AC Response to RC3

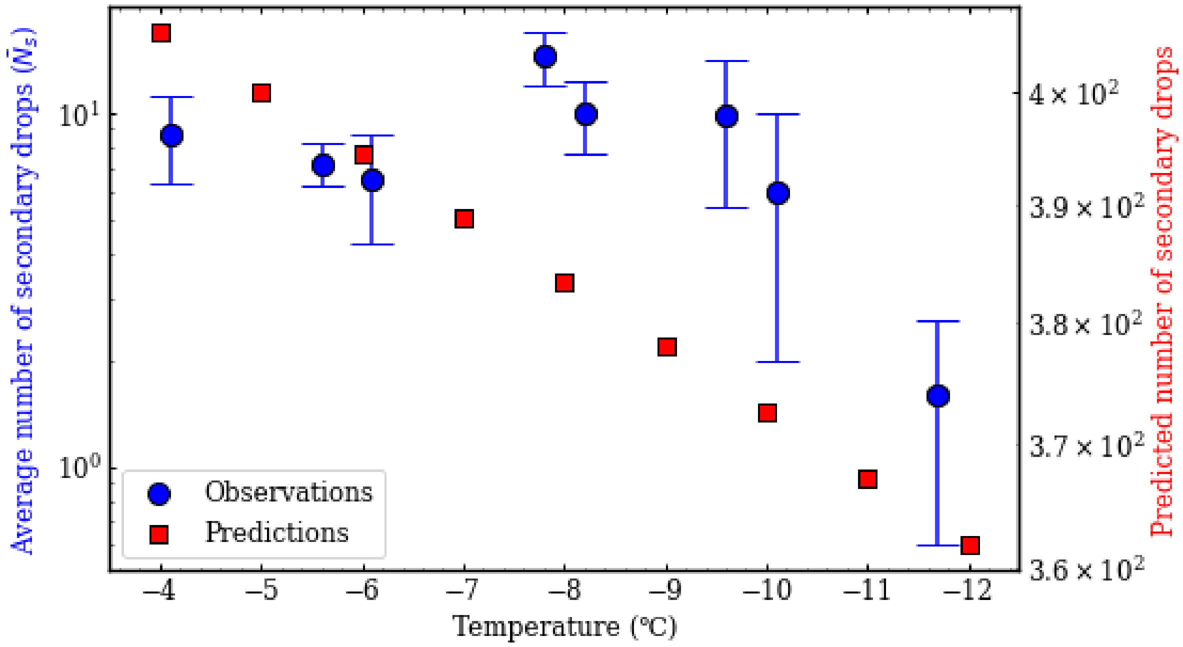
We thank Sylvia for taking the time to review our manuscript and appreciate the constructive feedback given.

RC: James, Phillips, and Connolly present experimental results on a secondary ice production (SIP) process involving liquid drop-ice crystal collisions. It is nice to see additional, and especially quantitative, laboratory results on SIP, and I support publication of the article. I feel, however, that several points should be elaborated and that some reorganization of sections would help with clarity.

Major Comments

- Given that a condensed version of the theoretical work in Phillips et al. 2018 was presented, I expected there to be some comparison of the observational results with this model. For example, can you calculate Ns as a function of temperature from the Phillips et al. model and overlay it on Figure 5? Can you say anything about the validity / assumptions of the model on the basis of these experiments?

AC: The figure below shows a comparison of the model with our experimental results.

