AC Response to RC2

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

RC: I enjoyed reading this paper, which presents significant new experimental results relating to secondary ice processes, and is certainly worth publishing. I have a few minor questions and suggestions for making the paper a bit stronger (see below).

Active between -3 °C ≤ T ≤ -8 °C, rime–splintering occurs when supercooled water drop diameters are < 13 µm or > 24µm (Hallett and Mossop, 1974; Mossop and Hallett, 1974; Mossop, 1978) Maybe I misremember the Mossop 1978 paper, but should the condition on droplet sizes here be "< 13 µm and> 24µm" rather than or?
AC: Changed.

Section 2

RC: Perhaps it could be useful just to elaborate a little bit on these formulae. Maybe showing a figure with graphs of Ns vs DE; Phi vs T; f(T) would help make the general behaviour clearer?
AC: We have removed Section 2 (Theory) due to comments from another reviewer, so the theory is no longer described in this paper.

RC: The typical freezing shape of the ice particle is shown in Fig. 1.

Is the shape of the ice particle likely to be a relevant factor here? How could you find out? What is it likely to be in the atmosphere?
AC: Added section in discussion:
'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.'

RC: the impact velocity (V0) for all experiments was 5.2m s−1

it's worth pointing out in the text that this is below terminal velocity for a 5mm drop(which would be closer to 9 m/s). However in the real atmosphere the ice particle would be moving as well, so the differential velocity may be more realistic than it might initially appear.
AC: We've added the following sentences in Section 2:
'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.

RC: In fluid dynamics, the Weber number, We = ρDV02/σ, and Reynolds number, Re = ρDV0/μ, are used to relate inertial forces to interfacial and viscous forces, respectively. Taking into account the temperature dependent values of surface tension and viscosity of the supercooled water between -4 °C ≤ T ≤ -12 °C, the We and Re number ranges obtained were 1747 ≤ We ≤ 1772 and 8781 ≤ Re ≤ 12240, respectively.

I think in both cases here, it would be good to clarify what We and Re refer to–or more specifically where these inertial, viscous, and interfacial forces are acting. Often in cloud physics we think about the inertial, viscous in the air surrounding the drop, while here (I think) you are considering them within the water

AC: We've added the following words highlighted in bold:
'…used to relate inertial forces of the **fluid** to its interfacial and viscous forces respectively.
And, added the following sentences to clarify:
'In this case, the fluid is the supercooled water drop. The inertial force is from the initial impact velocity of the supercooled water drop, and the interfacial (surface tension) and viscous forces are properties of the supercooled water drop.'

RC: Is it obvious what the length scale and velocity scale in We and Re should be? You have chosen V0 for the velocity scale, so that implies the water fluid parcels of interest are moving at this velocity. So are you considering the downward motion of the liquid water at the moment of impact on the ice particle? Or the lateral velocity of the liquid water as it spreads out? (are these velocity scales comparable?).

AC: In drop impact experiments it is typical that the length scale should the diameter of the initial drop before impact and the velocity scale is the normal impact velocity of the drop on the surface (e.g., see a review of drop impact by Josserand & Thorodssen 2016). So yes, this is the downward motion of the liquid water at the moment of impact on the ice particle, not the lateral velocity as the liquid water as it spreads out.
We have added the following to word in bold to clarify:
'…**initial** supercooled water drop diameter **before impact** (D)'
'…the **normal** impact velocity (V0)…'

It's difficult to say whether the impact and lateral velocity scales are comparable. Zhang et al. (2021) show in their Fig 9(a) that there is a linear relationship between the impact velocity and the lateral (spreading) velocity for water drops on flat surfaces with varying degrees of wettability. For a superhydrophobic surface (similar to our glass slide) with an impact velocity of 5 m/s the spreading velocity is ~9 m/s. However, our supercooled water spreads over an ice particle and the glass slide which will likely reduce the velocity.

RC: For the length scale, it's not obvious what to choose, when you have a liquid spreading over a solid surface. The depth of the water coating? D is probably not an unreasonable choice, but maybe you can make the argument a bit more explicit somehow. Again, it all comes down to what aspect of the flow of the water you are trying to characterise.

