Responses to Reviewer 1 Comments

I will start this review by confessing that I am an observationalist, not a modeler. I bring an obvious bias into this review, which is that I couch my evaluation of this work in terms of data collected in clouds, not numerical simulations of clouds. My main concern with this manuscript is that I cannot determine how this parcel model relates to an updraft in a cloud. Presumably, a parcel model is intended to represent the evolution of an undiluted parcel of cloud as it rises in the atmosphere. However, it is not clear what the prognostic microphysical variables are in the model. Presumably the model predicts N_{ice} for each of the categories because this is shown in the "ice generation function" equation, but what about mass? It's not even clearly stated if it's a bulk or bin microphysics scheme. It's also not clear what the "ice generation function" itself is, and what the units of G_{ice} are. I'm assuming this is dN_{ice}/dt from all microphysical processes, but it's not clear. The Sullivan (2017 – JGR) reference is in review and of no help. Even if the Sullivan JGR paper becomes available, at a minimum the manuscript should state what the model predicted variables are, and how they are being solved in the model numerically (e.g., what kind of time stepping method, the time step, etc.). It would also help if the manuscript gave the evolution equations for the model predicted variables.

Thank you for your careful reading and feedback. We initially limited discussion of model development, given that another manuscript treats those details thoroughly. We understand that this manuscript was not yet available upon your first reading, but it has now been accepted (doi:10.1002/2017JD026546). We have also expanded Section 2 to clarify the model description. To more generally make our results in each section relatable to observations, we include the table shown at the end of these responses in our summary and outlook section.

First, the prognostic microphysical variables are the hydrometeor number in each of the six classes. We rewrite: "*The model predicts the number in these six classes, denoted N_i, N_d, N_g, N_G, N_r, and N_R respectively.*" There is not an N_{ice} "for each of the categories", as you state. Rather, N_{ice} is the summation of the number in the three ice hydrometeor classes. This is stated in the Notation Appendix, and we repeat it for clarity early on in Section 2.

Then, there are no size distributions for the hydrometeors in each class. They are assumed monodisperse but their radii or major axes are evolved over time. To Section 2, we add, "*The hydrometeors in each class are assumed monodisperse, but their sizes are tracked over time as a function of temperature and supersaturation*." To emphasize that this is a bin microphysics scheme, with six hydrometeor classes, we write "*The **bin** microphysics consists of primary nucleation and …*"

In order to clarify the ice generation function G_{ice}, we have rewritten Equation 1:

$$G_{ice} = \left.\frac{dN_i}{dt}\right|_{NUC} + \left.\frac{dN_i}{dt}\right|_{BR} + \left.\frac{dN_i}{dt}\right|_{RS} + \left.\frac{dN_i}{dt}\right|_{DS} \quad (1)$$

$$= c_0 H(t) + \eta_{BR} K_{BR} \aleph_{BR} N_g N_G + \eta_{RS} \aleph_{RS} \left[K_{RS,g} N_g + K_{RS,G} N_G \right] N_R + \eta_{DS} \aleph_{DS} N_R \quad (2)$$

The ice generation function includes all sources of ice crystals, i.e., production of ice crystals from primary nucleation, collisional breakup, rime splintering, and frozen droplet shattering. We write this before Equations 1 and 2, along with the fact that the ice generation function has "*units of m^{-3} s^{-1}*", i.e., it is a number concentration of crystals generated per time. Then we have added an equation for the number balance looks in the ice crystal hydrometeor class:

$$\frac{dN_i}{dt} = G_{ice}(t) - G_{ice}(t - \tau_i) - \eta_{agg} K_{agg} N_i N_g$$

We hope that these adjustments clarify the relationship between the tendencies, generation function, and number balances: the source tendencies make up the generation function, and the generation function at different times makes up the number balance.

