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Comment on acp-2021-123

Anonymous Referee #2

Referee comment on "Comment on "Review of experimental studies of secondary ice production" by Korolev and Leisner (2020)" by Vaughan T. J. Phillips et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-123-RC2, 2021

This reviewer does not find the ideas and criticisms of KL2020 in manuscript particularly well organized or helpful with regard to evaluating the importance of ice-ice collisions to SIP. The discussion is largely qualitative and a repeat of what has been published previously by the authors. The language used is in several places exaggerated discussing the validation of both the Takahashi1995 experiments and the author's modeling results (presented in Yano2011, Yano2016, Yano2016b, Phillips2017, Philips2017b and Phillips 2018, hereafter termed "Y&P"). The Y&P work makes an important contribution to SIP research and several papers are cited in KL2020 but not described in detail as the focus of the review is experimental studies of SIP. In the early papers Y&P use a single parameter, the number of fragments produced per collision, extracted (and scaled) from the Takahashi1995 laboratory experiments to characterize the fragmentation. The parameter in Yano2011 was not temperature, humidity, LWC or particle size/character dependent. A more complex formulation is discussed in Phillips2017,2018 but this is not discussed here. At issue in this manuscript is the validity of the Takahashi1995 measurements and the simple scaling used in Yano2011,2016 to describe the in-cloud SIP process. Does the fragmentation observed by Takahashi1995 using 2 cm ice balls held on rods accurately simulate a SIP process in clouds? For several reasons I think this remains an open question.

I have the following comments on the manuscript:

1) Line 11: Shouldn't the word be "untested" rather than "erroneous" in describing the current situation? It seems KL2020 is not suggesting Takahashi1995 is erroneous. We have no basis to conclude the validity one way or another. Instead, there is concern these results do not simulate actual in-cloud collision process. Confirming experiments with free-falling and proper-size ice particles have not been done yet.

There is a hint in the manuscript (see lines 145, 281, 288, 301) that the authors believe the comparison of their model results with field-study measurements (like in Phillips2017b) is in sufficient agreement as to validate a fit-derived fragmentation parameter and are not sensitive to the value extracted from Takahashi1995. If this is the claim, then this point is important and needs to be discussed in its own section. Phillips2017b shows good simulation-observation agreement but it is unclear to what extent the fragmentation parameter is the only free-parameter. More discussion of how one can determine a fragmentation parameter from a SIP cloud model-field data comparison would be interesting.

2) Line 22: "Impossible" seems an exaggeration. The question is whether the laboratory result and scaling used in Yano2011 is a realistic description of in-cloud processes.

3) Line 23: "unreliable, which is not the case," - this statement needs to be explained. On what basis do the authors claim to know the Takahashi1995 data set is a reliable representation of in-cloud collisions? I have a similar comment to the text on Line 41 and several other places in the manuscript. Are the authors claiming a Phillips2017b-like analysis of other field data sets also shows good model-data agreement? Wasn't the breakup parameterization used here more complex than simply the size, velocity, and KE scaling of Takahashi1995? Perhaps provide more discussion of the later model refinements and the evidence for the Takahashi1995 extracted parameter used in Yano2011?

4) Lines 52-110: This material repeats much of what is stated in various places in Y&P. It is so scattered and un-quantitative as to provide few new insights. A more organized presentation would be appreciated.

5) I comment on the Takahashi1995 characterization: This reviewer doesn't find the Takahashi1995 result particularly compelling as a simulation of what occurs between free ice particles in clouds. Not to say it is erroneous, just on it's face not compelling. The reviewer's opinion isn't particularly important here but again a more organized quantitative analysis of the Takahashi method rather than the collection of scattered hand-waving arguments would be appreciated if this is to be one focus of the manuscript.

The SIP mechanism is unknown at this point. There have been several suggested ideas. The authors have published a model based on ice-ice fragmentation during collisions and claim their model can describe the process. The Takahashi1995 study collided two 1.8 cm diameter ice spheres, counted the crystals on a collector plate covering a fraction of the chamber bottom, and then multiplied that number by 4 as an estimate for the number of crystals ejected from their colliding spheres. Are these crude experimental finding for 2 cm sphere collisions an accurate representation of the ejection rate for ice crystals in clouds?

The scaling relation used in Yano2011 (to apply Takahashi1995 result to realistic cloud particles) considers only differences in particle diameter and fall velocity. Later in Y&P kinetic energy, growth time, vapor density, collision dynamics/type and stochastic considerations are all mixed into this scaling. But the scaling has not been confirmed by experiments. Korolev2010 mentions several questions in applying Takahashi1995 to actual cloud particle collision processes. I won't describe these considerations but will discuss several other considerations.

There are several ideas for how fragments occur during collisions. Apparently for some temperature, LWC, convection and humidity/riming conditions, cloud processes produce irregular or "fuzzy" ice spheres with fragile irregularities protruding from their surface. The idea presumes the protuberances grow with time and their fragility may (or may not) also increase. When a collision occurs involving at least one of these fuzzy particles some protuberances break off as fragments. This potential secondary ice production mechanism requires the fragments somehow find themselves a region with sufficient humidity to survive and grow thereby increasing the ice particle number density. Andy H's comment describes cases where the fragments likely will not survive. Phillips2017b describes a case where the fragments apparently do survive. Others will have different or more sophisticated ideas for the microphysics. But in this SIP mode the fragments originate at the surface of one or more of the colliding particles. The surface of the particles involve roughening or new crystallite nucleation such that the protuberances grow via riming or vapor deposition. These processes are temperature, RH, LWC and particle-size dependent. The surface roughness of the spheres in the Takahashi experiments were not characterized. At the surface during lumping/roughening or protuberance formation there will be epitaxial effects from the underlying crystallinity that likely will depend on the initial formation and growth process of the underly crystal. The 2 cm spheres in Takahashi1995 began as frozen liquid water inside a metal sphere. This freezing process is quite different from the variety of ice particle formation processes that occur in clouds undergoing SIP. Potential differences in the ice surface properties alone might call into question the relevance of the experiment to actual cloud particle-particle collisions.

Second, the collision itself is likely different from what occurs between cloud particles. In the experiment the 2 cm spheres move via a rod frozen into the center of the sphere. The rigidity of the rod is important to the amount of energy exchanged in the inelastic collision. A springy rod and axel will cause the balls to react much different than a ridged rod. Also during the collision, the strain along the axis of the rod will be different than in other directions causing perhaps larger amounts of gouging into the ice surface than would occur with free particles. An apparatus holding the ice on rods adds complications to evaluating the forces in each inelastic collision. One suspects the energy transfer and the potential for gouging into the ice surface are different from what occurs when free-particles collide. There are also aerodynamics considerations and charging effects for both the particles and the fragments created in colliding smaller crystals. Perhaps all these

potential effects wash out and the simple kinetic energy considerations are good enough to describe what is occurring. But it does seem fair that some experimental work using um scale and larger particles is needed before one can be confident the simple scaling idea, like that applied in the Yano2011 analysis, is valid.

This reviewer suggests the manuscript needs considerable re-work and clarification.