AC: The length scale typically used in drop dynamics is the diameter of the initial drop before impact. See response to question above for clarification made in the text.

RC: Section 3 –you used a high speed camera. What exposure time was used? It seems from the images like the splash itself (t=0) is quite blurred. Was this limited by the illumination?

AC: The exposure time was 929.36 µs. We believe the frames appear blurred because of difficulties in knowing where the drop would impact, which made it difficult to focus the camera.

RC: For figs 2,3,4 I did wonder whether adding some slightly more detailed description of what's happening in the various frames would help the reader interpret what they are seeing. It took me a while to get a sense of what was happening. Or maybe some extra annotation on the figures themselves?

AC: We've annotated Fig 3 (originally Fig 2) and Fig 4 (originally Fig 3) with letters to indicate the impact phase, spreading phase, secondary drop formation/ejection during the spreading phase, retraction phase, secondary drop formation due to receding break-up and partial rebound. Plus a clarifying sentence highlighted in yellow in the caption. See below:

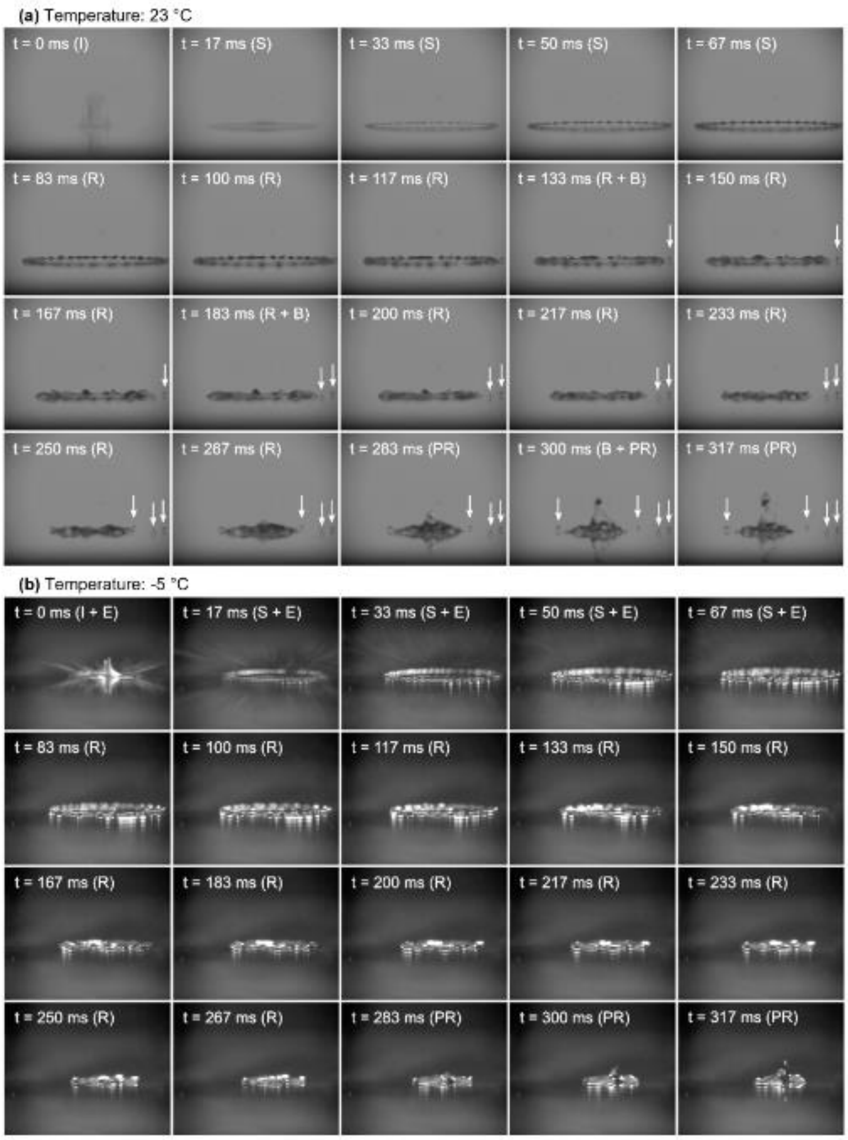


Figure 3. Frames from the high–speed camera configuration of a water drop impact on a glass slide when both water drop and glass slide are at (a) room temperature (23 °C) and (b) -5 °C. The impact phase (I), spreading phase (S), secondary drop formation/ejection during the spreading phase (E), retraction 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.

