AC Response to RC1

We thank the referee for their time in reviewing our manuscript and appreciate the constructive feedback given.

RC: 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.)
AC: We believe that the drops we observe on the substrate are only formed during the retraction phase of the supercooled water drop and froze on the substrate. We did test drop impacts on the substrate without the presence of the ice particle and did not observe freezing within 30 min, whereas, for drop impact onto the ice particle freezing was observed within 10 s after impact. Therefore, this suggests that the substrate does not cause supercooled drops to freeze.

RC: 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.
AC: We thank the referee for their insight. We have removed the paragraph where we suggest that freezing could be due to shear and have added the following paragraph:
'Whilst the freezing mechanisms of the secondary drops was not specifically studied in this work, we consider the following mechanism. The freezing of supercooled water drop occurs in two stages. The first stage is characterised by the formation of ice dendrites throughout the supercooled water drop.

The latent heat from the formation of the ice dendrites is released during this stage, warming the temperature of the supercooled water drop to ~0 °C. The second stage is characterised by the freezing of the remaining supercooled water drop and is controlled by the loss of latent heat due to the supercooled water drop surroundings. Stage 1 of freezing is fast and the time taken for this stage to complete (ti) can be estimated from the following equation (Macklin and Payne, 1967):

$$t_i \approx \frac{G}{\delta R}$$

where δR is the thickness of the layer of supercooled water on the ice particle and G is the growth velocity of ice which is temperature dependent.

From Fig. 4, we can estimate that the rim of the supercooled water drop, which is also the thickest part of the supercooled water drop, is approximately 0.78~mm. Taking this value for δR and given that the growth velocity of ice at -5 °C is approximately 1 cm s^{-1} (Pruppacherand and Klett, 1997, chapter 16) then $t_i = 0.078$ s. Figure 4 shows the time-scale for the retraction phase is of the order 0.1 s. It is plausible that the initial ice dendrites can propagate through the supercooled water drop and that water containing these dendrites may then break off during the retraction phase and initiate freezing. The second phase of freezing will take longer, but as long as the drop contains ice dendrites it will eventually freeze. This explanation is also proposed by Schremb et al. (2018) and Phillips et al. (2018) who suggest that seeding ice crystals are transported during the initial spreading phase. Alternative freezing mechanisms include the formation of a thin, unobserved film of liquid water present on the glass slide after the retraction phase. The contact between the thin film of water and the ice particle could induce freezing in the thin film, which could then trigger freezing in the seemingly detached secondary drop. Mechanical agitation or shock may also play a role in the freezing of the secondary drops (Alkezweeny, 1969, Czys, 1989). Regardless of the freezing mechanism, the glass slide will likely have some influence, and it will be pertinent to remove this in future investigations.

RC: 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.)
AC: Added following sentence:
'Alternatively, mechanical agitation or shock may play a role in the freezing of the secondary drops (Alkezweeny, 1969; Czys, 1989).'

RC: 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.
AC: We thank the referee for their comments and suggestions and have added clarification to the points raised above.

Minor points:

RC: 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.
AC: Added:

‘The terminal velocity of a 5 mm diameter drop is approximately 9 m s^{-1} (Gunn 1949). Initially, the impact velocity may seem unrealistic. However, the ice particle in these experiments was held stationary on a glass slide, but in the atmosphere the ice particle would also be falling. The terminal velocity will depend on the ice particle shape, but for aggregates of similar size it is typically around 1 m s^{-1} (Locatelli and Hobbs, 1974). The differential velocity between the supercooled water drop and ice particle will be less than 9 m s^{-1} dependent on the nature of the ice particle. While such large droplets are rare in the atmosphere the purpose here is to demonstrate that the process is a potential secondary ice mechanism.’

RC: Line 176: 11 C. Missing a negative sign here?
AC: Yes – added negative sign.

RC: 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…)
AC: We agree that this work raises many more questions and thank the referee again for their constructive review! In light of another reviewer’s comments, we have expanded on the conclusions section to include some suggestions on how we would further this work.

‘One of the main experimental challenges of this work was dropping the supercooled water drop consistently onto the ice particle which limited the amount of experiments we could perform. As shown in Table A1, the majority of the successful impacts were classified as partial hits despite the intention for them to be direct hits. While partial hits are expected in clouds, as well as direct hits, we also conducted many experiments where the supercooled water drop missed the ice particle. One method of achieving better control of the supercooled water drop impact could be via growth and supercooling of a water drop at the end of a needle similar to the system shown in Schremb et al. (2018). Compared to our current mechanism, which involved tilting a pipette to allow the supercooled water drop to roll off, the supercooled water drop would remain fixed to a certain point before detaching under gravity, making it easier to drop consistently in the same position.

Another experimental challenge we would like to address is quantifying the secondary drops formed during the spreading phase of the supercooled water drop during impact. Thoroddsen et al. (2012) quantified secondary drops ejected with velocities of up to 100m s^{-1} using an ultra-high-speed camera capable of recording at 1000000 fps, and we could use a similar setup. We could then exploit the birefringent properties of ice to determine whether these ejected secondary drops froze.

The number of secondary drops per collision is sensitive to geometry and material of collision, even for drops of the same size. We quantify about 10 per collision, Schremb et al. (2018) observed 10s of collision for impacts on elevated ice surface, Rozhkov et al. (2002) observe 100s for drop impacts on steel disks at room temperature, as do Villermaux and Bossa (2011) for drop impacts on iron cylinders at room temperatures. Consequently, after addressing the above challenges and elevating the ice particle off the glass surface, which may be achieved simply by fixing the ice particle on a wire, further work is needed to investigate, more systematically, this new SIP mechanism over a range of experimental parameters, not limited to: supercooled drop sizes, supercooled water drop-to-ice particle size ratios, ice particle shapes, temperatures, drop height (and hence impact velocity), airflow, relative humidity conditions and chemical compositions of the supercooled water drop.’

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