ANSWER TO REVIEWER 3

General Comments

This is a good paper overall, in its seeking to explore the role of different secondary ice formation mechanisms active under conditions present in clouds over the focused region near Antarctica. To the extent that I can tell, the findings with regard to BR parameterizations are solid and enlightening regarding the necessary conditions (especially heavy riming) for it to occur and the need for formulations of BR that account for the requisite size of colliding particles

We are grateful for the many insightful comments that have helped us improve our manuscript.

This paper was less forward and unsatisfying in conveying that end-to-end prediction of secondary ice formation actually still eludes the community. This is my inference from this paper and from the literature of the last few years represented by some of the team on this paper. This relates to the assumptions on primary nucleation, and the concentrations of ice that exists outside of regions where strong SIP ensues. There is still a gap here, one that is not really discussed, although this study seems to close it some. By this I mean that the authors have proposed to explain how ice concentrations go from values of 0.1 per liter to ones enhanced up to 100 times, and under conditions that the Hallett-Mossop process cannot explain such enhancement. The results cannot explain the inconsistency between recent INP measurements over the Southern Ocean or near Antarctica (e.g., McCluskey et al., 2018; Schmale et al., 2019; Welti et al., 2020) and the fact that Litkowski et al. (2017) required use of the DeMott et al. (2010) parameterization (with qualifications about its likely inapplicability to the region) in order to explain consistency with at least some of the ice concentration signature in the regional clouds.

The summer coastal low-level Antarctic clouds are dominated by supercooled liquid droplets while ice occurs in isolated large ice patches. Our statistics that focus on cloud ice properties by default refer to the ice-containing regions of the cloud. This indeed was not clear in the initial version of the manuscript and now is explicitly mentioned in the abstract, case description and conclusion section.

Although no primary ice scheme is likely suitable for polar conditions, DeMott et al. (2010) still performs better than all other parameterizations available in WRF (and generally in models with no prognostic aerosols). This was discussed in Listowski et al. (2017). Sensitivity tests however are considered to address the factor of 10 uncertainty in the chosen parameterization, but we acknowledge that the results of these tests were not emphasized enough in the main manuscript and particularly the large sensitivity to INP reductions. For this reason the whole section on primary ice nucleation has been moved from the Supplementary Material to the main text (section 4.4). The fact that a minimum primary ICNC concentration of 0.1 L^{-1} is needed to initiate break-up is now explicitly stated in both 'abstract' and 'conclusions' section. Potential suggestions on how this requirement might be met are offered in sections 4.4 and 5 (e.g. ice seeding as likely in our case, transport of terrestrial aerosols, more efficient H-M).

Moreover, the deficiencies of the applied parameterizations, such as the fact that they have been neither developed or calibrated for polar conditions, are also explicitly mentioned in the revised text (lines 399-404, 499-500).

The measurement issue is that it seems that many of the clouds remain supercooled or perhaps with ice in them at a level that could elude measurement due to detection limits. The initial transition to the point that calculations start in this study is unexplained. This does not greatly harm the paper, since it remains as a mystery for many investigators. However, some simulations performed (CNTRL case primary ice decreased by 10x) make it clear that BR would not occur in clouds with lower primary ice concentrations. This deserves some mention/discussion in the main manuscript. There must be "enough" ice, enough apparently being 0.1 per liter in the temperature regime between 0 and -9°C, and then BR can ensue and, if done properly, explain ice enhancements observed in many cases. But primary ice nucleation appears not capable of providing that starting ice concentrations, based on available information. I reiterate this point in some of the specific comments below.
The sensitivity in low INP conditions is now discussed more extensively in section 4.4 and the fact that there might be a 'triggering' primary ICNC threshold that can explain the observed ice patches within the predominately liquid clouds. Furthermore we discuss possible processes that can provide the necessary conditions to meet this requirement (e.g. ice seeding in the particular case). This finding is also repeated in the 'conclusions' section.

Specific Comments

Abstract:

Lines 27-28: In what studies has primary ice formation ever been constrained in the region of this study by aerosol measurements? To me it implies that someone has reliably connected aerosol measurements and ice nucleating particle measurements to primary ice formation measured in the region.
The abstract has been modified substantially and this sentence has been removed.

