Responses to Reviewer 2 Comments

The manuscript investigates the role of three secondary ice production mechanisms (rime splintering, frozen droplet shattering, and breakup), more specifically the evolution of the total ice number concentration depending on secondary ice production, the thermodynamic limitations on the secondary processes and the dependence on the chosen parameterization. The authors found that the evolution of the total ice number concentration is determined by the involvement of two phases and the non-linearity of the collision process. However, in case all processes are active none of them is dominant over the others. They also found that only breakup needs a minimum number of ice nuclei, all other processes are more sensitive to the number of cloud condensation nuclei and thermodynamic conditions. The results are summarized in Fig. 8 where they show in which thermodynamic region and also for which ice nuclei concentration and rates secondary ice formation is favorable. The manuscript adds some interesting aspects to the question what bridges the gap between ice nuclei and ice crystal measurements.

We thank the reviewer for the careful reading and thoughtful feedback.

Major Comments

The simulations show some interesting aspects of secondary ice production. However, the interpretation aspect could be stronger emphasized. What do the findings have for consequences in terms of modeling of mixed-phase clouds? How are the results connected to field observations? Do the findings agree with observations? Do the findings make sense in the general context / understanding of the microphysics of a mixed-phase cloud? Which further aspects would need to be investigated?

We have added a section before the final summary and outlook called "Comparison with experimental studies". It includes the table at the end of these responses to link field / laboratory measurements with our results in each section. The text in this section also more thoroughly addresses these interpretation questions:

"In Table 2, the parcel model results from the previous three sections are compared to those from field and laboratory measurements for each process. Similar time frames for enhancement, O(30 minutes), and favorable modest updrafts and warmer cloud base temperatures are present in both the observations and simulations. The same importance of N_{INP}^{Iim} to collisional breakup has been shown in other studies (Vardiman 1978).

An important limitation of the parcel model is the assumption of monodispersity. Large droplets shatter more effectively, and large graupel will have a larger sweep-out kernel for collisional breakup. Without the tails of a hydrometeor size distribution, these larger hydrometeors are omitted, and secondary production is underestimated. An ongoing study will implement similar formulations into a mesoscale meteorological model to understand the effect of this assumption. Ventilation effects, spatial phase separation, and continuous sedimentation are other, more advanced features of a real-world parcel that could also alter these parcel model results [Sullivan et al 2017]. For example, droplet or ice hydrometeor growth will be enhanced by the stronger vapor density gradient generated by their incloud motion. Again, omitting additional hydrometeor growth will underestimate secondary production. If `pockets' of ice phase exist within mixed-phase cloud, then the values of N_{INP}^{Iim} will be more influential as the collisional breakup contribution will increase relative to rime splintering or droplet shattering. If a continuous formulation of sedimentation were substituted for the threshold one used here, the largest enhancements in Figures 3 and 4 should shift to higher updrafts. Large hydrometeors would be held aloft by these higher updrafts and feed into the secondary production tendencies. The parcel model could be extended in future studies to investigate these effects." In regard to future aspects to consider, we also note at the end of Section 3.1, that "future work should also incorporate a dependence of p_{fr} on the number of submerged INP, rather than just on time and temperature."

Specific Comments

- Page 2, line 23-26: Could you not calculate the number of INP from the nucleation rates? Yes, you're correct. The absolute INP number could be calculated from the adjusted primary nucleation rates and the simulation duration. However, investigating the effect of primary nucleation rate on secondary production is still different than investigating the effect of the absolute INP number on secondary production. The wording about the Connolly et al. 2016a study is adjusted: "Connolly et al. 2006a found that rime splintering production increased with increasing primary nucleation rate but did not give an estimate of a threshold N_{INP} for this rime splintering to initiate."
- Page 3, line 4: How do you derive a nucleation rate from the INP concentration given in DeMott et al. 2010?

Some details of the model construction were omitted because they are covered extensively in another *Journal of Geophysical Research* manuscript (doi:10.1002/2017JD026546). The manuscript recently became available, and we have included more details in the revised manuscript. In particular, here, we note: "*More specifically, the nucleation rate is calculated as the product of updraft velocity, an assumed lapse rate of 6 K km⁻¹, and the temperature derivative of the INP concentration:* $u_z \Gamma d/dT [a_1 exp [a_2 (T - a_3)]$."

