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Initiation of secondary ice production in clouds

Sylvia C. Sullivan[1], Corinna Hoose[2], Alexei Kiselev[2], Thomas Leisner[2], and Athanasios Nenes[1,3,4,5]

[1]School of Chemical and Biomolecular Engineering, Georgia Institute of Technology, Atlanta, GA 30332, USA
[2]Institute of Meteorology and Climate Research, Karlsruhe Institute of Technology, Karlsruhe, Germany
[3]School of Earth and Atmospheric Sciences, Georgia Institute of Technology, Atlanta, GA 30332, USA
[4]ICE-HT, Foundation for Research and Technology, Hellas, 26504 Patras, Greece
[5]IERSD, National Observatory of Athens, P. Penteli, 15236, Athens, Greece

Correspondence to: A. Nenes (athanasios.nenes@gatech.edu)

Abstract. Disparities between the measured concentrations of ice-nucleating particles (INP) and in-cloud ice crystal number concentrations (ICNC) have led to the hypothesis that mechanisms other than primary nucleation form ice in the atmosphere. Here, we model three of these secondary production mechanisms – rime splintering, frozen droplet shattering, and breakup upon collision – with a six-hydrometeor-class parcel model. We perform three sets of simulations to understand temporal evolution of ice hydrometeor number (N_{ice}), thermodynamic limitations, and the impact of parametric uncertainty when secondary production is active. Output is assessed in terms of the number of primarily nucleated ice crystals that must exist before secondary production initiates ($N_{INP}^{(lim)}$), as well as the ICNC enhancement from secondary production and the timing of a 100-fold enhancement. N_{ice} evolution can be understood in terms of collision-based non-linearity and the 'phasedness' of the process, i.e., whether it involves ice hydrometeors, liquid ones, or both. Breakup is the only process for which a meaningful $N_{INP}^{(lim)}$ exists (0.002 L^{-1} up to 0.07 L^{-1}). For droplet shattering and rime splintering, a warm enough cloud base temperature and modest updraft are the more important criteria for initiation. The low values of $N_{INP}^{(lim)}$ here suggest that, under appropriate thermodynamic conditions for secondary ice production, perturbations in CCN concentrations are more influential on mixed-phase partitioning than those in INP concentrations.

1 Background

Number concentrations of ice-nucleating particles (N_{INP}) in the atmosphere span orders of magnitude from a few per cubic meter up to 100s per liter (e.g., DeMott et al., 2010). At temperatures greater than about -15°C, these concentrations remain low: only one particle in every 10^3 or 10^4 will nucleate an ice crystal Rogers et al. (1998); Chubb et al. (2013); DeMott et al. (2015). However, even when INP concentrations are low at warm subzero temperatures, in-cloud ice crystal number concentrations (ICNC) can be orders of magnitude higher (e.g., Hallett and Mossop, 1974; Heymsfield and Willis, 2014; Lasher-Trapp et al., 2016; Taylor et al., 2016; Ladino et al.), particularly in tropical maritime clouds Koenig (1963, 1965); Hobbs and Rangno (1990).

This discrepancy may be explained in some cases by shattering upon cloud probe tips (Field et al., 2003; Heymsfield; McFarquhar et al., 2007), but even as instrumentation and algorithms have been developed to minimize these artifacts (Korolev et al.;

Korolev and Field, 2015), the disparity has remained, supporting several hypothesized secondary ice production processes. Hallett and Mossop (1974) proposed rime splintering in which ice hydrometeors collide with and freeze supercooled droplets to form rime, which then splinters off as the hydrometeor continues to fall. Droplets in cases of rime splintering tend to be both less than 13 μm and greater than 25 μm in diameter, and temperatures fall between -3 and -8°C (Mossop, 1978; Heymsfield and Mossop, 1984; Mossop, 1985); however, ICNC enhancement, i.e., the increase in ICNC beyond that generated by primary nucleation, exists even outside of these conditions.

Another hypothesized mechanism is the shattering of droplets with a diameter of 50 to 100s of μm upon freezing (Mason and Maybank, 1960; Leisner et al., 2014; Lawson et al., 2015). At sufficiently cold temperatures, latent heat release leads to the formation of a liquid core-ice shell structure that eventually shatters upon internal pressure build-up. A third mechanism, independent of the liquid phase, is breakup upon mechanical collision of ice hydrometeors. Vardiman (1978) calculated the fragment number generated during breakup from a change in momentum, and Takahashi et al. (1995) later conducted experiments with a rotating ice sphere in a cloud chamber to estimate the number of ice crystals ejected versus temperature. Yano and Phillips (2011), and more recently Yano et al. (2015), have identified ‘explosive regimes’ defined by non-dimensional parameters, where breakup may enhance ICNC by as much as 10^4.

Laboratory and in-situ data of these processes, especially droplet shattering and breakup upon collision, are difficult to obtain, and their fragment generation functions and temperature dependence remain uncertain (Field et al., 2017). Small-scale models provide a good tool to estimate variability of secondarily-produced ICNC with these parameters and the minimum number of INP needed to initiate secondary production, called $N_{INP}^{(lim)}$ hereafter.

Some previous studies have estimated $N_{INP}^{(lim)}$ on the basis of in-situ data. For example in a study of ice initiation in cumulus, Beard (1992) found that a nucleated ICNC of 0.001 L^{-1} could trigger raindrop freezing around -5°C . More recently, Crawford et al. (2012), with Aerosol Properties, PRocesses And InFluenceS (APPRAISE) campaign data, and Huang et al. (2017), with Ice and Precipitation Initiation in Cumulus (ICEPIC) campaign data, identified a primarily nucleated ICNC of 0.01 L^{-1} as sufficient to initiate rime splintering. Connolly et al. (2006a) found that the rime splintering tendency increased with increasing primarily-nucleated ICNC, but this result was based upon adjusting the primary nucleation rate rather than the absolute N_{INP}. Clark et al. (2005) also adjusted the primary nucleation rate relative to the rime splintering one, but gave no approximate $N_{INP}^{(lim)}$ values or thermodynamic constraints. These studies have also considered only rime splintering, despite evidence that multiple processes occur simultaneously (Rangno and Hobbs, 2001). We provide more comprehensive estimates of $N_{INP}^{(lim)}$ here for three secondary production processes above over a range of fragment numbers and thermodynamic conditions.

2 Parcel model

To estimate ICNC enhancements and $N_{INP}^{(lim)}$, we run a parcel model with six hydrometeor classes for small ice crystals and droplets, small and large graupel, and medium and large droplets (Sullivan et al., 2017). The number in these classes is denoted N_i, N_d, N_g, N_G, N_r, and N_R respectively. The microphysics consists of primary nucleation and secondary production by breakup upon collision, rime splintering, and frozen droplet shattering, as described by an ice generation function:

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$$G_{ice} = c_0 H(t) + \eta_{BR} K_{BR} \aleph_{BR} N_g N_G + \eta_{RS} \aleph_{RS} \Big[K_{RS,g} N_g + K_{RS,G} N_G \Big] N_R + \eta_{DS} \aleph_{DS} N_R. \quad (1)$$

c_0 is the primary nucleation rate derived from the temperature dependence of the immersion INP concentration given in DeMott et al. (2010); H is a Heaviside function; η_X is the weighting for process X, either 100% when the process is active or 0% when it is inactive; K_X is a gravitational collection kernel for process X; and $\aleph_X$ is the fragment number generated by process X. The specific forms of $\aleph_X$ are given in Table S1; in particular, $\aleph_{DS}$ contains a product of droplet freezing and shattering probabilities, p_{fr} and p_{sh}. BR stands for breakup upon collision, DS for droplet shattering, and RS for rime splintering. Later, the droplet shattering tendency, denoted $DScoll$, is modified to represent a collisional process with a product of large droplet and ice crystal numbers. η_{RS} is set to 1% outside the optimal rime splintering temperature zone of -3 to -8°C. For the liquid phase, a droplet generation function consists simply of droplet activation, calculated from a Twomey power-law formulation. 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 and coalescence losses. The time delay quantifies how long depositional, riming, or condensational growth to the next hydrometeor class will take and is solved for approximately from growth equations. The six hydrometeor number tendencies are coupled to moist thermodynamic equations for pressure, temperature, supersaturation, mixing ratios, and hydrometeor sizes. In particular, newly produced ice crystals are assumed to be spherical with bulk ice density, while graupel is assumed to be spheroidal with a deposition density and non-unit capacitance as in Chen and Lamb (1994). The model microphysics is shown schematically in Figure S1, and parameter values and sources are given in Table S1. Model assumptions, explicit thermodynamic tendencies and correlations, and collection kernels are detailed in Sullivan et al. (2017).