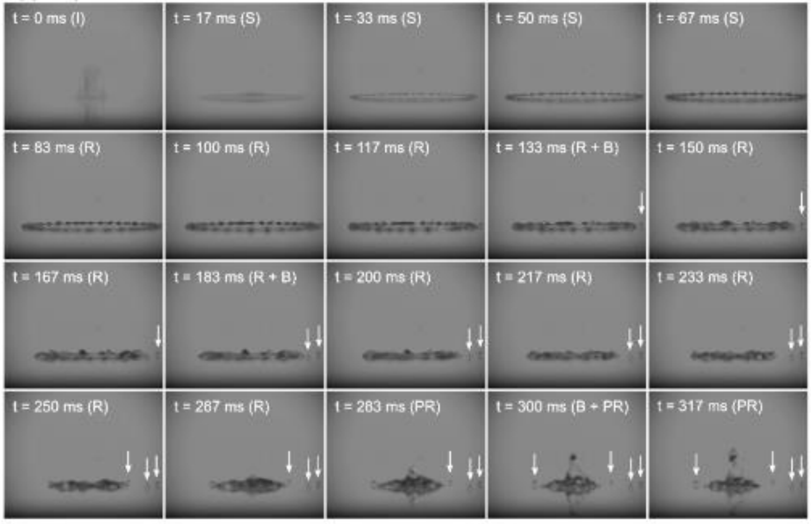


We have decided to remove Section 2 as we cannot currently quantify the majority of the secondary drops formed in our experiments. Therefore, we believe a comparison between our experimental results and the model presented in Phillips et al. (2018) does not provide anything beneficial at this time. We would like to provide quantification of the majority of secondary drops in the future through additional experiments and these will be compared with the model understand the validity/assumptions of the model.

RC: - I felt that the mechanistic discussion at the start of Section 5 would have been helpful prior to Sections 4.1 and 4.2, so that I had a better sense of what was physically happening in the experiments in Figs 2 and 3.

AC: Changed. We have also added some more detail about the drop impact phase in Figs 3 (originally 2) and 4 (originally 3). See below.

(a) Temperature: 23 °C



(b) Temperature: -5 °C

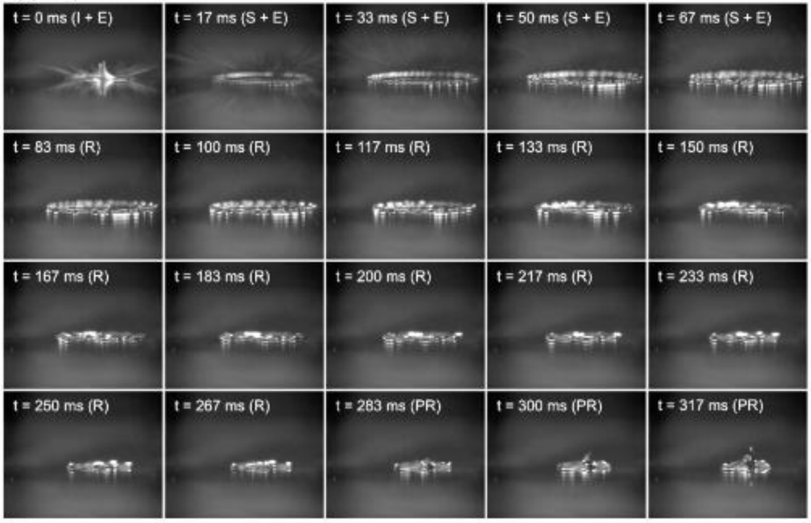


Figure 3. Frames from the high–speed camera configuration of a water drop impact on a glass slide when both water drop and glass slide are at (a) room temperature (23 °C) and (b) -5 °C. The impact phase (I), spreading phase (S), secondary drop formation/ejection during the spreading phase (E), retraction phase (R), secondary drop formation due to receding break-up (B) and partial rebound (PR) of the water drop are indicated in the frames. Arrows indicate secondary drop formation during the retraction phase of the water drop.

