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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Secondary ice production in summer clouds over the Antarctic coast: an underappreciated process in atmospheric models" *by* Georgia Sotiropoulou et al.

Anonymous Referee #3

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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. 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 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 ℃, 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.

Specific Comments

1) 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.

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.

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.

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.

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.

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 N_{ice}."

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.

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?

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.

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.

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?

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." 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.

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.

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 nucle. You mean an altered assumption is made on their size? Or how size impacts scavenging rate? Please explain.

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.

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?

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.

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.

References

McCluskey, C. S., Hill, T. C. J. , Humphries, R. S., Rauker, A. M., Moreau, S., Strutton, P. G., Chambers, S. D., Williams, A. G., McRobert, I., Ward, J., Keywood, M. D., Harnwell, J., Ponsonby, W., Loh , Z.M., Krummel, P. B., Protat, A., Kreidenweis, S.M., and DeMott, P.J.: Observations of ice nucleating particles over Southern Ocean waters. *Geophysical Research Letters*, 45, 11,989–11,997. https://doi.org/10.1029/2018GL079981.

Schmale, J., Baccarini, A., Thurnherr, I., Henning, S., Efraim, A., Regayre, L., Bolas, C., Hartmann, M.,Welti, A., Lehtipalo, K., Aemisegger, F., Tatzelt, C., Landwehr, S., Modini, R. L., Tummon, F., Johnson, J., Harris, N., Schnaiter, M., Toffoli, A., Derkani, M., Bukowiecki, 35 N., Stratmann, F., Dommen, J., Baltensperger, U., Wernli, H., Rosenfeld, D., Gysel-Beer, M., and Carslaw, K.: Overview of the Antarctic Circumnavigation Expedition: Study of Preindustrial-like Aerosols and Their Climate Effects (ACE-SPACE), *Bull. Amer. Meteor. Soc.*, 100 (11): 2260–2283, DOI:10.1175/BAMS-D-18-0187.1, 2019.

Welti, A., Bigg, E. K., DeMott, P. J., Gong, X., Hartmann, M., Harvey, M., Henning, S., Herenz, P., Hill, T. C. J., Hornblow, B., Leck, C., Löffler, M., McCluskey, C. S., Rauker, A. M., Schmale, J., Tatzelt, C., van Pinxteren, M., and Stratmann, F.: Ship-based measurements of ice nuclei concentrations over the Arctic, Atlantic, Pacific and Southern Ocean, *Atmos. Chem. Phys. Discuss.*, https://doi.org/10.5194/acp-2020-466, in review, 2020.

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