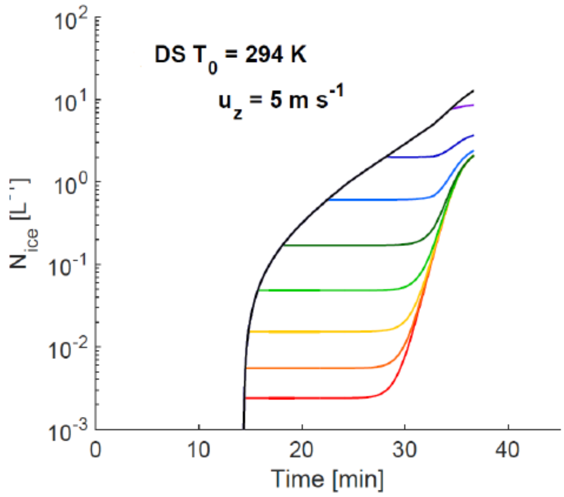
Finally we add the following description of the numerical solutions used: "*The six hydrometeor number tendencies are solved with an explicit Runge-Kutta (2,3) pair for delay differential equations [Bogacki 1989] and coupled to moist thermodynamic equations for pressure, temperature, supersaturation, mixing ratios, and hydrometeor sizes. This second set of equations is solved with a Rosenbrock formulat of order 2 [Rosenbrock 1963].*" We address the concern about ice mass below.

The manuscript shows no drop or ice particle size distriutions and no liquid water or ice water contents as a function of temperature. Also, the observations that I am most familiar with suggest that clouds with cloud-base temperatures colder or equal to 0 C, which are all of the cases examined here, do not produce cloud drops large enough to support drop shattering, and generally not even large enough to support rime splintering. Large drops (drizzle and rain drops) are what the literature (e.g., Koenig 1963, 1965; Hobbs and Rangno 1990, Rangno 2008, Lawson et al. 2015) associates with drop shattering and rapid glaciation. The data suggest that the formation of millimeter-diameter supercooled drops requires cloud base temperatures warmer than approximately +18 C (291 K) and broad (> 50 um diameter) cloud base drop distributions. Albeit, the requisite relationship between CCN and cloud base temperature is yet to be accurately quantified. Also, the coalescence process is key to the formation of supercooled large drops. Nowhere in the manuscript can I find how coalescence is handled in the model (except that K_X is a gravitational collection kernel in Eq. 1). One aspect of the simulations that does appear to be consistent with the observations is that rime-splintering takes place only in clouds with very weak updrafts (e.g., Heymsfield and Willis 2014). However, it is not clear in the manuscript exactly why this takes place in the simulations. Before I can recommend publication, the manuscript needs to provide an explicit description of the model, and the evolution of the parcel in terms of microphysical parameters (liquid and water size distributions, LWC, IWC as a function of temperature). I understand that this may be a bit artificial given the six categories of particles, but an attempt must be made, and the results should be compared with observations.

There are no drop or ice particle size distributions shown because the hydrometeors are assumed monodisperse in each class, and we have added this explicitly to the manuscript. For the coalescence formulation, we use a gravitational sweep-out kernel, as you note, and assume that the terminal velocity of the (small) collected droplets is negligible relative to that of the (medium) collector droplets. Then the coalescence efficiency between small and medium droplets is assumed to be unity, and that between droplets of the same size is assumed to be negligible. These assumptions are based upon the measurements of Klett and Davis 1973. We include these detail: "*The coalescence efficiency is assumed to be unity between small and medium droplets and negligible between two droplets of the same size*."

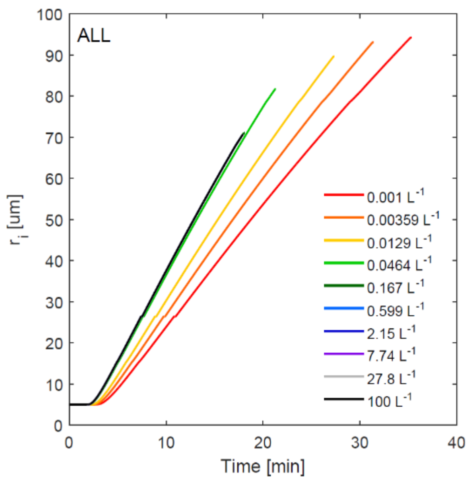
Your point about the cloud base temperatures is a good one. Indeed, more recent in-situ measurements where droplet shattering was thought to occur cite warmer cloud base temperatures. Given our choice of a less steep CCN spectrum (yielding fewer droplets) and a stronger updraft, large droplet formation should still reasonably occur for colder initial temperatures. As noted in Taylor et al. 2016, "the temperature at which large concentrations of drizzle and raindrops form depends [not only] on the cloud base temperature [but also on] cloud-drop number concentration and time-dependent factors such as updraft speed."