Lines 30-32: To be explicit, the parameterization bridges the gap between measured ice concentrations outside of SIP regions (perhaps, this is not clearly shown) and ice concentrations ultimately achieved in some areas.
By default ice properties are only calculated with the observed ice patches where SIP seems to occur. To avoid any confusion this is now explicitly stated in the abstract along with the fact that these ice patches are observed within predominantly liquid cloud layers.

Line 33: "Insensitive to uncertainties in one primary ice production parameterization." Or something to that effect. Without a clear understanding of primary ice formation in these clouds, one cannot claim that the uncertainties or concentrations are known. Given that some observations have not been mentioned, I suggest that qualification be made that end-to-end understanding still eludes the field. What the paper shows, and what is not made clear here, is that this is true as long as some other primary or secondary process is able to produce 0.1 per liter ice to begin with. Unfortunately, none is known to exist. It is wired into the model via use of a parameterization that has not been constrained by INP observations in this region.
We now make clear in both abstract and 'conclusions' section that a minimum

primary ice concentration of 0.1 L^{-1} is needed to initiate break-up. Potential contributions to this minimum value are also mentioned in section 4.4. and 5.

2) **Introduction: Line 53:** Suggest ": : :generation of ice crystals in a cloud that: : :" ...ensues following primary ice nucleation, but occurs in large excess to ice nucleating particle (INP) concentrations: : Otherwise this seems an awkward definition. It does require INPs or pre-existing ice from some nucleation process, but the key factor is that it far exceeds these initial values in concentration.
The whole paragraph has been revised (lines 51-55).

Line 77-78: Suggest that "it is" and "it" can be removed from this sentence for readability. Also, please note that neither of the referenced studies in this sentence were for the region of study, and the latter should be qualified as a modeling study.
Thank you, corrected

Line 103-104: This statement does not describe the results herein or in Young et al.(2019) in my opinion. They state, my emphasis added, "Under the assumption that primary ice is suitably represented by the model, we must enhance SIP by up to an order of magnitude to simulate observed Nice:" They did not observe INPs, correct? To be accurate, ": : :help to explain: : :"? Also it is important to note that this study will also not use observation of INPs as a basis for any sort of closure on complete understanding. The implementation in the Morrison scheme does require explaining the suitability of the inherent ice nucleation scheme, and so it was appreciated to have an Appendix relating to that.
We have modified this statement, which was inaccurate, by referring now to the discrepancy between observed and modeled ICNCs in line 101 (instead of INPs and ICNCs).

Line 149-154: ICNCs were substantially higher than what? The model ice? Expectations? Based on what? Just for reference, the ICNCs quoted here for the noted temperature range are not one order of magnitude higher than expectations based on more recently reported INP measurements in the Southern Hemisphere ocean region (references noted above), and are 3 orders of magnitude higher than Bigg (1973) at the nearest temperature of observation. Also, regarding noting the "frequency" of observations of low aerosol concentrations and high ICNCs, are you failing to acknowledge the other publications on the observational studies that indicate the apparently more frequent observations of supercooled water conditions? What I am getting at is being clear on what part of the discrepancies are endeavored to explain here, how one gets to these concentrations and from what level of ice presence. The model predicted values, or the diminishingly low values one might expect from observations of INPs? Is it possible to state what the lower detection limits for ice concentrations are in observations, and how this might affect the mean values or spread in ice concentrations?
Although clearly INPs can be substantially lower over the Southern Ocean, we do not have INP measurements for the particular case. For this reason we only state the aerosol concentrations available to indicate how clean is the measured atmosphere. Assuming that the average of observed INP from the literature is more representative for our case than INP diagnosed from direct aerosol observations (with uncertainty of one order of magnitude) is also subject to considerable uncertainty. Considering the high ICNC conditions in the observed cloud ice patches, surface INP measurements

may not account for ice-nucleating particles transported above/within the cloud.

When calculating ICNC statistics, by default this estimate concerns the ice-containing regions of the cloud. However, we acknowledge that this was not explicitly discussed in the previous version of the paper.