3 Simulations

The three rows of Table 1 show three sets of simulations with the parcel model. First we investigate the evolution of the total ice hydrometeor number, N_{ice}, i.e. the summation of N_i, N_g, and N_G, in default simulations with fixed fragment numbers and thermodynamic conditions. Simulation acronyms include BR for breakup upon collision, DS for droplet shattering, RS for rime splintering, or ALL if all processes are active (see also Table 1 caption). These runs address how the value of $N_{INP}^{(lim)}$ and enhancement magnitude or timing vary when different processes are active. We quantify enhancement from secondary production as the ratio of the total ICNC to the number generated by primary nucleation when the simulation ends, i.e., when the parcel becomes water subsaturated or reaches a temperature of 237 K where homogeneous nucleation may occur: $N_{ice}^{(max)}/N_{INP}(t_{end})$. An enhancement of 10 can be understood as *at least* a 10-fold increase in ICNC due to secondary production, as an aggregation sink is also active in the simulations. In the absence of secondary production, ICNC enhancement does not exceed one.

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Table 1. All simulations with parameters adjusted from the default values in Table S1. A control run with no secondary production, i.e., $\eta_{DS} = \eta_{BR} = \eta_{RS}$ = 0% is denoted INP below. Thermodynamic simulations are run with combinations (BRDSth, BRRSth, and DSRSth) or all (ALLth) of the processes and shown solely in the Supplement.

Run BR	***Run DS***	***Run RS***
	(Run DScoll)	
Breakup upon collision	Droplet shattering only	Rime splintering
only	(Collisional droplet shattering only)	only
$\eta_{DS} = \eta_{RS} = 0\%$	$\eta_{BR} = \eta_{RS} = 0\%$	$\eta_{BR} = \eta_{DS} = 0\%$
Run BRth	***Run DSth***	***Run RSth***
Thermodynamic variations	Thermodynamic variations	Thermodynamic variations
for breakup	for droplet shattering	for rime splintering
u_z = { 0.1, 0.5, 1, 1.5, 2, 2.5, 3, 3.5, 4 m s^{-1}}	T_0 = { 256, 258, 260, 262, 264, 268, 270, 272 K}	
Run BRpp	***Run DSpp***	***Run RSpp***
Parameter perturbations	Parameter perturbations	Parameter perturbations
for breakup	for droplet shattering	for rime splintering
F_{BR} = {0, 90, 140, 200, 280}	F_{DS} = {25, 75}x 10$^{-12}D^{-4 \text{ or } -3}$	F_{RS} = {9, 15, 30, 45, 80}
	(β, γ) = { (-0.016, 500), (-0.015, 400)}	x 10^{7} (kg rime)$^{-1}$
T_{min} = {246, 249, 252, ...	$p_{sh}^{(max)}$ = {1, 5, 10, 20, 30%}	
255, 258 K}		

The second set of simulations considers the effect of updraft velocity and initial temperature in the parcel; this set is denoted 'th' for thermodynamics. The updraft is varied from 0.1 up to 4 m s^{-1} to simulate both stratiform and convective conditions, while the initial parcel temperature is adjusted from just below freezing (272 K) down below the peak of the droplet shattering probability distribution (256 K). These conditions also ensure numerical stability, given the stiffness of the coupled equations. The final set, denoted 'pp', performs parameter perturbations. In particular, we vary the leading coefficient of the fragment number generated per collision, F_{BR}; the minimum temperature for which breakup occurs, T_{min}; the functional form of the fragment number generated per shattering droplet; and the maximum of the temperature-dependent droplet shattering probability distribution, $p_{sh}^{(max)}$. The effect of these parameters on the generated fragment numbers is shown in Figure S2, and the alternate sigmoid functional forms for $\aleph_{DS}$ are shown in Figure S3.

3.1 Hydrometeor number evolution

The temporal evolution of N_{ice} in the default simulations is shown in Figure 1. Each simulation is done for a range of total INP number within the parcel, $N_{INP}^{(tot)}$. The structure in the number evolution can be understood by considering whether the process is collisional and whether it involves hydrometeors in one or both phases (liquid or ice).

N_{ice} from breakup and rime splintering evolves non-linearly, as these processes involve a product of hydrometeor numbers. Independent of $N_{INP}^{(tot)}$, N_{ice} grows steadily throughout the simulation for these collisional secondary production processes. Even as graupel or large droplets are consumed, those hydrometeors still in the parcel continue to grow by deposition or condensation respectively. This ongoing hydrometeor growth increases the secondary production tendencies via their collection kernels, and this link itself is non-linear because both hydrometeor terminal velocity and collisional cross section increase with growth.

Droplet shattering is not modeled as a collisional process here, and its tendency is only proportional to a single hydrometeor number, N_R. As a result, N_{ice} does not grow steadily throughout the DS simulation, but rather exhibits threshold behavior when the temperature becomes cold enough for a non-negligible freezing probability according to (Bigg, 1953). A decrease in N_{ice} occurs right before the sudden increase for the DS simulation because large graupel begin to fall out of the parcel around 45 minutes. Below in Section 3.1.1, when we model collisional droplet shattering ($DScoll$), a steady increase appears again.

The ice phase is also influential for the enhancement timing from rime splintering or breakup. Because their tendencies involve graupel numbers, increasing $N_{INP}^{(tot)}$ boosts their rates of generation. For breakup, a parcel with 0.0129 L^{-1} INP reaches 10 $L^{-1} N_{ice}$ in 23 minutes, while that with 0.167 L^{-1} INP reaches the same value in 17 minutes. For rime splintering, the same increase in INP shifts the time to reach 10 L^{-1} N_{ice} from 30 minutes back to 25. While these differences in enhancement timing sound small, they can help infer which secondary production processes are active from in-situ N_{INP} and ICNC data. For example, ICNC on the order of hundreds per liter can form within 10 to 15 minutes (Hobbs and Rangno, 1990; Rangno and Hobbs, 1991, 1994). This timing is too rapid to be explained by rime splintering alone (Mason, 1996), in agreement with our RS simulation. Simulations with breakup and rime splintering in combination, on the other hand, *are* sufficiently rapid (Fig. S4b).

As with enhancement timing, its magnitude can be explained in terms of non-linearity and hydrometeor phases involved. The breakup tendency is proportional to the product of two ice hydrometeor numbers, N_g and N_G, so the impact of varying $N_{INP}^{(tot)}$ is most pronounced for the BR simulations. Increasing $N_{INP}^{(tot)}$ by two orders of magnitude (0.001 to 0.167 L^{-1}) increases $N_{ice}^{(max)}$ by four order of magnitude (0.0023 to 37.6 L^{-1}). The rime splintering and droplet shattering tendencies are proportional to N_R which is around 10^6 times as large as N_g or N_G, so the impact of $N_{INP}^{(tot)}$ for these processes is diluted. For the purely liquid-phase droplet shattering, the two-order-of-magnitude increase in $N_{INP}^{(tot)}$ has no significant impact on $N_{INP}^{(max)}$. For rime splintering, it actually translates to a two-fold decrease in $N_{ice}^{(max)}$ (30.58 to 16.67 L^{-1}). This decrease is the result of an increasing denominator in the $N_{ice}^{(max)}/N_{INP}(t_{end})$ expression (see also the RS panels of Figures 3 and 4 below). The rime splintering tendency is strong enough that it always generates additional ice crystals, so increasing $N_{INP}^{(tot)}$ actually decreases