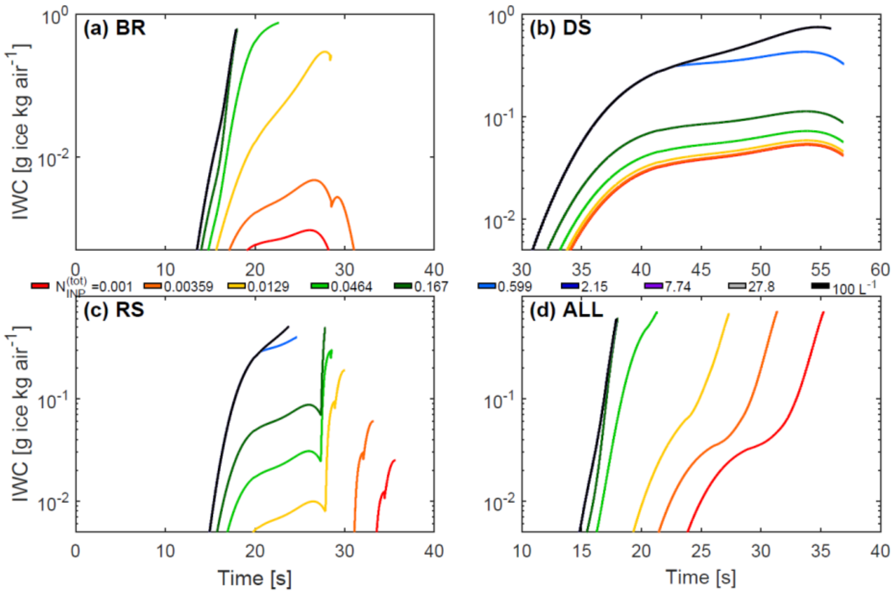
There are also measurements that indicate that the droplets need not be on the order of a millimeter to shatter effectively. Laboratory droplet levitation experiments shown in Leisner et al. 2014 indicate that shattering can occur even for droplets of diameter of 80 um. Lawson et al. 2015 note that droplets about 200 m above the cloud base, at temperatures where freezing and shattering could begin, have a diameter of 90 um. Nevertheless, we run two sensitivity tests initiated from T_0 of 294 K and with a more convective updraft of 5 m s^{-1} and include their N_{ice} evolution in the supplementary material:



In Section 3.1, we note this sensitivity test: "*In more recent measurements with evidence of droplet shattering, the cloud base temperature has been warmer, and the updraft stronger, than the default conditions in Table S1, e.g., Lawson et al. 2015, Taylor et al. 2016 (see Table 2). We show the N_{ice} evolution from a `warm-base-convective' sensitivity run in Figure S7. Here the same threshold behavior occurs once the parcel reaches cold enough temperatures for droplet freezing, but there is no N_{ice} decrease beforehand because ice nucleation begins later, and no graupel has begun to fall out.*" And to a paragraph early on in Section 3.2 on the thermodynamic simulations, we add that "*These cloud base temperatures are colder than those associated with most in-situ measurements of frozen droplet shattering [Lawson et al. 2015, Taylor et al. 2016]; however, our simulations still produce droplets of sufficient diameter to shatter, O (100 um), and a 'warm-base-convective sensitivity run is shown in Figure S7.*"