Regarding detection limits, the 2DS is a single particle instrument: it measures all particles that pass through its sample volume with size larger than ~10 μm. However phase identification cannot be conducted for particles with sizes lower than 80 μm. The 2DS sample volume depends on particle size and the data integration period. For example at 300 um the sample volume is 3.7 L/s. For 1 count measured within 1-sec averaging window, this equals a concentration of 0.27 L^{-1} (1 count/ (3.7 L/s $*$ 1s)). If a 10-sec window was chosen, then this value would go down to 0.027 L^{-1}. There is an uncertainty in the concentration due to the counting statistics (1/sqrt(counts)). For 1 count the uncertainty due to counting statistics is 100 %. This is now explained in section 2.1, lines 121-125. The total uncertainty in ICNCs is even larger but cannot be determined. However, what the applied thresholds ensure is that calculations concern ice patches, not liquid-only regions (line 243-244), and that this is consistent for both observations and model.

3) Modeling Methods General comment on this section: I found it very confusing to have two of the altered simulations refer to alteration of size dependencies, but in one for the limitation to be that crystals exceeded a certain size (FRAG1siz) and the other to required that size be scaled for a very large size to one much smaller (TAKAHsiz). I have no recommendation for describing size effects that are totally different.
We changed TAKAHsiz to TAKAHsc (to indicate scaling for size)

Line 226, end of Section 3: The abstract mentions sensitivity tests on primary ice nucleation. I understand that these are given in the Supplemental, but this should perhaps be mentioned here.
The whole primary ice section has been moved from the Supplementary Information to the main manuscript as section 4.4.

4) Results

Line 240: The CNTRL simulation underestimates mean ICNC, but it should be made clear that this is because of the misrepresentation of the highest ICNC, correct?
Yes, as now stated in lines 268-269 CNTRL cannot reproduce the whole spectrum and largely underestimates the frequency of ICNCs > 1 L^{-1}

Line 342-344; lines 352-353: This is not really the potential uncertainty in INPs, right? This is the uncertainty for the parameterization. That parameterization does not have representation in samples from either Antarctica nor from marine boundary layer regions. The reason this becomes important is the final statement ": : :as long as there are enough crystals to initiate this process."
The discussion in section 4.4 now addresses the fact that the parameterization is not constrained based on Antarctic measurements. The fact that a minimum primary concentration of 0.1 L^{-1} is required to initiate BR is now emphasized throughout the whole manuscript.

If INP number concentrations are exceedingly low, as they appear to be in some recent studies referenced above, is the BR process ever stimulated unless cloud tops get much colder? It seems not. It seems that some other process is needed to explain the presence of something on the order of 0.1 per liter already at all temperatures warmer than -9 ? Based on Fig. S4, I ultimately noticed that the basis of your statement must be in the simulations where INPs were decreased by a 10x factor. In that case, it appears that the BR process does not come into play. This is an important finding in my opinion. It deserves mention as another key result.
This is now mentioned as a key result. We also suggest processes that might result in conditions favorable for BR (e.g. ice seeding, aerosol transport). It is likely that understanding the interactions between such processes might explain the reason why large ice patches with substantially enhanced ICNCs are observed when supercooled liquid cloud conditions generally dominate.

5) Conclusions Lines 356-360: The results indicate that if sufficient ice concentrations are present already, or are parameterized to be at a certain level, then BR can do what is stated here. However, primary ice nucleation is not “limited” in this study. It is far in excess of existing INP measurements
The conclusion section now emphasizes that certain primary ice conditions are required to initiate BR. We also explicitly state that such INP conditions are not frequently found over Southern Ocean, which is likely the reason why supercooled droplets dominate in these clouds.

Appendix A

Lines 392-393: The Meyers et al. (1992) formulation for contact freezing is another parameterization of questionable applicability to this region. Ignoring that or whether it contributes at all or not, I am unclear on what is meant by the rates being further weighted by the effective diffusivity of the contact nuclei. You mean an altered assumption is made on their size? Or how size impacts scavenging rate? Please explain.
The effective diffusivity of the contact nuclei is estimated based on Brownian motion as (similar to Young 1974): $D_{ap} = R\ T\ (6\ p\ r_i\ N_A\ m\ )^{-1}\ [1 + 0.0737\ T\ (2880P)^{-1}\ r_i^{-1}]$, where R is the universal gas constant, N_A is Avogadro’s number, m is the dynamic viscosity of air, T is the temperature, P is the air pressure, and the radius of ice nuclei r_i is assumed to be 1×10^{-7} m. The factor in the brackets [] is a correction factor accounting for the mean free path of air molecules relative to the size of the ice nuclei (all units are MKS). This information is now provided in the revised manuscript in lines 488-493

Lines 397-398: If the parameterization for heterogeneous nucleation has been selected to compare better with in-cloud ice measurements over the Antarctic Peninsula, does that not “fix” the primary nucleation problem, without necessarily knowing if this is disguising other processes that must be at play to achieve such concentrations? Given what I mention already above, you might need to explain that this selection has been made to solve an issue or is perhaps based on some prior inference that these ICNCs must be the INP concentrations, not because it is the most appropriate thing to do. This is discussed in some of the papers that preceded this one, especially the fact that the parameterization selected was not relevant for application on sea spray particles.