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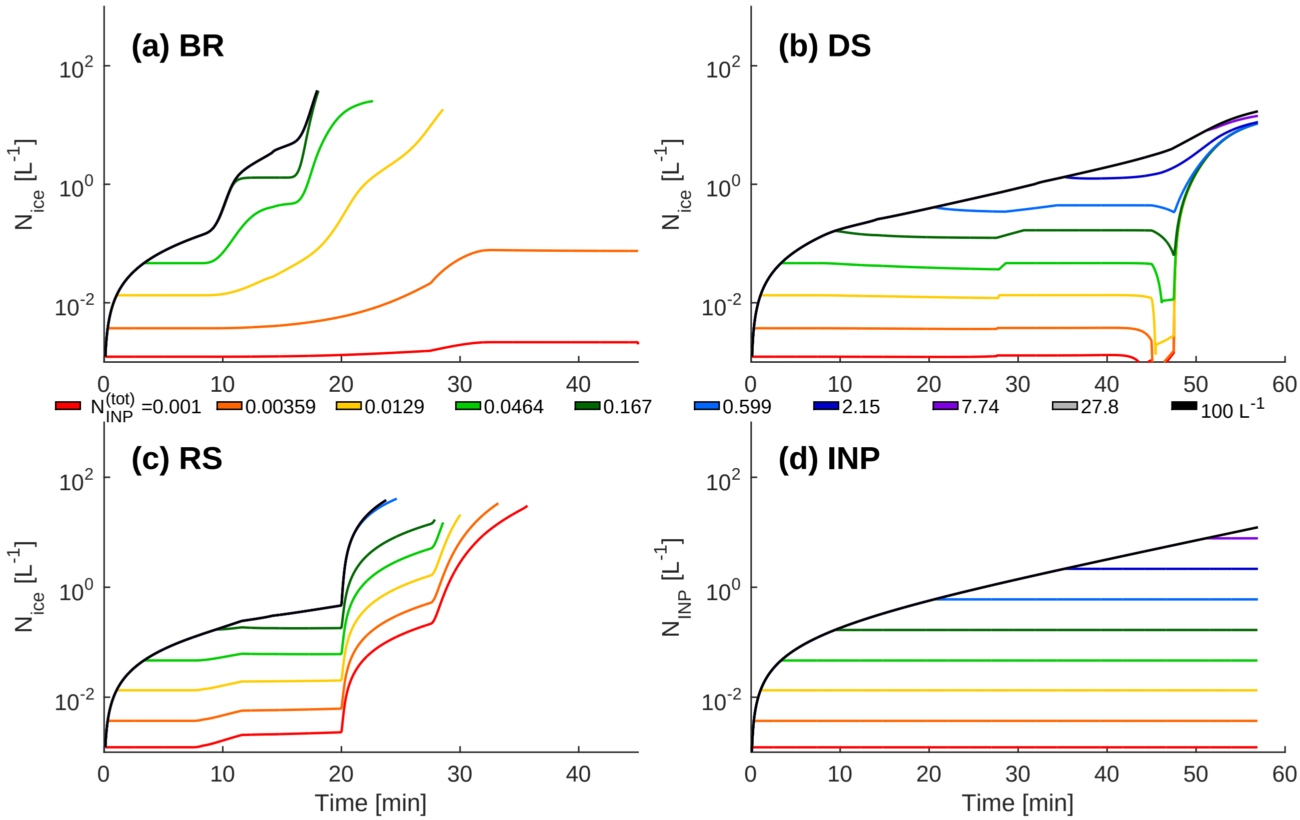

Figure 1. Evolution of the total ice hydrometeor (summation of ice crystal, small and large graupel numbers) number for default simulations with a range of $N_{INP}^{(tot)}$ from 0.001 L^{-1} up to 100 L^{-1}: **(a)** breakup upon collision only, **(b)** droplet shattering only, **(c)** rime splintering only, and **(d)** a control run when only primary nucleation is active. These default simulations are run for u_z of 2 m s^{-1} and T_0 of 272 K.

enhancement. The total INP number does, however, affect which rimers contribute to enhancement: when $N_{INP}^{(tot)}$ exceeds 0.167 L^{-1}, only rime splintering of small graupel can occur before subsaturation of the parcel.

Finally, increasing $N_{INP}^{(tot)}$ increases the ice generation rates from breakup and rime splintering only up to a certain point. Beyond an $N_{INP}^{(tot)}$ around 0.599 L^{-1}, additional INP do not increase $N_{ice}^{(max)}$. The parcel is in a supersaturation-limited regime, for which it becomes subsaturated before the effect of additional primary nucleation can be felt by secondary production.

3.1.1 Collisional droplet shattering

As an extension of the default simulations only, we consider N_{ice} evolution and enhancement from droplet shattering as a collisional process; no parameter perturbations or varying thermodynamics are run for this collisional formulation. In this case,

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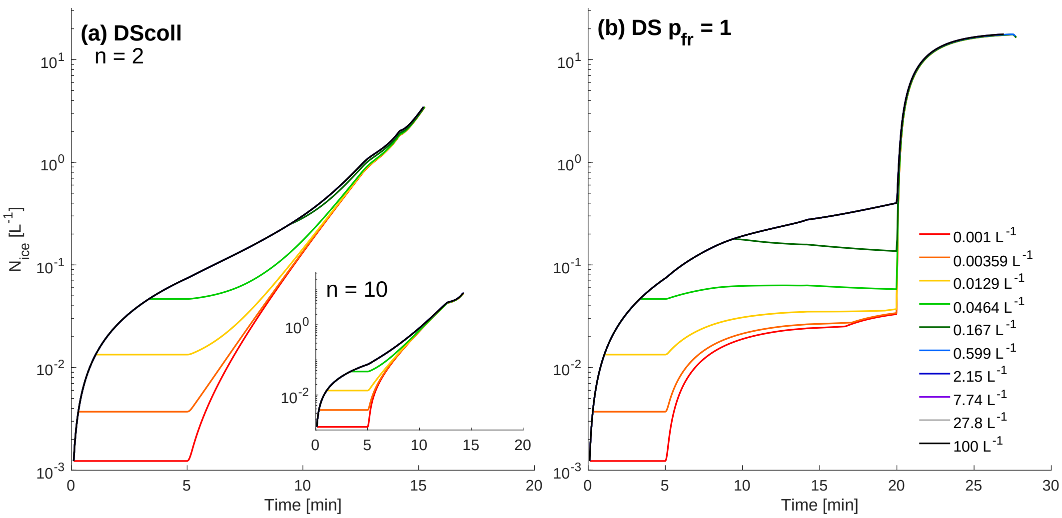

Figure 2. Evolution of N_{ice} for **(a)** collisional droplet shattering and **(b)** droplet shattering with p_{fr} of 1 over the same range of $N_{INP}^{(tot)}$ as in Figure 1. p_{sh} is set to the default value of 20%. For the main panel **(a)**, droplet shattering generates 2 fragments per collision, and for the inset, 10 fragments per collision. These extensions to the default simulations are run for u_z of 2 m s^{-1} and T_0 of 272 K.

the tendency is proportional to both N_R and N_i, rather than just N_R as in Equation 1:

$$\left.\frac{dN_i}{dt}\right|_{DS} = \eta_{DS} K_{DS} \aleph_{DS}^{(coll)} N_R N_i \qquad (2)$$

The fragment number from Lawson et al. (2015) ($F_{DS} D_R^4$) and p_{sh} are retained as in the DS simulation, but p_{fr} is removed with the understanding that the ice crystal-droplet collision initiates the freezing.

In Figure 2a, the threshold behavior of the enhancement from pure liquid droplet shattering is replaced by a steady increase similar to that from rime splintering or breakup. In fact, the growth in N_{ice} is now more gradual than that from RS or BR because N_i is also consumed by collisions now; there is effectively a linear increase in log space as $dN_i/dt \propto N_i$. This combined source and sink of N_i from droplet shattering also yields a smaller $N_{ice}^{(max)}$ of only 3.47 L^{-1} when 2 fragments are generated per collision and 7.87 L^{-1} when 10 are generated. The enhancement timing, on the other hand, has been much accelerated to about 14 (n = 10) or 15 (n = 2) minutes. $N_{INP}^{(tot)}$ still has no meaningful effect on either enhancement magnitude or timing.