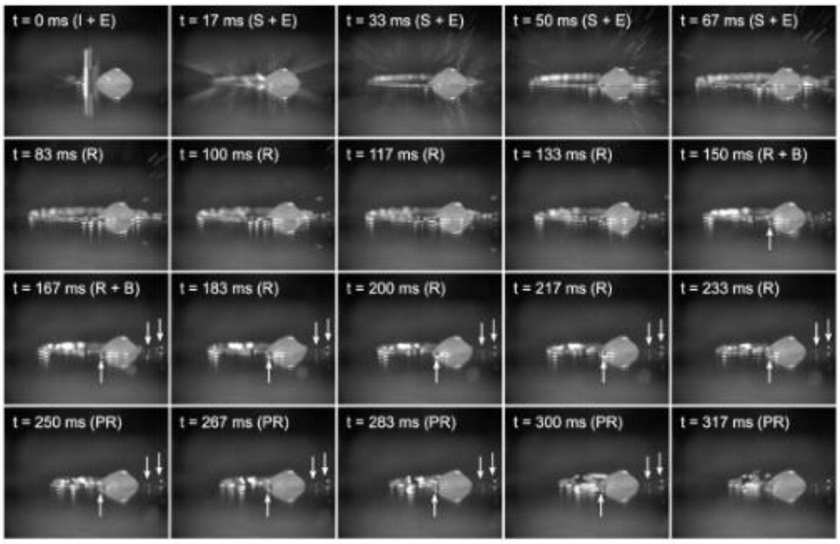


Figure 4. Frames from the high–speed camera configuration of a supercooled water drop impact on an ice particle when both drop and ice particle are at -5 °C. The impact phase (I), spreading phase (S), secondary drop formation/ejection during the spreading phase (E), retraction phase (R), secondary drop 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.

In Fig 4 we've added before impact, near impact, ~10s after impact and difference between before and after impact to the frames. We've also indicated that the top panel is from the RPicam with no polarising filter showing both liquid and solid phase water and that the bottom panel is from the RPicam with a polarising filter showing ice only. See below:

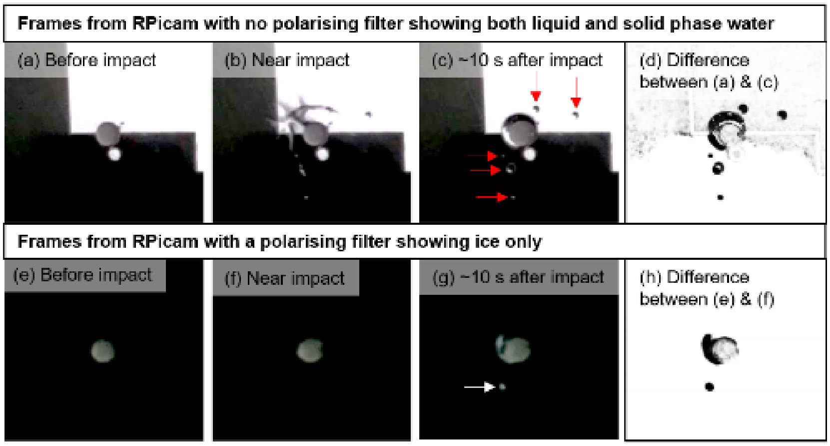


Figure 5. Selected frames from the impact of a supercooled water drop on an ice particle at -4°C using the RPicams configuration. Frames (a)–(c) before, at and ∼10 s after impact using the camera with no polarising filter. Red arrows in (c) indicate the number of secondary drops formed. Frame (d) shows the difference between (a) and (c). Frames (e)–(g) before, at and ∼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: Discussion -You mention the influence of the glass slide, and I agree the presence of the slide itself is definitely worth discussing. Another factor I can think of here is that the ice particle is effectively in a fixed vertical position, while in the atmosphere the ice particle is in free fall, and when the drop hits it, then the ice particle can move in response to that –so some of the drop's momentum can be carried to the ice particle. Would that change the way the water flows over the ice particle, and freezes?