- Page 3, line 4: Why a Heaviside function? The Heaviside function is used to "transfer" hydrometeors that have had sufficient time for depositional and riming growth to the next highest size bin. But this time delay Heaviside is applied later to all terms in the generation function. So Equation 1 is corrected by omitting the *H*(*t*).
- Page 3, line 9: How is the connection between *DS* and *DScoll*? From the description (*DScoll* = collision between large droplet and ice crystals?) it sounds like two different processes. Add more explanation to this point.

DS refers to the shattering of a droplet as it begins to freeze because of a submerged ice nucleating particle. *DScoll* refers to the shattering of a droplet after collision with an ice crystal. Indeed, these could be considered two different processes that act simultaneously. For now and for comparison's sake, we have only allowed one or the other to be active and label both as "droplet shattering" since that is the mechanism (just induced by different phenomena).

The following clarification is added to the manuscript: "Later, droplet shattering is induced by collision with an ice crystal (denoted DScoll), rather than by internal freezing on a submerged INP. In this case, the tendency includes a product of large droplet and ice crystal numbers."

- Page 3, line 9: Why is it 1% and not 0% outside of the temperature range? This is done to account for uncertainty about whether this optimal temperature range of -3 to -8°C is for the rimer surface temperature or ambient cloud temperature (e.g., Heymsfield and Mossop 1984). We assume that even when the ambient cloud temperature is lower, there may be locally warm enough regions on the non-leading edge of the hydrometeor for rime splintering to occur.
- Page 5, lines 3-4: Explain that more explicitly, example? This section is organized so that this initial idea, *"structure ... can be understood by considering whether the process is collisional and whether it involves ... one or both phases"*, is explained in the

proceeding paragraphs. To make this organization clearer, the following changes are made to the topic sentences:

"When the process involves a product of hydrometeor numbers, as for breakup and rime splintering, the N_{ice} evolution is non-linear."

"When the process involves a single hydrometeor number, as for this case of droplet shattering, the N_{ice} evolution is linear and does not grow steadily. Instead it exhibits threshold behavior when the temperature becomes cold enough for a non-negligible freezing probability according to Bigg 1953."

"Because breakup and rime splintering involve the ice phase, increasing N_{INP}^{tot} boosts their rates and yields large enhancements sooner."

"Finally non-linearity and hydrometeor phases involved determine enhancement timing."

• Page 7, Equation 2: What is the physical concept or idea behind this formula or approach? This equation is a reformulation of the ice crystal generation from droplet shattering, i.e., a replacement for the final term in Equation 1. It can then be interpreted like the other terms in Equation 1, where K_{DS} is a gravitational collection kernel, \aleph is the generated fragment number, and N_R and N_i are large droplet and small ice crystal numbers respectively. It is exactly analogous to the formulation of the rime splintering terms, but with N_i rather than N_g or N_G . To make this connection clearer, we expand Equation 1 as follows:

$$G_{ice} = \frac{dN_i}{dt}\Big|_{NUC} + \frac{dN_i}{dt}\Big|_{BR} + \frac{dN_i}{dt}\Big|_{RS} + \frac{dN_i}{dt}\Big|_{RS}$$
(1)
$$= c_0 H(t) + \eta_{BR} K_{BR} \aleph_{BR} N_g N_G + \eta_{RS} \aleph_{RS} \Big[K_{RS,g} N_g + K_{RS,G} N_G\Big] N_R + \eta_{DS} \aleph_{DS} N_R$$
(2)

(This adjustment makes the collisional droplet shattering tendency Equation 3.) Before the equation for collisional droplet shattering, we add

"the droplet shattering tendency is adjusted to be proportional to both N_R and N_i , rather than just N_R as in the final term of Equation 2:"