As an uppermost bound for the enhancement from droplet shattering, we rerun the DS simulation with p_{fr} set to 1. In this case, an $N_{ice}^{(max)}$ of 17.67 L^{-1} is obtained over 27.8 minutes, not as fast as the collisional droplet shattering but about twice as fast as DS with non-unity p_{fr}. The temperature-dependent freezing probability above delays the DS enhancement, and in cases where p_{fr} is higher, droplet shattering may occur much more rapidly. Future work should also incorporate a dependence of p_{fr} on the number of submerged INP Paukert et al. (2017), rather than just on time and temperature. Temperature and updraft dependencies are investigated in more detail next.

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3.2 Varying thermodynamics

Secondary enhancements from the simulations with varying thermodynamics are shown in Figures 3 and 4. Runs are performed for a range of updraft velocities and initial temperatures given in Table 1, but we focus on the extremes, as behavior in between is intermediate.

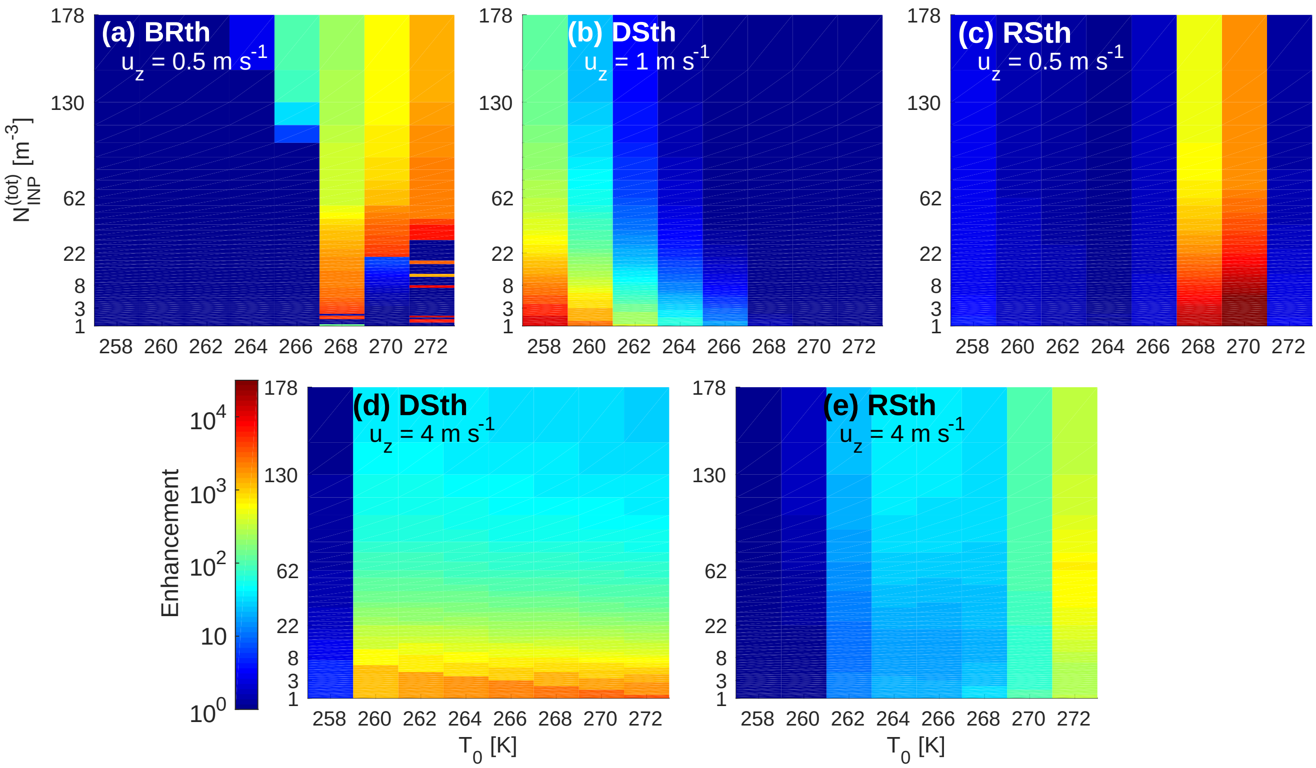

Figure 3. Ice crystal number concentration enhancement, i.e., $N_{ice}(t_{end})/N_{INP}(t_{end})$, for the thermodynamics simulations at various values of $N_{INP}^{(tot)}$, the total INP number in the parcel, and T_0, the initial temperature. Red indicates a larger enhancement per INP. Panels **(a)**, **(b)**, and **(c)** show the enhancement for breakup and rime splintering at a low, stratiform-like updraft of 0.5 m s^{-1}. Droplet shattering is shown at 1 m s^{-1} because only very small enhancements occur at lower u_z. Panels **(d)** and **(e)** show the enhancement for droplet shattering and rime splintering at a higher, convective-like updraft of 3.5 m s^{-1}. No meaningful enhancements are generated by breakup at this larger updraft.

The top panels of Figure 3 show enhancements for stratiform conditions, i.e. u_z of 0.5 or 1 m s^{-1}, and a range of cloud base temperatures T_0. $N_{INP}^{(lim)}$ values for breakup upon collision can be seen in panel **(a)**. As T_0 decreases from 272 to 270 to 268 K, $N_{INP}^{(lim)}$ drops from 32.8 to 21.5 to 2.1 m^{-3}. At 266 K, $N_{INP}^{(lim)}$ increases again, reaching an $\mathcal{O}(10^2)$ enhancement only for an INP concentration of 0.143 L^{-1}. Larger ICNC occur only at these warmer T_0 because the parcel remains in the mixed-phase

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temperature range long enough that large graupel can form. For droplet shattering and rime splintering, there is no $N_{INP}^{(lim)}$ value greater than 1 m^{-3}.

Then when u_z is increased to 4 m s^{-1} in the bottom panels, the T_0 range over which droplet shattering and rime splintering occur expands, while the enhancement magnitude shrinks. If T_0 is too cold and u_z is too strong, or conversely T_0 is too warm and u_z is too weak, the parcel does not remain in the appropriate temperature range for a long enough time to generate large hydrometeors that can shatter or collide. In particular, enhancement from breakup disappears for all T_0 values at a larger u_z because the parcel is too short-lived for graupel to form again. As the parcel moves faster, it is more likely to pass through the 'RS temperature zone' of 267 to 269 K or obtain higher p_{sh} or p_{fr}, but it also spends less time in these optimal zones.