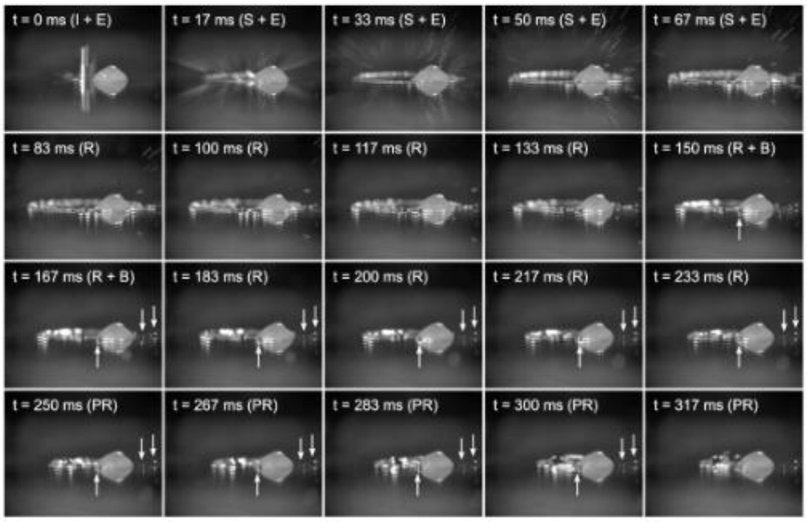


Figure 4. Frames from the high–speed camera configuration of a supercooled water drop impact on an ice particle when both drop and ice particle are at -5 °C. The impact phase (I), spreading phase (S), secondary drop formation/ejection during the spreading phase (E), retraction phase (R), secondary drop formation due to receding break-up (B) and partial rebound (PR) of the water drop are indicated in the frames. Arrows indicate secondary drop formation during the retraction phase of the supercooled water drop.

RC:- I appreciate that the limitation of the glass slide is acknowledged and secondary drop production in other setups is discussed (lines 159-171). But does the presence of the glass slide also mean that the mechanism shown in Figure 6 may need to be modified in the real atmosphere?

AC: Figure 6 (now Figure 2) represents a schematic of the experiments and is not representative of the process which would likely occur in the atmosphere. We have added the following words in bold in the caption to clarify:

'A schematic diagram of a supercooled water drop impact on an ice particle **on a glass slide**…'

RC: In particular, given that the secondary drops and frozen fraction have only been quantified in the retraction phase, should an equivalent retraction phase exist between two curved surfaces in relative motion?

AC: A retraction phase occurs over stationary curved surfaces, e.g. see Zhang et al. (2020). In fact, a curved surface is more likely to induce additional fragmentation mechanisms compared to a flat surface. In the atmosphere the ice particles are likely to be irregular in shape. We have included the below paragraph discussing the role of the shape of the ice particles:

'Another factor that will influence the generation of secondary drops is the ice particle shape. Currently, our ice particles have a pointed tip, as shown in Fig. 1, which is a typical shape formed when a liquid water drop is frozen on a cold substrate (Snoijer et al.,2012), but not representative of atmospheric ice particles. According to Phillips et al. (2018), who refer to this SIP mechanism `Mode 2', for it to occur, the supercooled water drops must have a diameter larger than 150 μm and the ice particle more massive still. In the atmosphere, ice particles which are larger than 150 μm are typically irregular in shape (Korolev and Sussman, 2000). A study by Zhang et al. (2020) shows that at room temperature, water drop impact on curved surfaces induce additional fragmentation mechanisms compared to flat surfaces. Therefore, we expect the irregular shape of an ice particle to affect the fragmentation mechanisms of the supercooled water drop and thus secondary drop formation. Exactly how irregular particle shapes will change the secondary drop formation is difficult to ascertain without further studies.'

In relative motion we would still expect to see secondary drop formation during the retraction phase as surface tension is responsible for the retraction of the drop. We do want to pursue investigating more realistic supercooled water drop collisions and ice particles in the future.

RC:- Section 5.1 read like introductory material, as it did not include discussion of any of the study's findings. It is valuable information, but I would integrate it into the introduction as general motivation to study SIP.
AC: We have integrated it into the introduction.