- Page 9, line 2: Where do I see that in the figure? "For droplet shattering and rime splintering, there is no N_{INP}^{Iim} value greater than $1 m^{-3}$ " because from the lowest value of N_{INP}^{tot} on the y-axis of Figures 3 and 4 (= 1 m⁻³), there is a color corresponding to a 100-fold ice crystal number enhancement or higher. Large enhancements are occurring from these processes even at low INP numbers. To the text, we add "there is no N_{INP}^{Iim} value greater than $1 m^{-3}$: the enhancement is largest at the lowest value of N_{INP}^{tot} in Figure 3 and decreases with higher values of N_{INP}^{tot} ."
- Page 9, line 9: Again where and how do I see that in the figure? Yes, thank you for pointing out that this result (*"breakup remains the only process with a defined N_{INP}^{lim}"*) is not immediately clear from the figure. We add the following: *"At the lowest values of N_{INP}^{tot} for the droplet shattering and rime splintering panels, enhancements are large and decrease with increasing N_{INP}^{tot}. Only for breakup does N_{INP}^{tot} need to surpass a threshold before a large enhancement occurs."*
- Page 10, line 14-17: What is the reason for these differences?

In fact, we want to mention that previous studies *agree* with our results here. The Connolly et al. study produced a decrease in ice production from freezing rain when primary nucleation was more efficient, and we also see a decrease in production from droplet shattering with additional nucleation. Then the Mossop, Hobbs and Rangno, etc. studies emphasize the importance of initial liquid hydrometeor formation, rather than ice nucleation, to secondary production. We clarify by writing *"Other studies have also emphasized the importance of initial liquid hydrometeor formation, rather than primary nucleation, to ice generation from rime splintering."*

• Section 3.3: Make it clear what process which paragraph is referring to, it starts with Breakup, page 11, line 4 DS...

Thank you for pointing this out. To the start of the first paragraph, we add "First the effect of nucleation rate is investigated on the N_{INP}^{lim} value for breakup is illustrated in Figure 5. The top panels show results from a default nucleation rate and ones reduced by factors of 10 and 100."

We adjust the start of the third paragraph also: "Then the effect of shattering probability and generated fragment number are investigated for droplet shattering." And the start of the last paragraph: "Finally, we investigate the impact of the fragment number from rime splintering."

- Page 13, line 10: Please also describe what can be seen in panel b. Thank you for pointing this out. The commentary is expanded as follows: "*F_{RS} is the more influential factor in timing. Its impact on N_{ice} evolution is shown in panel b, where a given enhancement is obtained over a shorter period for a higher F_{RS}. As for breakup, the effect of the parameter is much larger when N_{INP}^{tot} is smaller in the yellow traces.*"
- Page 14, line 1: You did not really explain the single versus two-phasedness before? It is an interesting aspect and maybe you could explain that a bit further (here or in the sections before). The notion of single versus two-phasedness is mentioned in the abstract and one of the two key aspects in the analysis of Section 3.1. To make this more clear, we adjust the terminology in the first paragraph of Section 3.1: "The structure in the number evolution can be understood by considering whether the process is collisional and its 'phasedness', i.e., whether it involves hydrometeors in the liquid or ice phase or both."
- Page 14, summary point 1: It could be interesting to illustrate this point in a table or figure. In figure 8, it is not really depicted for each process separately.
 Thank you. We agree and have added a "structure summary" in panel (a) of an additional figure (below).
- Page 14, line 23: What do you mean by emissions? Aerosol emissions? Yes, aerosol emissions. This is added in.
- Page 14: It could be interesting to plot the dominant regions of each process on a 2D-Plot with the vertical velocity and the temperature on the axis.
 Thank you. We agree and have added a "thermodynamic summary" in panel (b) of an additional figure:



• Page 15, line 13: You could add more references here, e.g., Conen et al. 2011 and Steinke et al. 2016.

Thank you. These references have been added in here.

- Figure 1: Panel d is not mentioned / explained in the text. Either remove it or explain it in the text as well.
 Thank you for noting this. To the first paragraph of Section 3.1, the following is added: "These varying values of N_{INP}^{tot} are shown in different colors in panel d of Figure 1."
- Figure 2: Explain in the caption what n = 2, n = 10 means (it is only indirectly explained in the text). In fact in the caption, it states "for the main panel (a), droplet shattering generates 2 fragments per collision, and for the inset, 10 fragments per collision." But the n's are changed to \varkappa 's to agree with the notation in the text, and ' $\varkappa = 2$ ' and ' $\varkappa = 10$ ' expressions are added in parentheses to the caption explanation.
- Figure 3: In the case of *BRth* and *RSth*: does the same argument yield as in *DSth* for choosing a velocity of 0.5 instead of the smallest value of 0.1?
 Yes, exactly. This is added to the caption: "The lowest updraft of 0.1 m s⁻¹ is not shown because only very small enhancements occur. Droplet shattering is shown at 1 m s⁻¹ for the same reason."
- Figure 3: Why are there no meaningful enhancements by breakup if the updraft is larger? Less collisions?