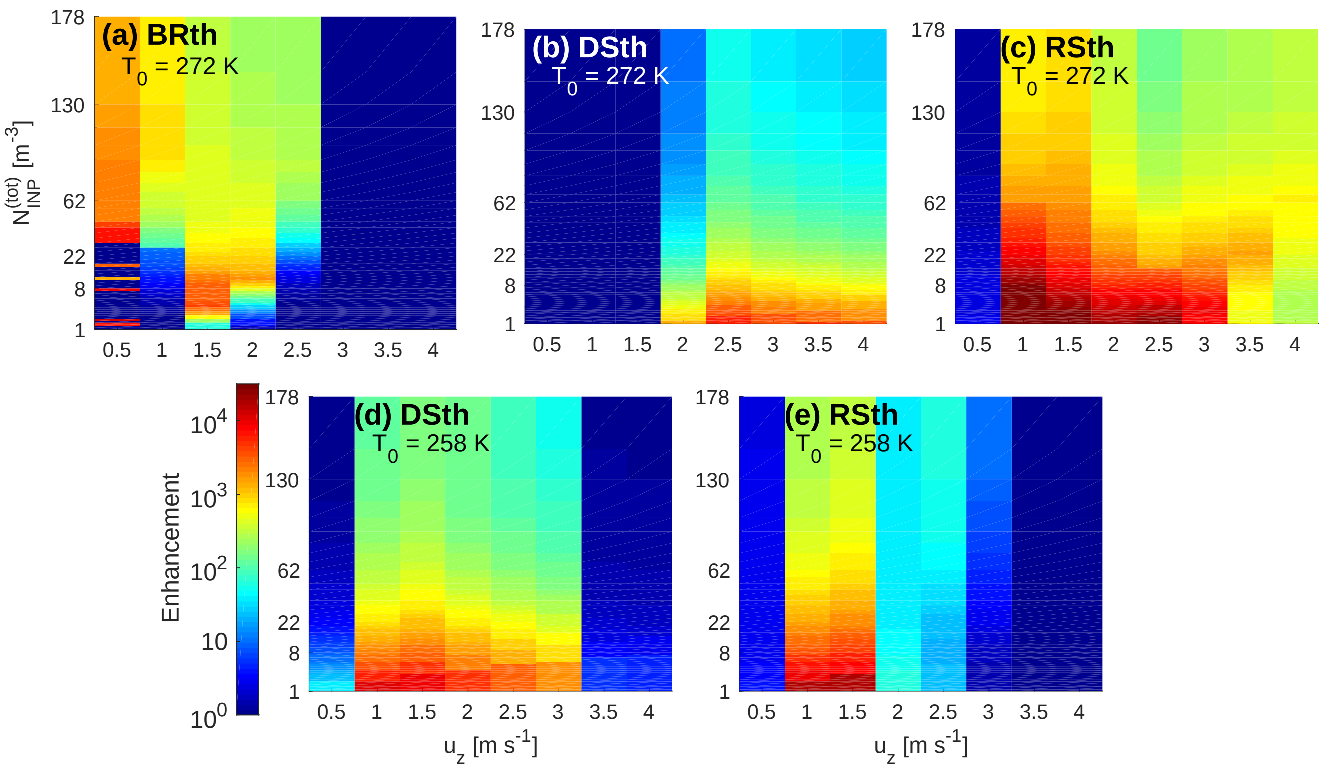

Figure 4. Ice crystal number concentration enhancement, i.e., $N_{ice}(t_{end})/N_{INP}(t_{end})$, for the thermodynamics simulations at various values of $N_{INP}^{(tot)}$, the total INP number in the parcel, and u_z, the updraft velocity. Red indicates a larger enhancement per INP. Panels **(a)**, **(b)**, and **(c)** show the enhancement for breakup, droplet shattering, and rime splintering only at a warmer cloud base temperature of 272 K. Panels **(d)** and **(e)** show the enhancement for droplet shattering and rime splintering at a colder cloud base temperature of 258 K. No meaningful enhancements are generated by breakup at this colder T_0.

If instead, we fix T_0 and look at a range of u_z as in Figure 4, breakup remains the only process with a defined $N_{INP}^{(lim)}$. This value decreases from 32.8 m^{-3} at 0.5 m s^{-1} down to 1.52 m^{-3} at 1.5 m s^{-1}. At 2.5 m s^{-1}, it increases back up to 50 m^{-3}, and at the fastest updraft velocities, no enhancement from breakup occurs.

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For colder T_0, the idea of a 'sweet spot' in u_z appears again. The updraft must be strong enough that large droplets form by condensational growth but modest enough that these droplets remain in an appropriate temperature range for long enough. These trends are summarized in the first panel of Figure 8 and agree generally with Mossop (1985) in which enhancement was possible down to 0.55 m s^{-1} but highest around 1.8 to 2 m s^{-1}. Mossop used a shell-fracture hypothesis to explain this optimum: too high a velocity and the riming drop spreads across the ice surface, rather than forming a fragile protuberance, and too small a velocity and an incomplete ice shell may form around the riming drop. Although not a validation of this hypothesis, the simplified model is, interestingly, able to reproduce this u_z behavior without such detailed rime physics.

Although there is no meaningful $N_{INP}^{(lim)}$ for droplet shattering or rime splintering, N_{INP} still affects enhancement from these processes. In fact, increasing $N_{INP}^{(tot)}$ generally decreases enhancement for all $u_z - T_0$ conditions. This can be understood in terms of a sort of INP efficiency: the highest ICNC per INP is produced when $N_{INP}^{(tot)}$ is lowest. Mathematically, increasing $N_{INP}^{(tot)}$ increases the denominator of the enhancement ratio without a corresponding increase in the numerator. Physically, a higher $N_{INP}^{(tot)}$ depletes supersaturation more rapidly, as many small ice crystals grow by deposition, or it may keep the parcel warmer with latent heating. Fragment numbers, $\aleph_{DS}$ and $\aleph_{RS}$, also depend on the large droplet radius or rimed mass, which are reduced at lower supersaturation. Previous work corroborates this understanding: Connolly et al. (2006a) found that increasing primary nucleation led to a decrease in the freezing of rain in cloud resolving simulations, while many studies have shown the importance of liquid phase properties to the rime splintering tendency (Mossop, 1978, 1985; Hobbs and Rangno, 1985; Heymsfield and Willis, 2014; Lawson et al., 2015).

Finally, Figures 3 and 4 show enhancement from a single process, but enhancement from multiple secondary production processes simultaneously can generally be understood as the linear combination of that from these single processes (Figures S5, S6, or S7). For example, the pattern of enhancement from ALLth in Figure S5 looks like the addition of the patterns from RSth, DSth, and BRth in Figure 3.

3.3 Parameter perturbations

Lastly we use the insight about N_{ice} evolution and approximate enhancements from the above simulations to investigate the impact of adjustable parameters. In particular, we look at the effect of generated fragment numbers and temperature dependencies on $N_{INP}^{(lim)}$ and enhancement magnitude or timing.

The top panels of Figure 5 show the effect on $N_{INP}^{(lim)}$ from breakup for the default nucleation rate and one reduced by factors of 10 and 100. The conditions for which no enhancement occurs are shown in black in Figure 5, and the number of these points increases dramatically as the nucleation rate decreases from left to right (8 to 32 to 84%). Then as T_{min} increases, the temperature range over which breakup occurs shrinks, and $N_{INP}^{(lim)}$ increases: more ice crystals are needed initially to reach a 100-fold enhancement ultimately. As F_{BR} increases, more fragments are formed per collision, and $N_{INP}^{(lim)}$ decreases. This second effect of F_{BR} is the larger of the two. These $N_{INP}^{(lim)}$ trends for breakup occur until a sufficiently low F_{BR} or sufficiently high T_{min}, beyond which enhancement does not occur for any value of $N_{INP}^{(tot)}$ (up to 300 L^{-1}).

The bottom panels show N_{ice} evolution for various values of F_{BR} and T_{min} and for $N_{INP}^{(tot)}$ of 0.0129 L^{-1} (in yellow) and 0.167 L^{-1} (in green). The effect of both parameters is much larger when $N_{INP}^{(tot)}$ is small. Increasing F_{BR} from 40 to 280

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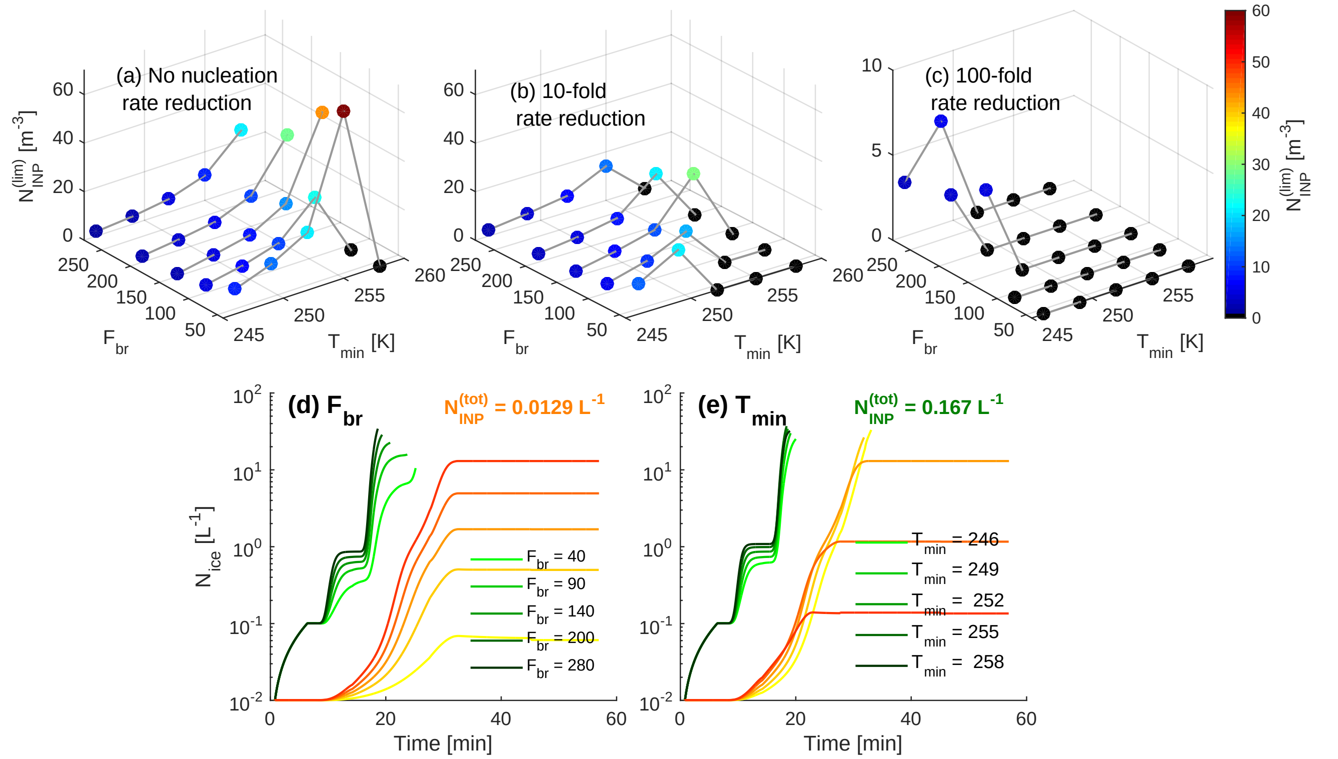