Finally, we have shown ice crystal numbers because "enhancement" from secondary ice production is generally discussed in the literature in terms of the orders-of-magnitude discrepancy between ice crystal and ice-nucleating particle numbers. But the mass is also important to consider, as you suggest. To the supplemental information, we add the two figures below, ice mass mixing ratio evolution for the default simulations and a sample ice crystal radius evolution. *At the top of Section , we state: The ice mass mixing ratio and ice crystal radius evolution are also shown in Figures S4 and S5, but analysis focuses on N_{ice} below.*"





SOME SPECIFIC COMMENTS

Following are some specific comments. Until the major comments are addressed, I am not willing to go through the manuscript with a fine-toothed comb, as I assume that the paper will be significantly modified. The comments below are intended to give the authors some idea of the type of modifications that are needed.

p. 1 Line 4: "Break Up" is not a good term for ice-ice collisions, because drops also break up. I suggest that you find a more descriptive term that applies only to ice. If the term "break up" has to be defined as ice-ice collisions here and everywhere else in the manuscript.
"*Breakup*" was used because preexisting work on this process generally employs this term, e.g., Yano and Phillips *JAS* 2001, Phillips et al. *JAS* 2017, Field et al. *Meteor. Mono.* 2017. But we understand that this terminology may cause confusion with droplet breakup. We have gone through and changed all instances of "*breakup*" to "*collisional breakup*".

p. 2, Line 7: Add references; there are several.
We have added Scott and Hobbs 1977, Phillips et al. 2001, and Fridlind et al. 2007 to the citations for frozen droplet shattering.

p. 2, Lines 16-17: This is contradictory. In the previous sentence, you reference Field as reporting many uncertainties in the physics of secondary ice production, and then go on to state that small-scale models provide a good tool to estimate variability in secondary-produced ice. The model is only as good as the physics it contains. With the acknowledged vast degree of uncertainties, how can one have any confidence in the model results? If the model results are to be useful, then the physical uncertainties have to be emphasized. Also, sensitivity tests should be run to show how the physical uncertainties impact the results. At a minimum, a disclaimer of this sort needs to be inserted at this point in the manuscript.
We do not believe that these statements are contradictory. Investigating how a given output varies with uncertain parameters is an important application of models. And particularly for small-scale, more controllable models, output variation with adjustable parameters can be well-understood. This kind of work allows experimentalists to focus on measuring the most influential parameters and provides a test-bed for parameterizations prior to implementation in large-scale models. This utility of small-scale models is summarized in the IPCC Assessment Report 5: "high-resolution models enhance our

understanding of cloud processes [as] an important tool in testing and improving parameterizations of cloud-controlling processes."

As you note, sensitivity tests should be run with the small-scale model to understand the process and parametric uncertainties. Sections 3.1.1 and 3.3 contain these tests. We run simulations for different formulations of the physics of frozen droplet shattering. And then we investigate the sensitivity to adjustable parameters in the fragment generation functions (particularly F_{BR}, T_{min}, sigmoid versus polynomial forms for droplet shattering, and $p_{sh}^{(max)}$).

We clarify the utility of small-scale models in this paragraph: "*Laboratory and in-situ data of these processes are difficult to obtain, and their fragment generation functions and temperature dependence remain uncertain [Field et al. 2017]. Given these uncertainties, implementation of secondary ice production parameterization in large-scale models would be premature. Instead, small-scale, more controllable models provide a means of estimating variability in output secondarily-produced ICNC with these parameters, as well as the minimum number of INP needed to initiate secondary production.*"

<u>p. 3, Eqn (1) and discussion</u>: Eqn (1) is far too arcane to understand what is going on in the model. The reference to Sullivan et al. (2017) is of no use since it is under review. There are several unanswered questions. What are the units of G_{ice}? What is the role of coalescence and how is it handled? What is the cloud base drop distribution? Are CCN included? If so, how? Why don't small ice and small drops appear in Eqn (1)? Also, the number of secondary ice particles produced is only one issue. The mass of ice is of equal if not more importance. If large (millimeter-diameter) supercooled drops are rapidly freezing, as seen in the observations, then the *conversion* of water to ice (and eventually back to water in the form of rain), is more significant than the number of ice particles. Show the results also in terms of water and ice mass.
As described above, we have worked to make the model description more clear without restating what has already been published in the model development manuscript. In particular, we have more clearly stated the purpose and the units of the ice generation function and expanded its mathematical explanation with two additional equations. Small ice and droplets do not appear in Equation 1 because they play no role in any of the processes that are a source of small ice crystals.