An explanation for no breakup enhancement at larger updrafts is given on page 9, lines 6-7 (in the original manuscript): "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." If the parcel does not exist long enough for the depositional and riming growth of small crystals to graupel, then breakup will simply not occur.

 Figure 4: Why are there no meaningful enhancements by breakup at colder T₀? Thank you for pointing this out. Here we have added in an explanation at the end of page 9 (in the original manuscript): "no enhancement from breakup occurs again because graupel does not form. In this case, not only is the parcel too short-lived for graupel formation; diffusional growth is also slowed significantly at such low temperatures."

- Figure 8: It is a nice summary of the outcome of the paper and can be quite a useful Figure. You could strengthen that a bit more. In the current version of the manuscript is not very prominent. We include additional mentions of this figure earlier in the results section.
- Figure 8: What is meant by diminished INP efficiency? The idea of diminished INP efficiency is discussed on page 10, line 10 of the original manuscript, i.e., that the largest ice crystals enhancements are at the lowest N_{INP}^{tot} values and that as the INP concentration increases the secondary enhancement per INP, a kind of efficiency, drops. This term is highlighted in Section 3.2 to draw attention to it, and it is mentioned again in the Figure 8 caption.
- Figure S1: You could add *BR*, *RS*, and *DS*. Ok, the abbreviations have been added in.
- Figure S1: The process rime-splintering is not clearly depicted (how does the ice multiplication happen?).
 The figure has been adjusted to show a rimed graupel intermediate between the colliding droplet and graupel and the secondarily-produced crystals.

Small remarks, typos:

- The INP subscript is not nice to read. Reduce the space between the letters or write it non-italic (which is standard for physical subscripts?).
 Ok, the INP subscript has been un-italicized throughout.
- Page 1, line 20: Year missing at citation Ladino et al. Thank you. '2017' added to the Ladino citation.
- Page 2, line 28: Delete above. Ok.
- Page 4, line 3: Replace freezing by melting. 272 K is the melting temperature. Freezing normally happens at lower temperatures.
 Ok.
- Page 4, line 6: Also F_{DS} and F_{RS}?
 Yes, thank you. F_{RS} was not mentioned and F_{DS} was not listed next to its description. These have been added in.
- Page 4, line 7: The parameters for the functional form are β and γ, if yes add in brackets after "... per shattering droplet"
 Yes, thank you. F_{DS} (β, γ) is listed thereafter now.
- Page 5, line 13: Remove brackets around citation. Ok, thank you.
- Page 5, line 15: Add "explained in" before Section ... Ok.
- Page 5, line 25: What does "its magnitude" refer to? The magnitude of the enhancement. The wording in this topic sentence has been changed.