Figure 5. Results from the parameter perturbation simulations with breakup upon collision. The top panels show $N_{INP}^{(lim)}$ to obtain a 100-fold enhancement in N_{ice} for various values of F_{BR} and T_{min} within the breakup parameterization. Dots are also colored by $N_{INP}^{(lim)}$, where black indicates no 100-fold enhancement ever occurring. From panel **(a)** to **(b)** to **(c)**, the nucleation rate decreases by two orders of magnitude; note that the y-axis in panel **(c)** has a smaller range than the others. The bottom panels show the temporal evolution of N_{ice} for the various values of F_{BR} and T_{min} with $N_{INP}^{(tot)}$ of 0.167 L^{-1} (green traces) and 0.012 L^{-1} (yellow traces). This coloring by $N_{INP}^{(tot)}$ is similar to that in Figure 1a above. These parameter perturbations are run for u_z of 2 m s^{-1} and T_0 of 272 K.

increases N_{ice} by a factor of 200 when $N_{INP}^{(tot)}$ is 0.0129 L^{-1} and by only a factor of 3 when $N_{INP}^{(tot)}$ is 0.167 L^{-1}. Similarly, decreasing T_{min} from 258 to 246 K increases N_{ice} by a factor of 230 when $N_{INP}^{(tot)}$ is 0.0129 L^{-1} and by only a factor of 1.5 when $N_{INP}^{(tot)}$ is 0.167 L^{-1}. The parameters also mostly affect the enhancement magnitude not its timing.

We next consider variations in $p_{sh}^{(max)}$ and the functional form for the fragments generated from droplet shattering. We triple the leading coefficient F_{DS} and alter the diameter dependence from quartic to cubic within the Lawson et al. (2015) formulation. Then we use two sigmoids shown in Figure S3, which generate higher $\aleph_{DS}$ at small D_R and lower $\aleph_{DS}$ at large D_R relative to Lawson et al. (2015). As above, there is no meaningful $N_{INP}^{(lim)}$ here, so we focus on the maximum enhancement from these various cases, shown in Figure 6.

In panel a, by far the smallest enhancements occur for a D_R^3 dependence in $\aleph_{DS}$. Independent of $p_{sh}^{(max)}$ these simulations never produce an ICNC enhancement greater than about 50. Larger enhancements occur for the D_R^4 dependence in $\aleph_{DS}$ than for a sigmoidal dependence on D_R. Interestingly for the largest leading coefficient, F_{DS} of 7.5 x 10^{-11}, higher $p_{sh}^{(max)}$ does not monotonically increase enhancement. Another kind of 'sweet spot' exists here, and too rapid initial fragment generation may actually deplete cloud liquid faster and limit ultimate ice crystal generation (Beheng, 1987; Connolly et al., 2006b; Field et al., 2017). Elsewhere, increasing $p_{sh}^{(max)}$ does yield higher enhancement, up to about 2500 for the sigmoidal $\aleph_{DS}$ and an order of magnitude more for the default D_R^4 $\aleph_{DS}$.

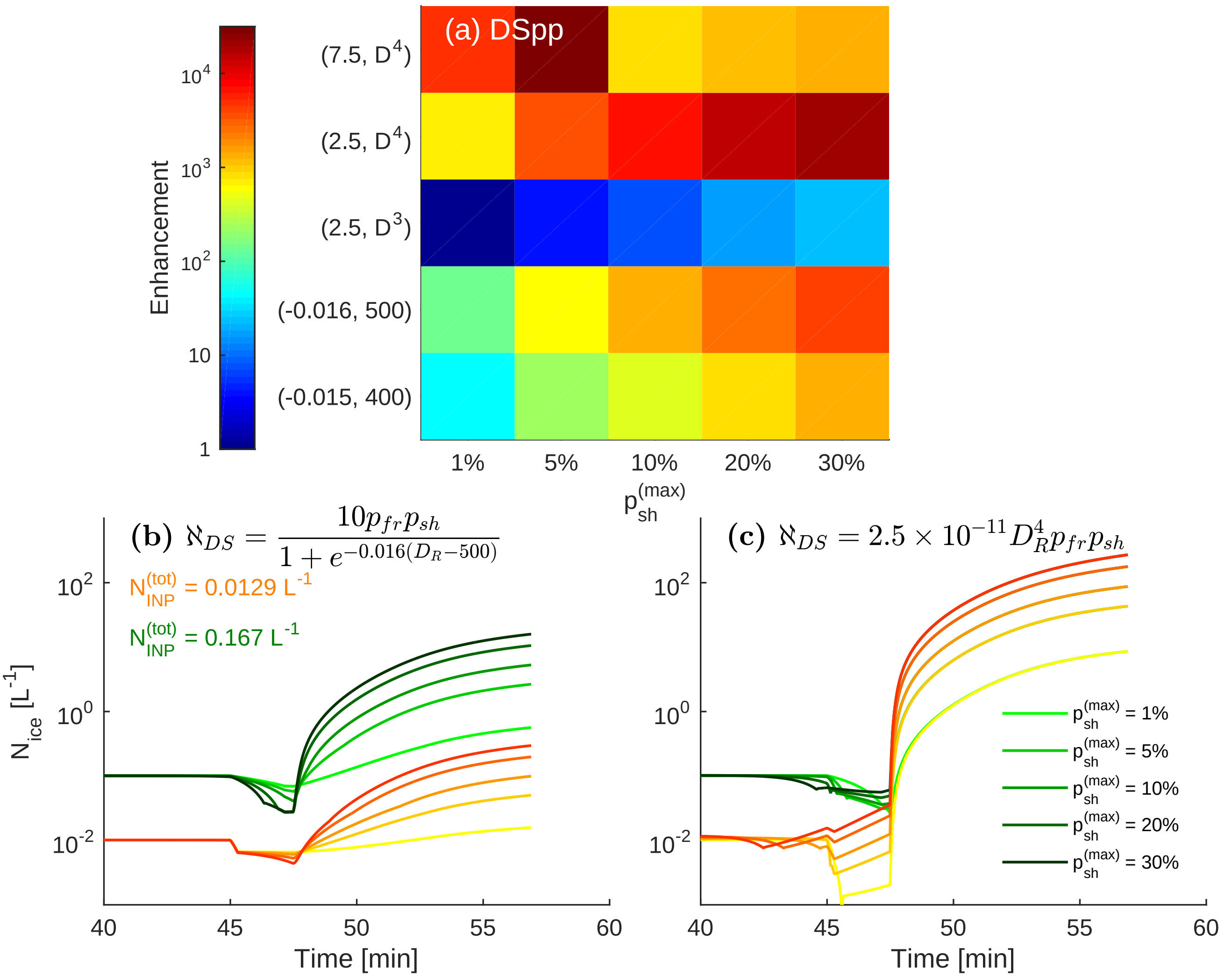

Figure 6. Results from the parameter perturbation simulations with droplet shattering. Panel **(a)** shows how the enhancement magnitude shifts with the various values of F_{DS} and p_{max}. Panels **(b)** and **(c)** show the temporal evolution of N_{ice} for the various values of F_{DS} and p_{max} with $N_{INP}^{(tot)}$ of 0.167 L^{-1} (green traces) and 0.012 L^{-1} (yellow traces). This coloring by $N_{INP}^{(tot)}$ is similar to that in Figure 1b above. These parameter perturbations are run for u_z of 2 m s^{-1} and T_0 of 272 K.

