\(^{-1}\) over 56.0 min. Because the initial number of hydrometeors is lower, the parcel remains saturated for a longer period of time, allowing larger hydrometeors and secondary ice production, in particular breakup, to occur. But once these microphysics begin to enhance ice crystal number, the weaker updraft is insufficient to maintain supersaturation.

Many of the enhancements included in Table 1 have been observed in maritime convective systems. The NAMMA campaign took place in the Pacific from the Cape Verde archipelago off the coast of West Africa, while the ICE-T campaign focused on the cumuli in the Caribbean around St. Croix and HAIC-HIWC occurred over the tropical Atlantic off the coast of French Guiana. And while Lawson et al. [2015] have found that strong updrafts may favor frozen droplet shattering, many other studies report only modest updrafts of 1 to 2 m s\(^{-1}\).

5. Discussion

The model microphysics is intentionally limited to facilitate analysis. With fewer linkages between the liquid and ice phase hydrometeor number tendencies, we can better elucidate how increases in one tendency affect another. Nonetheless, it is worthwhile to consider how the inclusion of additional microphysics would affect the results. One important limitation is the use of a time threshold for sedimentation. Including continuous sedimentation will reduce the large hydrometeor number concentrations and increase the magnitude of the secondary production tendencies and simulation duration. Without the continual loss of large hydrometeors, the depositional growth sink of supersaturation is overestimated. A second important limitation is the monodispersity of each hydrometeor class. Larger droplets and graupel, represented by the tail of a size distribution, are those that shatter more effectively, and their omission should also lead to underestimation of the secondary production tendencies. The same rime splintering and breakup parameterizations are being implemented in the more sophisticated COSMO framework, a mesoscale model with double-moment cloud microphysics [Seifert et al., 2012], to assess this effect. Future work will present output of these parameterizations for a case study.

Then simulations are stopped when the parcel becomes subsaturated with respect to water. If this limit were removed, \( N_{eq} \) should increase at the expense of \( N_{ice} \). Larger \( N_g \) and \( N_{LG} \) and smaller \( N_{ice} \) would then shift contributions toward breakup as it depends only on the ice phase. The parcel has also been assumed spatially homogeneous. If “pockets” of one phase or another were to form, then mixed-phase secondary production
through a process like rime splintering would decrease. The contribution of breakup, as a single-phase secondary production process, could again become more significant. Other studies suggest that frozen droplet shattering could be another influential mixed-phase secondary production process ([Lawson et al., 2015; Taylor et al., 2016]). A follow-up study incorporates this process ([Sullivan et al., 2017]).

We could also consider the effect of entrainment of subsaturated air by small-scale eddies. This can have three effects. If the air is subsaturated with respect to both water and ice, and the motions are strong enough to induce homogeneous mixing, supersaturation will drop and may ultimately lead to liquid droplet evaporation and ice crystal sublimation, affecting primarily hydrometeor size not number [e.g., Devenish et al., 2012]. The secondary production collection kernels would be reduced. If the motions are not strong enough to induce homogeneous mixing, only those droplets or ice crystals near the entrained air will evaporate or sublimate, affecting primarily hydrometeor number not size. Finally, if the entrained air is subsaturated only with respect to water, the Bergeron process will generate larger ice hydrometeors more quickly, at the expense of the liquid phase. The breakup contribution in this case could be large, assuming the larger-scale motion is strong enough to keep these large hydrometeors aloft.

A final consideration is the ventilation effect mentioned above. Estimates of a ventilation coefficient from the Froessling equation (\(Sh/2 = 1 + 0.276(Re)^{1/2}/(Sc)^{1/3} = \bar{f}_v\)) indicate that it may be on the order of 10 for the graupel classes. Convectively enhanced mass transfer will generate the large hydrometeors more quickly, fueling secondary production if the updraft is sufficient to keep them aloft and maintain supersaturation. But including this effect does not seem to counterbalance the diminished secondary production from spherical ice hydrometeors in section 4.2. While coefficients are larger for spherical hydrometeors than nonspherical ones due to a larger characteristic velocity and density in the Reynolds number, these effects are somewhat offset by a smaller characteristic length, and our estimates indicate that the two values would be within 1% of one another for the default simulations.

6. Conclusions

We have performed 15 simulations, detailed in Tables 3 and 4, with a six-hydrometeor-class, mixed-phase microphysics parcel model. These consider the effect of microphysical process weightings, graupel nonsphericity, and turbulence on the total ice hydrometeor number, \(N_{ice}\), and process contributions, \(p_X\), in the parcel. In particular, we have shown the following:

1. The largest ice crystal number enhancement occurs for intermediate conditions, when all microphysical processes are moderately active. Case 2 and Case MC produce the largest \(N_{ice,max}\) from the process weighting and cloud state simulations, respectively. These simulations are the intermediate cases in which neither the process weightings nor the dynamic-aerosol proxy conditions are too extreme. The large \(N_{ice,max}\) in Case 2 indicates that secondary ice production is promoted when the microphysics which produce larger hydrometeors are also active. For Case MC, secondary ice production is promoted by a balance of moderate updraft and moderate aerosol loading, \(u_z\) is strong enough and \(N_{tot}\) is low enough to maintain supersaturation throughout secondary ice production. The addition of a continual sedimentation sink could, however, shift the favorable zone to higher updrafts which keep large hydrometeors aloft longer.