Then we have emphasized that there are no size distributions involved; the monodisperse radius or axis of each hydrometeor class is evolved in time. The model contains no explicit aerosol. We add this statement and an in-line equation for primary nucleation before the statement that "*the droplet generation function consists simply of droplet activation, calculated from a Twomey power-law formulation*." So droplet number is calculated from supersaturation rather than a CCN number. Then we have added more detail for the coalescence formulation to Section 2, as discussed in the response to your major comments. And additional supplemental figures now show the ice mass mixing ratio for all default simulations, as well as the ice crystal radius evolution.

<u>p. 3, line 27</u>: 237 K is not the homogeneous freezing temperature of pure water. The generally accepted value in the literature is 235.15 K. The AMS Glossary of Meteorology states that homogeneous nucleation occurs near 233.15 K.
Thank you for pointing this out. We write "*or a reaches a temperature of 237 K above which no homogeneous nucleation occurs.*"

<u>p. 7, Fig. 2 Captions</u>: How were the values of 2 and 10 fragments per drop chosen? How is the dependence on drop size handled?
Two was chosen as the minimum number of fragments into which a droplet could fragment. Ten was chosen as an upper bound because it represents an order of magnitude increase upon each fragmentation. In what was formerly Equation 2 (now Equation 4), $\aleph_{DS}^{(coll)}$ contains the droplet size

dependence: $\aleph_{DS}^{(coll)} = F_{DS} (2r_R)^4 p_{sh} (T)$. So the fragment number is quartic in droplet size, as in Lawson et al. 2015. This equation was also given in Table S1.

p. 10, Line 17: Lawson et al. 2015 explicitly state that rime splintering is not responsible for the observed secondary ice process. Delete this reference.
Yes, thank you for catching this. Lawson et al. 2015 did emphasize the importance of the liquid phase to secondary ice production, but not to secondary ice production from rime splintering.

p. 11, Lines 4-7: What are the justifications for these assumptions and modifications?
Droplet levitation experiments at the Karlsruhe Institute of Technology are the basis for these modifications to the fragment generation function. In particular, these experiments indicate that the Lawson et al. parameterizations underestimates the fragment number generated for smaller droplets (D ~ 100 um) and overestimates the number for larger droplets (D ~ 1 mm). The sigmoid function addresses both of these concerns. Changing the exponent in the polynomial form addresses a potential overestimation for larger droplets only.

In Table S1, where we give the explicit functional forms of these modified fragment generation functions, we cite “Droplet levitation experiments”, but we also point this out in the text now.

p. 12, Lines 1-5: The production of ice in this scenario may be of some interest, but of more interest to cloud physicists is how the ice and water mass budgets evolve. Please show these.

p. 14, Line 5: This is the first mention of CCN. Were CCN used in the model, and if so, how?
To the statement that *“the droplet generation function consists of droplet activation, calculated from a Twomey power-law formulation*”, we have added in Section 2 that “*droplet number is calculated solely from supersaturation rather than a CCN number*” because aerosol is not treated explicitly in our framework.

p. 16, Line 8: “warm cloud base”. All cloud bases cited in the paper < 273 K, so there are no warm cloud bases.
Yes, accurate wording here would be “***warmer** cloud base”*, i.e., those parcels that are initiated from relatively warmer subzero temperatures. We have changed this to “*warmer subzero cloud base temperatures*” in a few places.

Table 2. Comparison of parcel model results in each section with results from in-situ and laboratory measurements not used to constrain the model formulations.