AC: We thank the reviewer for making this good point. We think that the ability of the ice particle to move upon collision will have some effect on the way it fragments, and it is certainly something we would like to explore in the future. However, we don't really know whether it will increase or decrease the fragmentation of the supercooled water drop without first investigating the fragmentation mechanism of the supercooled water drop without the glass slide. We know from a study by Zhang et al. (2020) that curved surfaces can cause additional fragmentation mechanisms. Another factor will be the freezing mechanism which we also need to investigate further. If freezing is initiated by the formation of the ice dendrites from contact with the ice particle, which can occur on the millisecond scale, then the ice dendrites will still be able to propagate through the supercooled water drop even if contact time is reduced. If freezing is via mechanical agitation/shock then the momentum transfer to the ice particle from the supercooled water drop will likely have a more significant effect on freezing. We have added the following paragraph below:

'In addition, the ice particle in our experiments is in a fixed position on the glass slide, whereas, in the atmosphere, the ice particle is in free fall. When the faster-moving supercooled water drop collides with the ice particle, the ice particle will move in response to the collision, likely affecting the formation of the secondary drops and their subsequent freezing. However, currently, it is difficult to ascertain how this will influence secondary drop formation and freezing without further investigations into the mechanisms of secondary drop formation on an elevated ice particle.'

RC: In figure 5 I think it's important to clarify what the error bars represent in the caption, and in the text. Is it the variation from one experiment to the next, in the "same" conditions? Or is it the uncertainty on the mean value?

AC: Added the following to caption:
'The error bars represent the standard error in the temperature intervals which are listed in Table A2 & A3.'

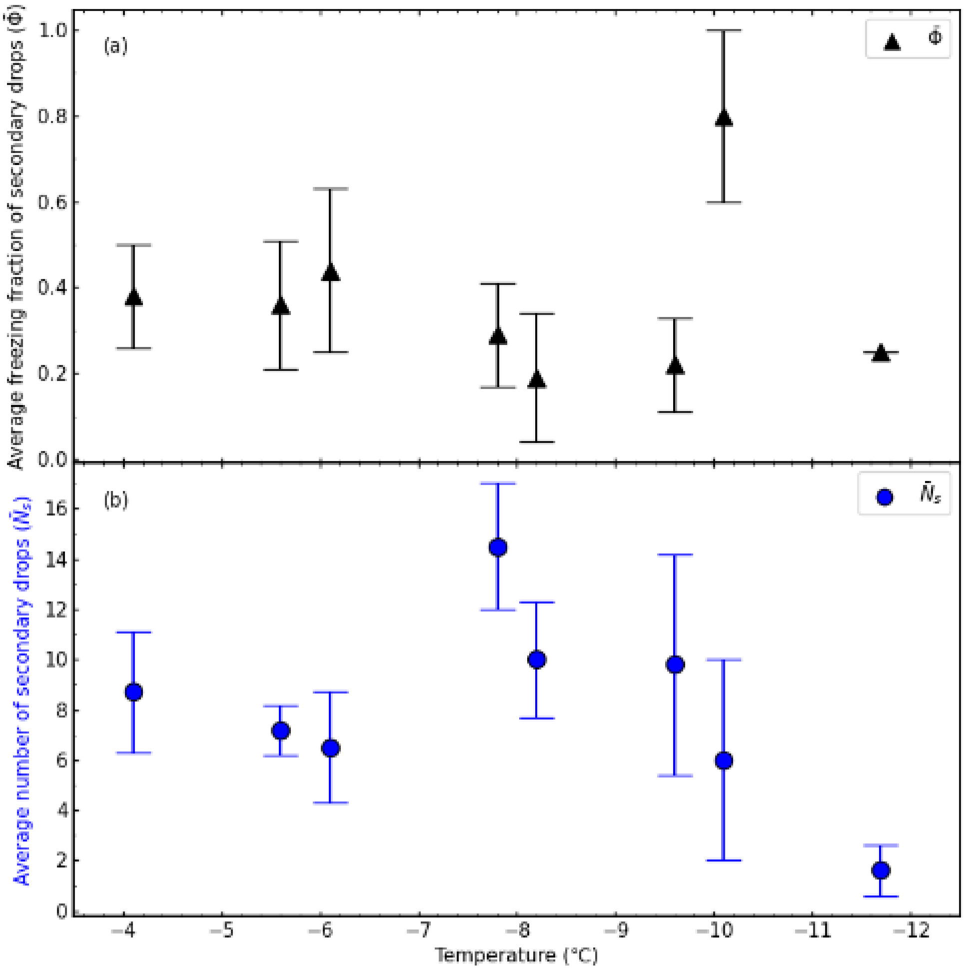
RC: Connected to this is Table A2 –the values of phi, sigma, and sigma_phi_bar are all quoted to the nearest 0.1, which seems a bit coarse. Might be worth 1 extra significant figure?

