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Discussions

# *Interactive comment on* "Review of experimental studies on secondary ice production" *by* Alexei Korolev and Thomas Leisner

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This review article fills an important void in the atmospheric chemistry and physics literature by aggregating and synthesizing the broad range of literature relevant to secondary ice production in clouds. The article is thorough in its review of the literature and is helpful in going beyond merely reporting prior results, but placing them within the context of the full body of work and the current understanding. I had two personal impressions while reading the review. First, we as a community have strayed too far from our roots, and while there is some excellent laboratory work still taking place, it is disproportionately small compared to the vast efforts currently focused on field and computational work. I agree with the authors' perhaps provocative statement that laboratory work cannot be replaced by field work if we hope to achieve physically-based



understanding and parameterization of SIP processes. Rather, these efforts need to take place hand in hand. Second, there is clear value in bringing all of the relevant experimental results together for a cohesive review, with the result being much more impactful than simply the sum of individual studies. Meaningful theoretical progress rests on the observations from these collective experiments. To put it another way, taking electromagnetism as an example, there would have been no Maxwell without a Faraday. I hope these impressions come through clearly to other readers of this review. I would go so far as to say that the authors should take the liberty of editorializing somewhat more along these lines in the Concluding Remarks section; that is their prerogative, though, and I only share it as my opinion. In any case, I consider this to be a well written and important contribution that will help in providing a deeper understanding of what we know about SIP, and motivation for further work on this topic. Figure 16 is a distillation of the key findings and illustrates the mechanisms in a clear, graphical way that should help the main results be accessible to a broad range of readers, including those from other communities who may have overlapping interests with the subject (e.g., materials science, turbulence, etc.). Variations of this figure will likely appear in future cloud physics textbooks.

The following suggestions should be considered in revising the review. I have listed them roughly in order of priority.

1. In the "Way Forward" section it would be very helpful to summarize some of the key points that came up throughout the paper, regarding what aspects need to be carefully considered in future laboratory experiments. I came up with the following list, but may have missed some points. Laboratory experiments on SIP mechanisms likely need to consider the following variables, in order to ensure that the results are of atmospheric relevance:

- Particle fall speed and its influence on enhancement of diffusive fluxes, mixing in turbulent wake (ventilation effects).

- Relative velocity and impact parameter for mechanisms involving particle-particle interactions.

- Ambient atmospheric pressure, temperature and gas properties.

- Thermal equilibration of particles with the surrounding atmosphere (or realistic values of thermal lag for typical atmospheric temperature profiles and turbulence properties).

- Presence of dissolved gases and other impurities; in particular, unrealistically high concentrations of gases such as CO2 should be avoided.

2. One possibility not considered in defining primary vs secondary ice production: Could there be "primary" mechanisms that do not involve INP, or that strongly enhance properties of otherwise ineffective INP? Might these be relevant since they could lead to apparent discrepancy between expected and observed number of ice crystals, even without the operation of secondary ice processes? I have in mind pressure perturbations and electrical effects, as examples (citing work with which I am familiar… I am sure there is much more… consider Yang et al. 2015, Applied Physics Letters "Ice nucleation at the contact line triggered by transient electrowetting fields" and Yang et al. 2018, Phys Rev E "Nonthermal ice nucleation observed at distorted contact lines of supercooled water drops").

3. It also would seem relevant to discuss laboratory or cloud chamber experiments in which no secondary ice production was observed. Knowing the conditions under which SIP is not required is valuable for determining what part of parameter space should be searched. Again, drawing from familiar work, and acknowledging that there must be more, I have in mind the cloud chamber study of Desai et al. 2019, GRL "AerosolâĂŘMediated Glaciation of MixedâĂŘPhase Clouds: SteadyâĂŘState Laboratory Measurements", in which agreement was observed between injected INP and observed ice crystal concentration, to within experimental uncertainties. Interestingly, although it was not emphasized in the paper, multiple images of "pac-man" shaped ice crystals were observed in that study (see their Figure 1), and one can speculate that



they are fragmented frozen cloud droplets. And yet they do not seem to have contributed significantly to ice budgets under the existing experimental conditions (limited to relatively small particle sizes with only 1 meter of vertical distance for fallout and thereby limited particle lifetimes).

4. My first impression was that there is a lack of balance between the ice shattering mechanism covered in section 2 compared to the other sections. Upon reflection, though, I realize that it is a result of more literature being available in that area. A word on this at the beginning of section 2 would help orient the reader, allowing to understand that the fundamentals of ice growth in supercooled liquid, etc., have been thoroughly studied and are of relevance to the drop shattering problem covered later in the section.

5. The discussion on page 4 (especially near lines 391-392) raises the question of gas equilibration time. For natural cloud droplets grown by condensation the gas content may be substantially different than for droplets generated in a laboratory from atomizing a bulk liquid that is presumably in equilibrium with ambient gases. Could this be a relevant factor to consider?

6. Clarify on lines 58-61 that you are referring to artificial ice shattering that results from sampling/measurement (as opposed to ice shattering from natural processes, which are also considered in this review).