RC:- In place of Section 5.1, I think a new (small) "Atmospheric Implications" section or additional sentences throughout the Discussion / Conclusion would be helpful to discuss the atmospheric conditions under which the experiments are representative, i.e. In what regions / synoptic conditions / cloud systems, would ice particles of diameter 6 mm and raindrop of diameter 5 mm coexist? For what range of ice particle and raindrop sizes, does 5.2 m s-1 represent a realistic relative terminal velocity? Would it be possible to use the We and Re characterizing these experiments to identify regimes in in-situ data for which this mode 2 fragmentation could occur? etc.
AC: We don't think that supercooled water drops with diameters of 5 mm and ice particles with diameters of 6 mm are necessarily representative of cloud conditions. Practically, it was easier for us to work with supercooled water drops of this size. The intention of this paper is to demonstrate that the collisions of supercooled water drops with ice particles could be a new SIP. We have removed the Atmospheric Implications section and added the following paragraph at the end of the Discussion session.
'As a proof-of-concept investigation, we studied supercooled water drops with diameters of 5 mm and ice particles with diameters of 6 mm as larger sizes of supercooled water drops were easier to work with experimentally. While these sizes are not necessarily representative of cloud conditions, theoretically, this new SIP mechanism should occur where supercooled water drop diameters are > 150 µm and the ice particles more massive still. Supercooled water drops and ice particles are present within a variety of different clouds. For example, Hobbs and Rangno (1990) presented aircraft observations in small polar--maritime cumuli that displayed ice enhancement. Their discussion highlighted that ice enhancement proceeded in two stages. The first stage consisted of the formation of frozen drops, < 400 µm diameter, and small graupel particles, < 1 mm diameter. The second stage was characterised by the appearance of high concentrations of vapour--grown ice crystals in the upper regions of the cloud. A key finding of this series of papers was that high concentrations of small ice particles appeared simultaneously with frozen drizzle drops. Furthermore, Rangno and Hobbs (2001) showed that large supercooled drops were often a requirement for ice enhancement in moderately cooled Arctic stratiform clouds, and ice enhancement was often coincident with observations of large supercooled raindrops. Supercooled drizzle drops and raindrops are common in convective clouds (e.g. Crawford et al., 2012, Taylor et al., 2016), as are large ice particles. Hence, because there is a broad continuum of drizzle and raindrop sizes, where the larger drops freeze first, followed by accretion of the smaller unfrozen drops that the collision of supercooled water drops with ice particles more massive may be of importance in a wide range of clouds.'

Minor Comments

RC: Line 15 – "where subzero temperatures"
AC: Added.

RC: Lines 17-18 – "typically fall between 1 x 10-5 L-1 and 1 L-1 at temperatures T~-10 deg C" (looking at Fig. 1-10 from Kanji et al. 2017)
AC: Changed.

RC: Line 24 – "NWP models underestimate the concentrations of ice particles" – It would be nice to include an order-of-magnitude range for these underestimates.
AC: Upon reflection we think that this is probably a too broad a statement about NWP models underestimating ice particle concentrations and have removed it. Some NWP model can get the right order of magnitude, but sometimes for the wrong reasons. They often use parameterisations that generate too many crystals by primary ice nucleation (e.g. the Cooper, 1986 nucleation description), when secondary ice may be responsible. The issue with over estimating primary IN is that it then leads to a glaciation of the clouds and underestimation of supercooled liquid water, which then leads to an underestimation of secondary ice mechanisms that rely on supercooled water being present (there are other reasons for the underestimation of supercooled liquid water too, related to the resolution of the models). There are instances where NWP models underestimate ice particle concentrations, such as in Crawford et al. (2012), where observed concentrations of were ~ 100/L ice particles, but the NWP model, Weather, Research and Forecasting, estimates only up to 30 /L, and more broadly 5/L.

RC: Line 28 – "supercooled water drop diameters are < 13 um and > 24 um" In Hallett and Mossop 1974, both droplet sizes should coexist.
AC: Changed.

RC: Line 31 – Along with the temperature range for frozen droplet shattering, it would be worthwhile to include a droplet size range as well, since droplet size will be discussed later as a parameter of the current experiments (e.g. 280-350 um in Keinert et al. 2020)
AC: Added the following:
'A range in diameters of freezing supercooled water drops has also been investigated between laboratory studies from 4 μm to 1000 μm (see Table 1 of Korolev and Leisner, 2020, for a summary).'