AC: Added another significant figure and updated Fig. 5 to reflect this change. See below:

Table A2:

T interval (°C)		$\bar{T}$ (°C)	$\bar{\Phi}$	σ	n	$\sigma_{\bar{\Phi}}$
-3.8	-4.3	-4.1	0.38	0.26	5	0.12
-5.3	-5.8	-5.6	0.36	0.34	5	0.15
-6.0	-6.1	-6.1	0.44	0.38	4	0.19
-7.7	-7.8	-7.8	0.29	0.17	2	0.12
-8.0	-8.5	-8.2	0.19	0.30	4	0.15
-9.4	-9.9	-9.7	0.22	0.19	3	0.11
-10.0	-10.1	-10.1	0.80	0.28	2	0.20
-11.3	-11.9	-11.7	0.25	0.00	2	0.00

Figure 5:



RC: The number of experiments is fairly small, given the variability in phi that's shown. I'm guessing these are quite time consuming to conduct and analyse. Perhaps you can discuss that a bit? In general I would enjoy seeing an expansion of the future work in section 6 to talk about how the experiment could be improved and elaborated. Likewise saying "no quantification of the freezing fraction of the secondary ice drops [from the jet of smaller droplets] can currently be made" is fine, but it would be good to discuss what you would need to do to quantify it, or study it in more detail.
AC: We have added the following paragraphs:

'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.'

References

Gunn and Kinzer, The terminal velocity of fall for water droplets in stagnant air, J. Atmos. Sci., 1949, 6, 243 – 24.

Josserand and Thoroddsen, Drop Impact on a Solid Surface, Annu. Rev. Fluid Mech., 2016, 48, 365-391.

Korolev and Sussman, A Technique for Habit Classification of Cloud Particles, J. Atmos. Ocean. Technol., 2000, 17, 1048 – 1057.

Locatelli, J. D. and Hobbs, P. V.: Fall speeds and masses of solid precipitation particles, J. Geophys. Res., 1976, 79,2185–2197.

Phillips et al., Secondary Ice Production by Fragmentation of Freezing Drops: Formulation and Theory, J. Atmos. Sci., 2018, 76, 3031–3070.

Rozhkov et al., Impact of water drops on small targets, Phys. Fluids, 2002, 14, 3485-3501

Schremb et al., Normal impact of supercooled water drops onto a smooth ice surface: experiments and modelling, J. Fluid Mech., 2018, 835, 1087–1107.

Snoijer and Brunet, Pointy ice-drops: How water freezes into a singular shape, Am. J. Phys., 2012, 80, 764–771.

Thoroddsen et al., Micro-splashing by drop impacts, J. Fluid Mech., 2012, 706, 560–570.

Villermaux and Bossa, Drop fragmentation on impact, J. Fluid Mech., 2011, 668, 412—-435.

Zhang et al., Asymmetric splash and breakup of drops impacting on cylindrical superhydrophobic surfaces, Phys. Fluids, 2020, 32, 122 108.

Zhang et al., Effect of wettability on droplet impact: Spreading and splashing, Exp. Therm. Fluid Sci., 2021, 124, 110369.