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Panels **(b)** and **(c)** show N_{ice} evolution for various values of $p_{sh}^{(max)}$ and the sigmoidal and default D_R^4 and $\aleph_{DS}$ forms respectively. The yellow traces show this evolution for $N_{INP}^{(tot)}$ of 0.0129 L^{-1} and the green for 0.167 L^{-1}, but these INP concentrations do not make a significant difference. Again it is clear that the D_R^4 dependence generates more ice crystals. And increasing $p_{sh}^{(max)}$ by a factor of 10 from 1 to 10% translates linearly to a factor 10 increase in $N_{ice}^{(max)}$.

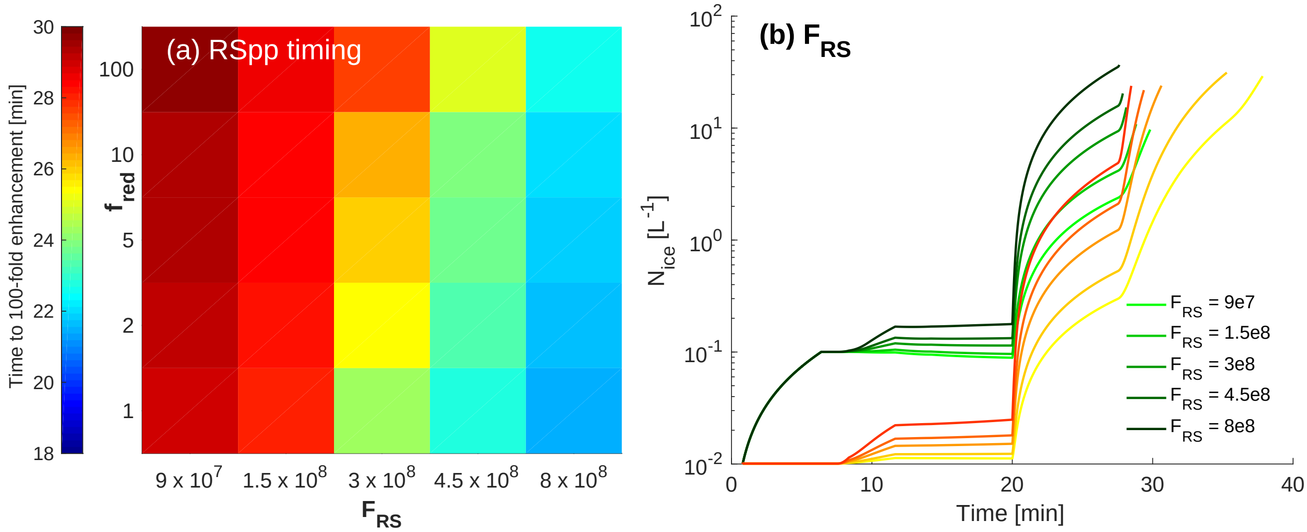

Figure 7. Results from the parameter perturbation simulations with rime splintering. Panel **(a)** shows how time of a 100-fold enhancement shifts with the fragment number per kilogram rime F_{RS} and the nucleation reduction rate f_{red}. Panel **(b)** shows the temporal evolution of N_{ice} for various values of F_{RS} with $N_{INP}^{(tot)}$ of 0.167 L^{-1} (green traces) and 0.012 L^{-1} (yellow traces). This coloring of $N_{INP}^{(tot)}$ is as in Figure 1c above. These parameter perturbations are run for u_z of 2 m s^{-1} and T_0 of 272 K.

Finally, Figure 7 shows the impact of fragment number per kilogram of rime, F_{RS}. Here we consider enhancement timing because the thermodynamic simulations show that there is no meaningful $N_{INP}^{(lim)}$ and the default ones show that the enhancement magnitude stays more or less constant. Panel **(a)** shows how the enhancement timing varies with the nucleation rate and fragment number F_{RS}. Slower nucleation rates are quantified by a reduction factor f_{red} on the y-axis. Along with lower F_{RS}, slower nucleation yields longer enhancement times, but only by about 8 minutes relative to the highest nucleation rate and F_{RS}. F_{RS} is also the more influential factor in timing, and its impact on N_{ice} evolution is shown in panel **(b)**.

4 Summary and Outlook

We have performed three sets of simulations with a six hydrometeor class parcel model, considering the effect of thermodynamics and parameter perturbations on $N_{INP}^{(lim)}$, as well as ICNC enhancement and timing. Our findings can be summarized in three points:

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1. *The evolution of N_{ice} from secondary production is determined by collision-based non-linearity and single versus two-phasedness.*

   N_{ice} increases gradually for the collision-based processes of breakup and rime splintering, whereas for non-collisional droplet shattering, N_{ice} increases significantly and suddenly, only when p_{fr} becomes large enough at cold enough temperatures. $N_{INP}^{(tot)}$ affects both the enhancement magnitude and timing for breakup. For rime splintering, $N_{INP}^{(tot)}$ affects timing to obtain a given $N_{ice}(t_{end})$, while for droplet shattering, it has almost no impact on either magnitude or timing.

2. *$N_{INP}^{(lim)}$ can be as large as 0.07 L^{-1} for breakup. Rime splintering or droplet shattering enhancement is determined by a thermodynamic 'sweet spot' rather than by $N_{INP}^{(lim)}$.*

   $N_{INP}^{(lim)}$ increases for breakup as the fragment number decreases or the temperature range shrinks, particularly for $N_{INP}^{(tot)}$ of 0.01 L^{-1} or less. At faster nucleation rates, the fragment number and temperature range are also more influential: enhancement occurs for 90% of the parameter space at a default nucleation rate, and just 10% of the space at a rate 100 times slower. These trends are visualized in the 'primary ice' panel of the summary schematic (Fig. 8).

   For rime splintering or droplet shattering, ICNC enhancements of 10^4 are possible even for slow nucleation rates and $N_{INP}^{(tot)}$ as low as 1 m^{-3}. For these processes involving the liquid phase, an intermediate updraft for which hydrometeors grow fast enough but also spend long enough in the appropriate temperature zone is more important. The cloud base temperature must also be warm enough, i.e., greater than 260 K in our simulations.

3. *When multiple secondary production processes are active, no single process dominates ICNC enhancement.*

   At higher nucleation rates, low u_z, and warm T_0, the contribution from breakup is large. If INP are limited, u_z is somewhat higher, or T_0 is somewhat colder, droplet shattering should be more important. Or if temperature falls in the optimal zone of 268 to 270 and u_z is intermediate, the rime splintering contribution will be large. A large p_{fr} for droplet shattering, however, throws off this balance. If p_{fr} is closer to unity, non-collisional droplet shattering dominates, as it depends on liquid hydrometeors only and has less stringent temperature dependence than rime splintering.