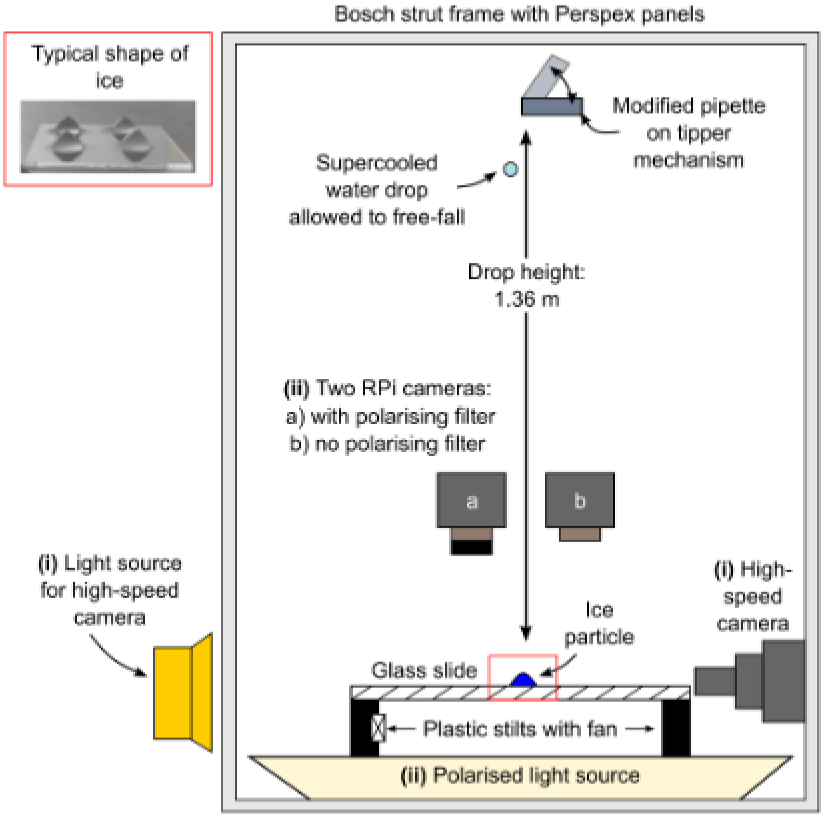
RC: Line 34 – I would suggest to rephrase as "the attention of laboratory studies has overwhelmingly focused on the rime-splintering...", since a growing body of recent work has look at breakup parameterizations (e.g. Hoarau et al. 2018, Sotiropoulou et al. 2020, Sotiropoulou et al. 2021, Dedekindet al. 2021, etc.)
AC: Changed.

RC: Line 52 – If you note that 'Mode 2' of frozen droplet fragmentation is studied, it would be helpful to know what 'Mode 1' is also.
AC: We have removed Section 2 so this is no longer in the paper. However, we have clarified 'Mode 1' and 'Mode 2' where the first reference to Phillips et al. (2018) is made:
'This SIP mechanism has been investigated via a theoretical study by Phillips et al. (2018) and referred to as 'Mode 2' as it involves collisions of supercooled water drops with more massive ice particles resulting in fragmentation of the supercooled water drop. Ice contained in some of the secondary drops was assumed to initiate freezing, yielding secondary ice fragments. By contrast, 'Mode 1' involved either collisions of supercooled water drops with less massive ice particles resulting in spherical freezing of the supercooled water drop or activation of immersed INPs, with a quasi-spherical outer ice shell that fragments. '

RC: Line 64 - “DEcrit” (not Dcrit). Also you have not yet defined the freezing stages when you mention “stage1 of freezing” here and again in Line 68.
AC: Thank you. We have removed Section 2 so this is no longer in the paper.

