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Comment on acp-2021-557

Anonymous Referee #1

Referee comment on "Secondary ice production during the break-up of freezing water drops on impact with ice particles" by Rachel L. James et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-557-RC1, 2021

Secondary ice production is an important topic, so I support publication of this paper. The authors have shown, convincingly, that collision of a supercooled droplet of water with a larger ice particle can produce secondary droplets. The authors have shown, less convincingly in my opinion, that those secondary droplets might freeze.

In essence, the experiments described in this paper are a refinement of those described in the JFM paper by Schremb et al. (the authors cite this paper). The authors here have documented some aspects of liquid water-ice collisions that Schremb et al. did not, and have quantified some others. My primary concern in the interpretation of these results is the freezing of the secondary drops. I am convinced, both from this paper and from Schremb's, that secondary drops are produced from instabilities in the rim of the water drop as it splashes across the ice. The filament structures shown in Fig. 6 are also a potential source of secondary droplets, though the number produced wasn't/couldn't be quantified in this study.

It wasn't clear to me from the manuscript whether the secondary drops froze in the air, or whether they froze once they landed on the substrate. If they froze once on the substrate, it is highly likely that freezing was because they are on the substrate. The probability that droplets of that size will freeze at temperatures in the range of approximately -10 C is very low. (This is true if they are on the substrate, but especially true if they are not.) Because the original ice is sitting on the substrate, there's the chance that a very thin film of ice can propagate along the surface from the ice to the supercooled liquid of the secondary droplet and cause freezing. (The ice that propagated along the substrate might not be apparent.)

The authors do address one possibility of how those smaller, secondary drops might freeze at temperatures as high as their experiments – shear at the ice-liquid interface which breaks off an ice embryo which causes freezing in the secondary droplet. I find this explanation unconvincing. (Something like this is also alluded to in Schremb's paper. I find it unconvincing there too.) If we impose a no-slip boundary condition at the solid-liquid

interface, which we usually do, there's no shear at the interface. The shear is all in the liquid. If there's a frost-like layer on the ice, pieces of that might break off into the liquid that becomes a secondary droplet, I suppose. If that were to the case, I would expect freezing of the front, not necessarily freezing of the secondary droplet once it detaches. If the freezing mechanism is in fact shearing of an embryo into the secondary droplet, you can estimate some typical time scales. You know the time scale for detachment from the measurements. See section 16.1.4 in Pruppacher and Klett for some thoughts on freezing time for droplets.

Perhaps mechanical agitation could trigger a freezing event. There are reports in the literature of freezing catalyzed by collisions. (See Alkwezeeny 1969 and Czys 1989.)

To re-emphasize my earlier point… I am in favor of publication of this paper, despite my misgivings about some of the interpretation of the data. Secondary ice is an important topic, and I think we need to consider a very wide range of possibilities and mechanisms. Also, to be clear, I'm not asking for more experiments for this paper. Some clarification on the points I raised above would be good, but I think it is enough to acknowledge these points in the present work and leave further work for further work.

Minor points:

The authors note that the falling drops were all the same size and fell from the same height, so that the impact velocity was 5.2 m/s. A comment here on how representative that might be as a closing velocity in the atmosphere (where it is most likely ice overtaking more slowly falling liquid drops) is warranted.

Line 176: 11 C. Missing a negative sign here?

Final sentence of the paper: "Further work is needed..." I agree. This manuscript is an interesting addition to the literature in my opinion, but opens a lot of questions as well. (Many of the best papers do…)

References

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