2. The relative contribution of rime splintering versus breakup is determined primarily by when large hydrometeors form in the liquid versus ice phase. Secondary ice production occurs after the formation of larger hydrometeors, which can rime or break up. If conditions favor large droplet formation, the rime splintering contribution will dominate. If conditions favor large graupel formation, the breakup contribution will dominate. This finding extends the first: intermediate conditions favor secondary ice production because they allow the development of larger hydrometeors in one phase or the other. For example, large droplets form quickly when coalescence is efficient. Broad droplet size distributions accelerate coalescence and result from an intermediate aerosol loading, with enough CCN for sufficient initial activation but also few enough for growth of some droplets to large sizes. The number of large hydrometeors in either phase may be underestimated without a size distribution for each class, so future work will assess the effect of these parameterizations in a more complete bulk microphysics scheme.

3. Including graupel nonsphericity significantly increases the secondary ice production rate. Representing graupel growth with spheroidal, rather than spherical, geometry leads to the development of larger maximum dimensions and to an increase in the aggregation, breakup, and rime splintering collection kernels,
predominantly through the collisional cross section. The faster ice production rates in the default simulations indicate that inclusion of ice hydrometeor nonsphericity leads to faster cloud glaciation.

4. **Lower updraft velocity may cause a “dynamically limited case” with diminished secondary production.** When the average updraft is lower, nucleation rate and the fragments generated upon breakup are decreased because of the temperature dependence of these factors. Fewer ice crystals are then formed, decreasing graupel numbers and the secondary production tendencies. In this case, the parcel remains supersaturated for a long time and has lower $N_{\text{ice, max}}$ and later $P_{\text{br, max}}$. Here the addition of continuous sedimentation would exaggerate the updraft limitation since any large hydrometeors would also be quickly lost to this sink. A consideration outside the scope of this zero-dimensional model but interesting for future study is the effect of phase separation within the cloud. Turbulence could entrain subsaturated air and initiate the Bergeron process, which could lead to larger contributions from breakup.

### Notation

- $a_g$: major axis of spheroidal, small graupel
- $a_{G}$: major axis of spheroidal, large graupel
- $B_0$: parameter within the hydrometeor terminal velocity calculation from Mitchell and Heymsfield [2005]
- $c_0$: rate of primary ice production as a function of temperature from DeMott et al. [2010]
- $c_p$: heat capacity of ambient air
- $c_X$: capacitance of ice hydrometeor $X$
- $d_0$: rate of new droplet activation as a function of supersaturation from Twomey [1959]
- $D$: diffusivity of water vapor in air as a function of temperature and pressure
- $e_w$: saturation vapor pressure with respect to water
- $e_i$: saturation vapor pressure with respect to ice
- $\eta_X$: weighting for process $X$ from 0 to 100%; $X = (R)ime (S)plintering; (coal)escence; (agg)regation; (br)eakup
- $f$: nucleation reduction or the factor used to reduce primary ice production from DeMott et al. [2010] for warmer subzero temperatures
- $F$: leading coefficient in the temperature-dependent function for fragments generated from breakup
- $G_{\text{dop}}$: droplet number generation function
- $G_{\text{ice}}$: ice crystal number generation function
- $H$: heaviside function
- $k_a$: thermal conductivity of air
- $K_X$: gravitational collection kernel for process $X$
- $N_{\text{br}}(T)$: number of ice fragments generated by breakup upon collision, a chi-square-distribution function of temperature based on data from Takahashi et al. [1995]
- $N_d$: small liquid droplet number
- $N_g$: small graupel number
- $N_G$: large graupel number
- $N_i$: ice crystal number
- $N_{\text{ice}}$: total ice phase hydrometeor number, $N_i + N_g + N_G$
- $N_{\text{eq}}$: total liquid phase hydrometeor number, $N_d + N_r + N_R$
- $N_r$: medium liquid droplet number
- $N_R$: large liquid droplet number
- $N_{\text{RS}}$: number of ice fragments generated by rime splintering as a function of rimed liquid mass as in Mossop and Hallett [1974]
- $p_X$: contribution of a microphysical process $X$ to total $N_{\text{ice}}$
- $q_i$: mixing ratio of ice
- $q_{lw}$: mixing ratio of liquid water
- $r_d$: small liquid droplet radius
- $r_i$: ice crystal radius
- $r_r$: medium liquid droplet radius
- $r_R$: large liquid droplet radius
- $R_a$: specific gas constant for moist air
- $R_v$: specific gas constant for water vapor
- $s_w$: supersaturation with respect to water
References


Δt_{dur} simulation duration, or the time until the parcel becomes water-subsaturated

Δt_{max} lower temperature bound beyond which no fragments are generated from breakup upon collision

V_{iX} terminal velocity of hydrometeor type i

τ_{iX} characteristic time for hydrometeor number in class X to grow by deposition, riming, or condensation to the next largest class or to fall out

γ adjustable parameter to control the decay rate of fragments generated from breakup on collision at warmer subzero temperatures

Γ adiabatic lapse rate

Γ_{IG} inherent growth factor

μ viscosity of air

ρ_a density of air

ρ_i density of bulk ice

ρ_w density of liquid water

ρ_{de} deposition density for graupel as in Chen and Lamb [1994]

ξ ratio of saturation vapor pressure with respect to water over saturation vapor pressure with respect to ice

ΔH_v heat of sublimation for water

ΔH_v heat of vaporization for water

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