RC: Line 70 – “Finally, Phillips et al. 2018 hypothesised that Φ(T) = min[4f(t),1] such that Φ = 0.5 at -10 C” Stated like this, it sounds rather ad hoc. Perhaps an additional sentence can clarify where this form comes from or how it is constrained.
AC: Thank you. We have removed Section 2 so this is no longer in the paper.

RC: Line 94 – I do not know how important it is, but it was not clear to me what the “x-y translator (modified 3D printer)” was in the setup.
AC: The pipette could be moved to different positions using an x-y translator so that multiple drop collision experiments could be performed on one glass slide as, at the time, we didn’t have access to multiple glass slides of the correct size. In hindsight, we would opt for a fixed design. This detail is not important to the experiment, so we have removed mention of it from the text and simplified Fig. 1 to avoid any confusion. See below:



RC: Line 99 – Surface tension of the liquid water is presented as γ in line 61 and σ here; viscosity is presented as λ here and μ in the equation for the Reynolds number.
AC: Changed surface tension to σ throughout text and viscosity to μ.

RC: Lines 113-114 – It was not clear to me why the filament-like structures do not form when the colliding droplet spreads on the glass slide at room temperature. Is there a physical explanation for why this only occurs at colder temperatures?
We are also curious about why the subzero glass slide experiments exhibit filament-like structures upon impact. Drop impact on superhydrophobic surfaces at room temperature do not usually

appear to form filaments. As the temperature of water decreases the viscosity and surface temperature change so the appearance of filaments at supercooling could be something to do with the change in properties of supercooled water.

RC: Section 4.1 – Somewhere in this section or perhaps in the preceding Section 3, it would be helpful to have already referred to Table A1, so that it is clear from how many experiments the results come, e.g. only two glass slide collisions total were performed at 23 and -5 C?
AC: We only conducted a few experiments on the bare glass slide using the high-speed camera at 23 and -5 °C as this was for qualitative purposes to gain an understanding of the water drop fragmented and how the ice particle influenced this fragmentation. We have added the following sentence at the end of Section 3:
"We conducted 32 experiments using the RPicams configuration during quantification of the freezing fraction of secondary drops and the data is given in Table A1."

RC: Lines 133-134 – Was anything learned from the partial versus direct collisions? Does one or the other produce more secondary drops or higher frozen fraction? I guess there may be no robust difference, given the difficulty of performing these direct collisions.
AC: With the current dataset, we didn't observe a discernible difference between partial and direct collisions. It is something we would like to investigate further by adapting the setup to make it easier to get more direct collisions. We have expanded on this at the end of the conclusions with the following paragraph:

'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.

RC: Also was Figure 5 produced from all data (both partial and direct) in Table A1? This should be specified in the caption.
AC: Figure 5 was produced from all data. We have added the following to Fig. 5 caption: "Average data included both direct and partial collisions."

RC: Lines 136-138 – "The smaller secondary drops observed at impact ... were not observed." This seems like it may be an important limitation. Is there the possibility to improve RPicam resolution in future work? This should be mentioned in the conclusions / future work if so.
AC: We would more likely try to quantify the smaller secondary drops formed during impact/spreading phase with the high-speed camera. We have added the following sentences in the conclusions/future work:

'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 100 m 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.'

RC: Line 148 – “Surface tension and viscosity forces were considered negligible during the spreading phase of the drop” I am confused by this statement. Where / in which calculations are these forces being considered negligible?

AC: These forces are considered in the Weber and Reynolds number calculations given in the second to last paragraph of Section 2. The Weber number relates the inertia to surface tension, and the Reynolds number relates inertia to viscosity. When We and Re numbers are over a critical value then inertia is the dominating force during spreading, and surface tension and viscosity are considered negligible.

We’ve changed the sentence to: “Surface tension and viscosity forces were therefore considered negligible during the spreading phase of the drop”

RC: Lines 150-151 – I have not seen the prompt-type / corona-type splash terminology before; I would define these terms more completely from the citations in these lines.