Previous comparisons of this scheme and other primary ice nucleation schemes available in WRF for Antarctic Clouds have shown that the Cooper parameterization performs worse (Listowski et al. 2017). This is also the case for Arctic clouds (Young et al. 2017). Nevertheless, DeMott's and Cooper's schemes produce similar primary ice over the temperature range covered by the observations, but the latter provides unrealistically high values at lower temperatures (see Young et al. 2017 and Supplementary Information of Young et al. 2019). This is now explained in Appendix A (lines 496-504).

Figures

Figure 2: It is only here that some limitation on ice at 0.005 per liter is mentioned. Apparently, this is the minimum ice concentration accessible at some measurement frequency, due to sample volumes I assume. This is nowhere explained, but it should be reiterated here. This limit also sets the limit on what is referred to as supercooled water versus ice. What if this were set at 0.00005 per liter, which may be closer to what could be available as INPs? Yet in this figure, it somehow looks like there is a threshold around 0.2 per liter. Why is that?

1 count measured by 2DS within 1-sec averaging window corresponds to a concentration of 0.27 L^{-1}. This was the minimum concentration in our previous manuscript. Discrepancies between the data presented in the previous manuscript version and Young et al. (2019) are due to different data processing methods. In our case, we first applied the 0.005 L^{-1} threshold to our dataset and then interpolated ICNCs to the time resolution of temperature measurements. Young et al. (2019) interpolated ICNCs first and then applied the cut-off ICNC threshold. Their interpolation resulted sometimes in lower values, as interpolation between non-ice containing and ice containing regions occurred. The minimum value in their data however is 0.007 L^{-1}, still higher than the applied threshold. In other words applying even lower thresholds would not impact observational statistics. Nevertheless, for consistency between the two papers we now use the exact same data-processing method as in Young et al. (2019) and describe this in lines 242-244.

As far as modeled ICNCs are concerned, ice processes are calculated only if meaningful ice content is present ($Q_i > 10^{-8}$ kg kg^{-1}). The minimum ICNC (sizes >80 μm) corresponding to these Qi conditions found in the CNTRL model dataset is 0.0008 L^{-1}. This means that no meaningful ice is produced for INPs ~ 0.00005 L^{-1}, which corresponds to supercooled liquid-only conditions in the model. Considering the high frequency of liquid-only clouds in the region, this is the expected behaviour.

Supporting Information

Text S4: The statement here about the uncertainty in the parameterization used is correct, unlike the inference in the main manuscript. But again here, the 0.005 per liter threshold is mentioned in how it impacts the results of decreasing primary ice nucleation. Does this threshold need to exist? Again, it needs explanation, somewhere in this paper.

Yes, this threshold separates ice patches from liquid-only regions of the cloud. This is now stated in lines 243-244. Including non-ice containing clouds in the calculation of ice microphysical properties would cause a significant bias in the results.

Figure S1 and others: Is there a real reason why the scale needs to change? I was quite confused by these figures until I read the last statement of each caption.
Supplementary Figures have been moved to the main text (Figure 5 and 6). The same scaling is now used in all plots to avoid confusion.

Reference:

Young, K. C., 1974: The Role of Contact Nucleation in Ice Phase Initiation in Clouds. J. Atmos. Sci., 31, 768–776, https://doi.org/10.1175/1520-0469(1974)031<0768:TROCNI>2.0.CO;2.

Young, G., Connolly, P. J., Jones, H. M., and Choularton, T. W.: Microphysical sensitivity of coupled springtime Arctic stratocumulus to modelled primary ice over the ice pack, marginal ice, and ocean, Atmos. Chem. Phys., 17, 4209–4227, https://doi.org/10.5194/acp-17-4209-2017, 2017.