- Page 5, line 30: Has to be N_{ice}^{max} instead of N_{INP}^{max}? Yes, thank you for your careful reading.
- Page 7, line 16: Add brackets around the citation "(Paukert et al. 2017)" instead of "Paukert..." Ok.
- Page 14, line 19: Should or is? Yes, *is* is appropriate. The wording is changed.
- Page 14, line 28: The brackets are strange. These are fixed.
- Page 15, line 2: The term "supercooled liquid fraction" might need a sentence of explanation. The wording is changed to "the liquid portion of a mixed-phase cloud could also..".
- Page 16, line 22: This is also a leading coefficient? Yes, the wording is changed to correspond to that for *F*_{BR} and *F*_{DS}: "Leading coefficient of the fragment number per kilogram of rime..."
- Page 17, line 17: *D* (diameter) is missing in the variable list. Either add it or exchange it here with *r*.
 The diameter variable is exchanged for the radius.
- Table 1: In the Caption there is a run mentioned denoted INP below, which does not exist in the Table?
 Yes, thank you. The *INP* run is not described in the table, only shown in Figure 1d. The wording in the caption is fixed to reflect this.
- Table 1: Reformulate in the Caption (since thermodynamic simulations is ambiguous, *BRth* etc. is also a thermodynamic simulation): "Thermodynamic simulations run with ... are shown solely ..." Ok, thank you.
- For the simulations only shown in the Supplement (*BRDSth...*) no Table with conditions of the simulations exists. However this could be helpful in comparison to Table 1.
 A Table S2 has been added to supplementary information with the details of the BRRS, BRDS, DSRS, BRRSth, BRDSth, and DSRSth simulations, and the caption of Table 1 is modified to note this.
- Table 1: Run *DSpp*: What does the *D* mean in the range of values for *F_{DS}*? *D* indicated the diameter of the freezing droplets. Since the *D* is replaced by *r*, as you noted above, the same is done here. This is corrected throughout Table S1 as well.
- Table S1: *F_{RS}* and *F_{DS}*: What is frag? Fragments? Yes. This abbreviation is removed from the *F_{RS}* and *F_{DS}* expressions to avoid confusion.
- Table S1: p_{sh}: What is N?
 N indicates the normal distribution. This is now noted in the caption.
- Table S1: p_{fr}: What is A?
 A is a constant. It is substituted for its specific value, 0.82.

- Legend Figure 5 (a), (b), (c), Fig. 6 (a) and Fig. 7 (a) needs to be bold to be consistent (matters most in case of Fig. 5).
 The letters of these subpanels have been bolded.
- Figure 5 a, b, c: It is a bit unlucky that most points are in the bluish range of the color scale. It is quite difficult to differentiate the different color tones of blue. Independent of what color scale is used, the points will fall in a similar color range and be difficult to differentiate because they correspond to similar values.
- Figure 5 and 6 and 7: Is the color scale in the legend the same for the green traces and the yellow traces (also only color of green traces is shown)? Mention in the Caption or add the colors for the yellow traces also to the legend.
 Yes, the gradient from dark to light in both green and yellow corresponds to the same parameter values. There is not room in the subpanels to include a larger legend, so this is added to the caption.
- Figure 7: The coloring is only similar to Fig. 1 c) for the first color of the green and yellow traces? I found this comment a bit confusing and it did not add necessary information, so maybe delete it. Ok, it has been deleted.
- Figure 8: Are the arrows from one panel to the other needed? What do they symbolize? For now, we have left these in place, as an indication that each panel contains subsequent "criteria" for secondary production to occur.
- Figure 8: What is *s*_M? What is *D*_V? (left panel) These variables are the supersaturation with respect to water and the water vapor diffusion coefficient in air. Both have been defined in the Notation Appendix.
- Figure S2 Caption: line 1: add on *BRpp* in the end. This figure shows the effect of adjustments for the parameter perturbations with both breakup and droplet shattering, so "*BRpp and DSpp*" are added to the caption.
- Figure S2 Caption: line 2: (b) shows the effect of the minimum... function on... Ok.
- Figure S2 Caption: line 3: ... droplet due to F_{DS}?
 The caption text is adjusted: "Panel (c) shows the effect of the leading coefficient within the droplet shattering fragment generation function, while ..."
- Figure S2 Caption: (d) What is plotted here? Freezing probabilities? Does not fit to the plot. The caption states that panel (d) *"shows various temperature-dependent freezing probability distributions."* All temperature-dependent curves in this Figure are shown for a temperature range of 237 to 273 K since these are the only relevant values for secondary production.
- Figure S3: Replace um with μm. Ok.
- Figure S3 Caption: Add: in dependence of *D_R*. Ok.
- Figure S3 Caption: How is the second sentence connected to this figure? Yes, you're right. This is not relevant here; it has been removed.

- Figure S4 Caption: Shift bracket behind "number". Ok.
- Figure S7: Difficult to read legend (a). The coloring here makes the legend quite difficult: either black will not be visible on the blue or white will not be visible on the cyan. We tried to compromise with a gray.

