

Replies to the reviewer's comments (Andrew Heymsfield) on "Review of experimental studies on secondary ice production" by A. Korolev and T. Leisner

From Authors: The authors appreciate the reviewer's time spent to read the paper and provide a diligent review. We found the comments very helpful in improving the manuscript. Below are point-by-point replies to the comments.

This is an excellent review article of a process that is very important for precipitation development, Secondary Ice Production (SIP). Based on observations I've made and those reported on by others, the process is particularly important when 1) cloud top temperatures are relatively warm, 2) there are relative few but some ice nuclei active at the cloud temperatures, 3) the cloud droplet sizes are relatively large, and the updraft velocities, although present, and not too strong, one to a few meters per second. SIP is therefore likely to be most active and important over relatively warm oceanic areas. Although several SIP mechanisms have been proposed, it is unclear when specific ones are active and under what conditions they occur.

This article discusses the pluses and minuses of the following SIP that have been proposed: (1) shattering during droplet freezing; (2) the rime splintering (Hallett-Mossop) process; (3) fragmentation due to ice-ice collision; (4) ice particle fragmentation due to thermal shock; (5) fragmentation of sublimating ice; and (6) activation of ice nucleating particles in transient supersaturation around freezing drops. The article focuses on laboratory studies, although some field observations are presented. Obviously, laboratory studies benefit from the ability to repeat experiments and narrow down possible processes by modifying the experiments appropriately.

I have relatively few comments because the article is very well written and extremely thorough.

Line 34: Schaefer

Reply: Corrected.

Line 59: "shattering" to "fragmentation"

Reply: Corrected.

Section 2.2, Eqs. (1) and (2), Freezing Fraction. Shouldn't ventilation enter into this discussion? It is factored into Eq. (3).

Reply: The ventilation factor in Eq.1 is included in the term ΔQ which describes the heat loss due to thermal exchange with the environment. Due to a very short duration of the recalescence stage ($10^{-5}\text{s} < t_1 < 10^{-1}\text{s}$ depending on D and ΔT) the thermal exchange between the surrounding environment and droplet is much smaller compared to the energy of the latent heat released during freezing. Accurate assessment of ΔQ suggests that it is much smaller other terms in Eq.1 and it is usually neglected. Eq.3 employs $u(T)$ obtained from experimental measurements. As before the due to a very short time of the recalescence stage the effect of ventilation in calculation of t_1 is neglected.

Section 2.7, Fragmentation during freezing. A table summarizing your discussion of shattering during drop/droplet freezing would be very helpful.

Reply: Table 1 summarizing shattering during drop/droplet freezing was added in the text following the reviewer's comment. In addition, we also added Table 2 summarizing laboratory results of the studies of the HM-process.

Table 1. Summary of experimental studies of droplet fragmentation during freezing by different research groups. The table covers only works that quantified the parameters included in the table.

Reference	Diameter (mm)	Temperature (°C)	Droplet suspension	Method of nucleation	Maximum SIP frequency (%)	max number fragments per drop	Temperature of maximum SIP rate
Mason and Maybank, 1960	30-1000	-2 to -25 ¹	stagnant (fiber)	various ²	47	200	-10C
Adkins, 1960	4-13	n/a ³	free fall	natural ⁴	0	0	n/a
Hobbs and Alkezweeny, 1968	20-150	-8 to -32	free fall	various ⁵	>5	n/a	no temperature dependence
Brownscombe and Thorndike, 1968	50-90	-5, -10, -15	free fall	tiny ice crystals	14	12	-15°C
Dye and Hobbs, 1968	1000	-3 to -15	stagnant (fiber)	tiny ice crystals	0	1	no temperature dependence
Johnson and Hallett, 1968	1000	-5 to -20	stagnant (fiber) +ventilation	tiny ice crystals	>1	n/a	no temperature dependence
Takahashi and Yamashita, 1969	600-800	-18 to -25	free fall	immersion ⁶	11	n/a	-15°C
Takahashi and Yamashita, 1970	75-350	0 to -30	free fall	tiny ice crystals	37	n/a	-15°C
Takahashi, 1975	45-765	-4 to -24	free fall	tiny ice crystals	35	n/a	-16°C
Pruppacher and Schlamp, 1975	410	-7 to -23	airflow	contact ⁷	15	>3	-11°C to -15°C
Bader et al., 1974	30, 42, 84 ⁸	-10 to -30	free fall	immersion ⁹	?	10	n/a
Kolomeychuk et al., 1975	1600	-12 to -25	airflow ¹⁰	natural ⁴	35	142	-15°C to -18°C
Lauber et al., 2018	300 -320	-5 to -30	stagnant (EDB)	tiny ice crystals	35	12	-7C to -13C
Keinert et al., 2020	300- 320	-1 to-30	stagnant (EDB), airflow	tiny ice crystals	1	3	-10C to -15C

1. ice nucleation temperature 0°C to -15°C
2. natural nucleation, silver iodide, contact tiny small ice crystals
3. not available
4. no special efforts were made to nucleate droplets
5. natural or immersed silver iodide
6. kaolinite or silver iodide
7. kaolinite or montmorillonite
8. mean volume diameter
9. silver iodide
10. flow of humidified nitrogen

Line 445: During the freezing process, the surface of the droplet is sublimating, perhaps affecting the fragmentation process.

Reply: The referee is right, roughly 1 % of the droplet mass evaporate per 1K of initial supercooling. This is a small amount and most of this evaporation occurs during the early stages of the second freezing process (Keinert et al. 2020), while the SIP processes occur during the later stages of the second freezing process. It seems, that it is unlikely that evaporation will affect SIP much, and therefore, we do not discuss it in the manuscript.

Line 649: undersaturated>subsaturated.

Reply: Corrected.

Line 732 "bigger" to "larger".

Reply: Corrected.

Line 783: the existence of shattering.

Reply: Corrected.

Line 824: concentration.

Reply: Corrected.

Line 825: You could mention the airborne studies of Mossop and Bigg and the use of balloon borne replicators, etc that can shed light on the problem. Many other airborne studies (Heymsfield and Willis, Lawson et al., Lasher-Trapp etc are directed towards the SIP problem. At the beginning of this section, re-emphasize that this review article is mostly directed towards laboratory and theoretical studies.

Reply: Several early Mossop's airborne studies (1970, 1972, 1985) were referenced in the introduction. The references on the studies of Heymsfield and Willis, Lawson et al., Lasher-Trapp et al. were provided in the concluding section to address the reviewer's comment.

The Bigg (1996) studied IFNs in Arctic clouds. There is a brief mentioning of SIP among other possible reasons to explain one of the observations of discrepancy between ice particle and IFN concentrations. This does not provide strong evidences of SIP compared to the early studies, such as that by Koenig, Hobbs, Mossop, and others mentioned in the introduction. The authors consider that referencing of the Bigg (1996) study would be destructive as not directly relevant to this study.

At the beginning of the section 9 the following text was added to address the reviewer's comment regarding laboratory studies:

“In this section we discuss results of experimental studies of artificial fragmentation of ice particles during in-situ sampling.”