	In-situ measurements	***Laboratory studies***	***Parcel model simulations***
Temporal evolution of N_i	**BR** and **DS**: 20 min to form drizzle drops and 12-15 min to glaciation after first ice (Taylor et al., 2016); **DS**: 2-3 min to glaciation after first ice (Lawson et al., 2015); **RS**: 10^2 enhancements within 10-15 min (Hobbs and Rangno, 1990), 8 L^{-1} over 32 min (Heymsfield and Willis, 2014)	**BR**: 20 min to increase ICNC by a factor of 10 with initial ICNC of 3 L^{-1} (Vardiman, 1978) (his Fig. 7); **DS**: only 50 seconds to fragmentation after equilibration and nucleation time (Johnson and Hallett, 1968); **RS**: Linear increase starting between 10 and 20 min (Hallett and Mossop, 1974) (their Fig. 1)	**BR**: Superexponential increase based on $N_{\mathrm{INP}}^{(tot)}$; **DS**: threshold increase based on p_{fr}; **DScoll** Exponential increase based on N_R; **RS**: Superexponential increase based on N_R
Limiting INP or thermodynamics	**BR**: T_{top} between -10° and -18°C with N_{ice} from 0.1 to 5 L^{-1} (Rangno and Hobbs, 2001); **DS** and **RS**: Taylor et al. (2016) cite the importance of the warm rain process through T_0, CDNC, u_z, and cell lifetime; **DS**: N_{INP} of 10^{-4} to 10^{-2} L^{-1} for $\overline{N_i}$ of 572 L^{-1} (Lawson et al., 2015); **RS**: $N_{\mathrm{INP}}^{(lim)}$ of 0.01 L^{-1} (Crawford et al., 2012)	**BR**: Strong modulation of ultimate ICNC by initial ICNC (Vardiman, 1978) (his Figure 1); **DS**: $N_{\mathrm{INP}}^{(lim)}$ of 1 m^{-3} (Beard, 1992), Favorable temperatures colder than those for RS (Korolev et al., 2004); **RS**: optimal temperatures between -3 and -8°C (Hallett and Mossop, 1974), modest updrafts are most favorable (Mossop, 1985; Heymsfield and Willis, 2014)	**BR**: $N_{\mathrm{INP}}^{(lim)}$ from 2 up to 70 m^{-3}, possible only at warmer T_0 and slower u_z; **DS**: no meaningful $N_{\mathrm{INP}}^{(lim)}$, favored at colder T_0 down to 258 K as u_z slows; **RS**: no meaningful $N_{\mathrm{INP}}^{(lim)}$, favored for 268-270 K but this range widens as u_z increases
Parametric uncertainty	**BR**: In-cloud graupel collision rate of 1 m^{-3} s^{-1} (Mizuno and Matsuo, 1992), 10% of ice particles were fragmented (Rangno and Hobbs, 2001); **DS**: 10^{-9} fragments per kg liquid (Lawson et al., 2015), 10% of drops frozen by -6°C (Brownscombe and Thorndike, 1968); **RS**: 1.4 L^{-1} s^{-1} (Taylor et al., 2016), 50 crystals s^{-1} (Heymsfield and Willis, 2014)	**BR**: Fragment generation rate K_0 of 0.00081 up to 0.01 L^{-1} s^{-1} (Vardiman, 1978); **DS**: Shattering frequencies of 10 to 37% between 50 and 120 μm (Brownscombe and Thorndike, 1968; Takahashi, 1976); **RS**: 250-700 splinters per mg rime at u_z = 1.5 m s^{-1}, 200-400 at 2 m s^{-1} (Hallett and Mossop, 1974) (their Fig. 3), 90-350 (Mossop, 1985)	**BR**: 10^2-fold enhancement increasing F_{BR} from 40 to 280 at $N_{\mathrm{INP}}^{(tot)}$ of 0.17 L^{-1}; **DS**: 10-fold enhancement increasing $p_{sh}^{(max)}$ from 1 to 30% independent of $N_{\mathrm{INP}}^{(tot)}$; **RS**: 10 minute sooner enhancement increasing F_{RS} from 3 x 10^8 to 3 x 10^9 for all $N_{\mathrm{INP}}^{(tot)}$