AC: In prompt splashing, secondary drops are formed from the break-up of the advancing thin film. Whereas, in corona splashing the thin film forms a bowl-like structure which then breaks up to form secondary drops. In general it is difficult to discern between these mechanisms (see paper by Josserand and Thoroddsen, 2016) and our set-up is not designed to study this splashing mechanism. As adding this terminology will probably create more confusion, we have decided to remove this sentence from the manuscript.

RC: Figure 7 – Are the top versus bottom panels also with and without the polarising filter?

AC: Yes. We’ve added the following sentence in the caption:
“The top panel shows frames from the RPicam with no polarising filter and the bottom panel shows frames from the RPicam with a polarising filter.”

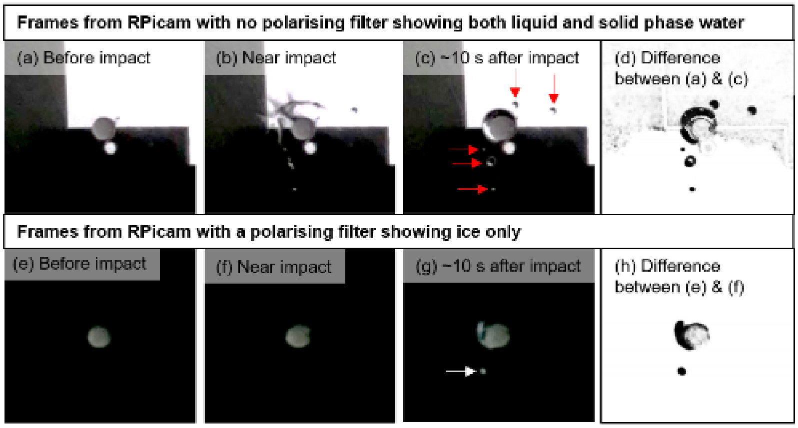


Figure 5. Selected frames from the impact of a supercooled water drop on an ice particle at -4°C using the RPicams configuration. Frames (a)–(c) before, at and $\sim$10 s after impact using the camera with no polarising filter. Red arrows in (c) indicate the number of secondary drops formed. Frame (d) shows the difference between (a) and (c). Frames (e)–(g) before, at and $\sim$10 s after impact using the camera with a polarising filter. The white arrow in (h) indicates the frozen secondary drop. Frame (h) shows the difference between (e) and (g).

RC: Line 176 – “T less than equal to -11 deg C”

AC: Added.

RC: Lines 180-187 – I find the arguments here difficult to follow. The takeaway is that temperature dependence of frozen fraction is caused by a liquid-ice interaction time scale? Could the authors reword somehow for clarity?

AC: We’ve reworded the paragraph to the following:
‘We observed a decrease in the number of secondary drops formed during receding break-up as

temperatures decreased below -8 ° C. Figure 7 shows the frames after a supercooled water drop impact with an ice particle for the experiments between -11 °C and -12 °C which was the range where the smallest number of secondary drops formed. At these temperatures, the supercooled water drop froze either during the spreading phase or in the early stages of the retraction phase. As the growth velocity of ice in supercooled water increases with decreasing temperature, e.g. at -2 °C it is around 0.2 cm s^{-1}, whereas at -10 °C it is around 5 cm s^{-1} (see Pruppacher and Klett, 1997, chapter 16), which may explain why a decrease in secondary drops was observed. We believe the decrease in secondary drop formation at temperatures below -8 °C may be due to the artificially flat geometry presented by the glass slide and to the large size of the incident drop, both factors which prolonged the interaction time between the supercooled water drop and ice. For example, the supplementary videos from Schremb et al. (2018) showed several secondary drops forming at -14 °C after impact on an elevated ice target, more than we observed at our lowest temperature of -12 °C.'

RC: Line 189 – "We believe that the freezing fraction of the secondary drops is independent of the number of drops formed." Is there a reason for this belief? I would expect temperature dependence to dominate also, but I could also imagine that when a fixed fraction of the colliding droplet mass produces secondary drops, and more such secondary drops form, they are smaller and freeze faster..?
AC: We have removed this paragraph in part due to another referee's comment.

References

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