7. Clarify that equation 4 is for the assumption of a spherical droplet in air. Also, for people in other fields who might be accessing this review, provide a reference for the ventilation coefficient and specify that it is a function of terminal speed and therefore of diameter.

8. Clarify on lines 176-178 what is meant by the "spatial scale of the ice crystals".

9. The meaning of Figure 5 and "the diameter of the monocrystalline frozen drops decreases with the increase of supercooling" is not clear to me. How do I interpret a

data point at a specific radius and supercooling? Does it mean that at lower supercoolings the frozen drops are single crystals and at higher supercoolings the drops are polycrystals? I would have assumed that there is a probability of single versus multiple crystals. Is the data point the probability of 0.5? More explanation is needed here.

10. In a few places there should be more acknowledgement of uncertainty, such as line 275 where it is probably more reasonable to say "Such a high rate of splinter production may be an important factor in the INP economy"... since it surely depends on many other factors as well. On the other hand, I see at least one place where the view of the field may be overly pessimistic: line 432 "remains poorly understood or unknown" would seem more reasonable to be "remains only partially understood."

11. In the caption of Figure 10 you refer multiple times to INP, but in this case you are referring to an ice particle colliding with a supercooled droplet. Strictly speaking, yes, the ice particle could be considered an INP, but to me it seems misleading. If we consider INP as usually defined, then this refers more to heterogeneous (primary) ice nucleation. Indeed the question of surface versus volume crystallization is intriguing, but it is more closely related to primary ice formation.

12. In the paragraph discussing the paper of Baker (1991), near line 750, it should be made clearer that Baker considered static drops during the transient freezing process, whereas others such as Prabhakaran et al. account for continuous production of supersaturation in the wake of a falling particle that is riming (wet growth) or melting. In the next paragraph I would also suggest that it would be helpful to have a better understanding of how INP behave at very high liquid-water supersaturations, since this is a regime not typically achieved with current instruments (Fukuta had a wedge method that produced very high supersaturations and indeed observed higher INP efficiency in that regime).

13. My initial reaction was that Section 8 does not really fit with the main theme of the review. It is relevant in the sense that spurious ice crystals may have contributed to



the apparent conflict between measured INP and measured ice crystal concentrations. But then it begs the question why other field measurements are not reviewed as well. One aspect that could be emphasized to strengthen the connection to the laboratory focus of the paper is that the high speed videos in the Koroleve et al. papers were obtained in a wind tunnel setting (at least that is my recollection). Perhaps this is a good place to emphasize that lab "experiments" have contributed not only to understanding of fundamental mechanisms, but also to the evaluation of measurement techniques applied in the field. Those videos captured in a controlled lab environment settled the question of shattered ice crystals in the minds of many in the community. I would also point out that this section misses an important reference to Jackson et al. 2014, JAOTech, who made a full assessment of measurement-induced ice shattering based on intercomparison of multiple instruments.

14. In Section 9.1 it could be useful to elaborate more on "the most striking outcome of this review", that there is such a wide range of results for each SIP mechanism. In conversation with colleagues I have encountered a sense of exasperation that lab experiments sometimes show bewildering complexity. I even remember a story from an individual involved with experiments in the Hobbs lab in the late '60s that suggested that the CO2 contamination they identified as a cause of drop shattering is one factor that motivated Prof. Hobbs to shift his group's emphasis to field work. The result of the "striking outcome", however, should be that we carry out more, not less experimental work, in order to clarify the various unrecognized factors and ultimately to gain a full understanding of the relevant processes.

A thorough check for grammatical and typographical errors should be made. Overall the writing is excellent, but there are multiple places where small errors appear. I summarize the ones I found, although I probably did not catch all while reading:

Title: It sounds more natural to my ear to say "experimental studies of secondary ice production" rather than "on secondary ice production". But I'm not a grammar expert, so I would not go so far as to say it is incorrect.

Line 34: Schaefer

Caption of Figure 4: \Delta T = 14.5 C (should not be negative as currently shown).

Line 205 and several other places: A temperature is high or low (not warm or cold, which is only for an object).

Line 208: the Visagie experiments.

Lines 335-336: experiments that had a droplet suspended (no “is” needed).

Caption of Figure 8: I do not find Lauber et al. 2015 in the references. Should it be 2018?

Line 425: number of parameters.

Line 489: in a cloud chamber.

Line 511: studies of Hallett and Mossop.

Line 526: rapid growth of the ice shell.

Line 581: Phillips (also this sentence might be clearer is written “The studies of Hobbs and Farber, Vardiman, and Phillips et al. were based on the consideration...”).

Line 633: thickness of.

Caption of Figure 14: AgI and Snomax were used as ice nucleating particles.

Lines 722-723: aerosols introduced into the ambient air.

Line 737: ice sphere in humid air.

Line 738: The word “study” appears twice... should be rephrased.

Line 765: Not sure if “feedbacking” is a word.

Line 783: the existence of shattering.



Line 843: a complete quantitative theoretical description.

Line 848: systematic basis in weather prediction models.

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