	In-situ measurements	Laboratory studies	Parcel model simulations
Temporal evo-	BR and DS: 20 min to form drizzle drops	BR: 20 min to increase ICNC by a fac-	BR: Superexponential increase based on
lution of N _i	and 12-15 min to glaciation after first	tor of 10 with initial ICNC of 3 $\rm L^{-1}$	$N_{\mathrm{INP}}^{(tot)}$; DS: threshold increase based on
	ice (Taylor et al., 2016); DS: 2-3 min to	(Vardiman, 1978) (his Fig. 7); DS: only	p_{fr} ; DScoll Exponential increase based
	glaciation after first ice (Lawson et al.,	50 seconds to fragmentation after equili-	on N_R ; RS : Superexponential increase
	2015); RS: 10 ² enhancements within 10-	bration and nucleation time (Johnson and	based on N _R
	15 min (Hobbs and Rangno, 1990), 8	Hallett, 1968); RS: Linear increase start-	
	L-1 over 32 min (Heymsfield and Willis,	ing between 10 and 20 min (Hallett and	
	2014)	Mossop, 1974) (their Fig. 1)	
Limiting INP	BR: T_{top} between -10° and -18°C with	BR: Strong modulation of ultimate ICNC	BR: N ^(lim) _{INP} from 2 up to 70 m ⁻³ , possi-
or thermody-	$N_{\rm lim}$ from 0.1 to 5 $\rm L^{-1}$ (Rangno and	by initial ICNC (Vardiman, 1978) (his	ble only at warmer T_0 and slower u_x ; DS:
namics	Hobbs, 2001); DS and RS: Taylor et al.	Figure 1); DS: $N_{\text{INP}}^{(lim)}$ of 1 m ⁻³ (Beard,	no meaningful $N_{\rm INP}^{(lim)}$, favored at colder
nunics	(2016) cite the importance of the warm	1992), Favorable temperatures colder	T_0 down to 258 K as $u_{\rm z}$ slows; RS: no
	rain process through T_0 , CDNC, u_x , and	than those for RS (Korolev et al., 2004);	meaningful $N_{\rm INP}^{(lim)}$, favored for 268-270
	cell lifetime; DS: $N_{\rm INP}$ of 10^{-4} to 10^{-2}	RS: optimal temperatures between -3 and	K but this range widens as u_x increases
	L^{-1} for $\overline{N_i}$ of 572 L^{-1} (Lawson et al.,	-8°C (Hallett and Mossop, 1974), mod-	
	2015); RS: $N_{INP}^{(lim)}$ of 0.01 L ⁻¹ (Craw-	est updrafts are most favorable (Mossop,	
	ford et al., 2012)	1985; Heymsfield and Willis, 2014)	
Parametric	BR: In-cloud graupel collision rate of 1	BR : Fragment generation rate K_0 of	BR: 102-fold enhancement increasing
uncertainty	$m^{-3}\ s^{-1}$ (Mizuno and Matsuo, 1992),	0.00081 up to $0.01 L^{-1} s^{-1}$ (Vardiman,	F_{BR} from 40 to 280 at $N_{\rm INP}^{(tot)}$ of 0.17
-	10% of ice particles were fragmented	1978); DS: Shattering frequencies of 10	L-1; DS: 10-fold enhancement increas-
	(Rangno and Hobbs, 2001); DS: 10-9	to 37% between 50 and 120 μm (Brown-	ing $p_{sh}^{(max)}$ from 1 to 30% independent
	fragments per kg liquid (Lawson et al.,	scombe and Thorndike, 1968; Takahashi,	of N ^(tot) ; RS: 10 minute sooner enhance-
	2015), 10% of drops frozen by $-6^\circ C$	1976); RS: 250-700 splinters per mg rime	ment increasing F_{RS} from 3 x 10 ⁸ to 3 x
	(Brownscombe and Thorndike, 1968);	at $u_x = 1.5 \text{ m s}^{-1}$, 200-400 at 2 m s ⁻¹	10^9 for all $N_{\rm INP}^{(tot)}$
	RS : 1.4 L ⁻¹ s ⁻¹ (Taylor et al., 2016),	(Hallett and Mossop, 1974) (their Fig. 3),	
	50 crystals s ⁻¹ (Heymsfield and Willis,	90-350 (Mossop, 1985)	
	2014)		

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.