More generally, the role of ice-nucleating particles in secondary production reflects how changing emissions will affect cloud phase partitioning. The low or non-existent values of $N_{INP}^{(lim)}$ calculated in this study indicate that perturbations in CCN concentrations are more influential on mixed-phase partitioning than those in INP concentrations, with the caveat that thermodynamic conditions are appropriate for secondary production. If the mixed-phase cloud is polluted by more CCN, the higher droplet number will mean that fewer droplets reach a sufficient size to shatter or rime efficiently (This last factor has been called the riming indirect effect (Borys et al., 2003; Lance et al., 2011; Lohmann, 2017).) And in these cases, the supercooled liquid fraction remains higher, and the cloud reflects more shortwave radiation. More pollution by CCN could also yield a thermodynamic indirect effect in which latent heat is released at high altitudes and strengthens the upward movement

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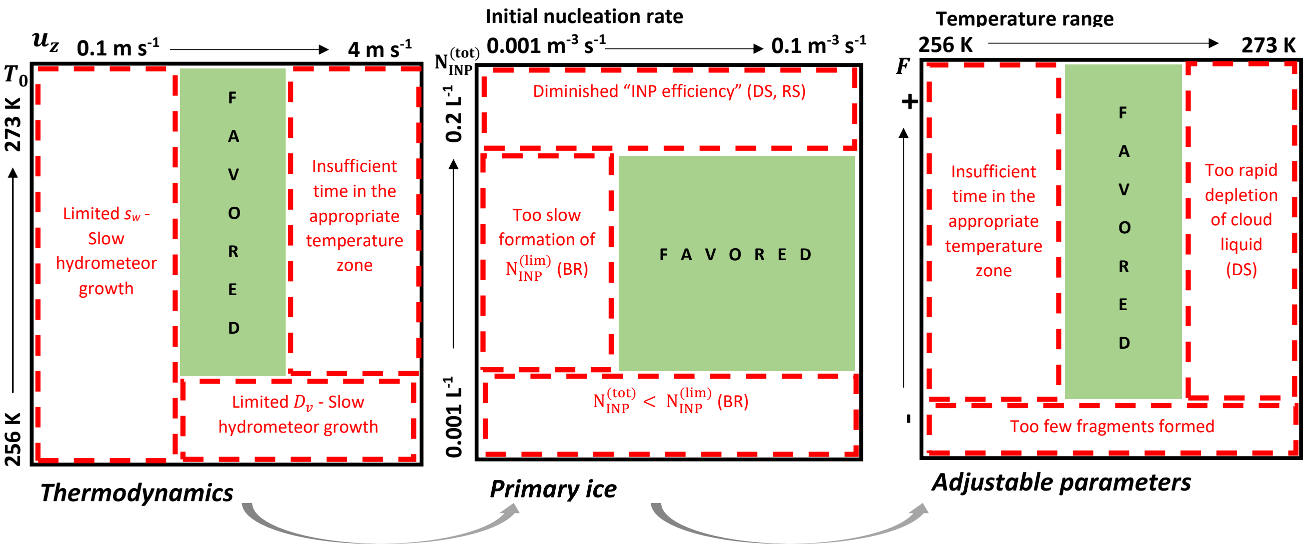

Figure 8. Summary of thermodynamic, primary ice, and adjustable parameter trends affecting ICNC enhancement from secondary production. F denotes the leading coefficient of a fragment number function for process X, $\aleph_X$. Regions in red indicate that secondary production may be limited, and those in green indicate that conditions are favorable. If the limitation is applicable only to one process, this is indicated in parentheses.

of the cloud; Koren et al. (2005) have called this cloud invigoration. Our simulations have shown that beyond a certain updraft, secondary production is no longer favored. In this way, the supercooled liquid fraction could also remain higher.

The impact of INP concentrations could be larger for deep convective clouds in which anvil spreading is caused by generation of many small crystals at cloud top (Fan et al., 2013). If the cloud is polluted by more INP, more vigorous secondary production by breakup may occur under conditions of fast enough nucleation rate but modest enough updraft and warm enough cloud base. These limited conditions can be found in deep convective clouds, along with other regions favorable for secondary production like the "mixing regions" at the edges of rising turrets or tops of eroding ones (Beard, 1992). In contrast to the riming or thermodynamic indirect effects mentioned above, an ICNC increase at the deep convective cloud top, a kind of 'anvil enhancement effect', would radiatively warm the surface.

A systematic quantification of $N_{INP}^{(lim)}$ is also relevant for the growing field of bioaerosol. Primary biological aerosol particles (PBAP) exist in the atmosphere at much lower number concentrations than dust or black carbon. But they also nucleate at warmer subzero temperatures (Hoose and Möhler, 2012; Fröhlich-Nowoisky et al., 2016), and small biological residues can intermix with dust particles to boost ice nucleation activity (O'Sullivan et al., 2015). Even when their contribution to primarily nucleated ICNC is small, they may remain influential via initiation of secondary ice production. For example, the ice active fraction of 10^{-4} for *Pseudomonas syringae* measured by Möhler et al. (2008) around -8°C could provide the 0.01 L^{-1} seed concentration from Crawford et al. (2012) for concentrations of 10^5 m^{-3}, although this is an upper bound for

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bioaerosol number. From our calculations, it could also provide the $N_{INP}^{(lim)}$ necessary for breakup to occur. Bioaerosol could also be sufficient to initiate rime splintering, given that this process occurs even for N_{INP} below 1 m^{-3} in our simulations. A climatically important linkage has also been hypothesized between PBAP, in-cloud ICNC, and cold phase-initiated rain and is often termed the 'bioprecipitation feedback' (Huffman et al., 2013; Morris et al., 2014). The possibility of secondary production with a low $N_{INP}^{(lim)}$ means that even a few bioaerosol could trigger generation of many small ice hydrometeors from larger droplets or graupel and suppress precipitation.

As a summary of our findings, we present an organizational framework for future studies of secondary production in Figure 8. Favorable conditions for large ICNC enhancements are shown in green, e.g., warm cloud base and intermediate updraft in the thermodynamic panel or higher nucleation rate for breakup in the primary ice panel. This classification can be used to determine where, within in-situ or remote sensing data, signatures of secondary production are likely to be found. And as more experimental studies to quantify the fragment number and temperature dependencies of these processes are done, more quantitative bounds can be established in the final adjustable parameter panel.

Code availability. No data was used in producing this manuscript. Various model version codes are available upon request.

Appendix: Notation

a_X Spheroidal axis of hydrometeor of type X

β Adjustable parameter in the sigmoidal function for the fragment number generated from shattering

c_0 Primary ice nucleation rate based upon DeMott et al. (2010)

$\mathcal{D}_v$ Diffusion coefficient of water vapor

F_{BR} Leading coefficient of the fragment number generated per collision based upon data from Takahashi et al. (1995)

F_{DS} Leading coefficient of the fragment number generated per shattering droplet as in Lawson et al. (2015)

f_{red} Factor for nucleation rate reduction

F_{RS} Fragment number per kilogram of rime as in Hallett and Mossop (1974)

γ Adjustable parameter in the sigmoidal function for the fragment number generated from shattering

ICNC In-cloud ice crystal number concentration

INP Ice-nucleating particle

K_X Gravitational collection kernel for process X

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$\aleph_{BR}$ Fragment number from breakup upon collision per large and small graupel number

N_d Small droplet number concentration in the parcel

$\aleph_{DS}$ Fragment number from droplet shattering per large droplet number

$\aleph_{DS}^{(coll)}$ Fragment number from collisional droplet shattering per large droplet and small ice crystal number

N_i Ice crystal number concentration in the parcel

N_{ice} Total ice hydrometeor number within the parcel, i.e. the summation of ice crystal, small and large graupel numbers

$N_{ice}^{(max)}$ Maximum N_{ice} formed within the parcel during a given simulation

$N_{INP}^{(lim)}$ Limiting ice nucleating particle number concentration to initiate secondary production

$N_{INP}^{(tot)}$ Total number of ice nucleating particles within the parcel available for primary nucleation. This value is fixed by the user beforehand.

N_g Small graupel number concentration in the parcel

N_G Large graupel number concentration in the parcel

N_r Medium droplet number concentration in the parcel

N_R Large droplet number concentration in the parcel

$\aleph_{RS}$ Fragment number from rime splintering per large droplet and large or small graupel number

ρ_w Density of liquid water

$p_{fr}(t,T,D)$ Temperature-, time-, and size-dependent probability that a large droplet freezes as in Bigg (1953)

$p_{sh}(T)$ Temperature-dependent probability that a frozen large droplet shatters with $p_{sh}^{(max)}$ being the maximum of this distribution

r_X Radius of hydrometeor of type X

s_w Supersaturation with respect to liquid water in the parcel

T_0 Cloud base temperature or the initial temperature of the parcel

t_{end} Time when the simulation is terminated, either because the parcel has become water subsaturated or the temperature has reached 237 K where homogeneous nucleation can occur

T_{min} Minimum temperature for breakup upon collision to occur

u_z Updraft velocity of the parcel

Competing interests. The authors declare no competing interests.

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