<u>Review of "A bin-microphysics parcel model investigation of secondary ice formation in an idealised shallow convective cloud" by James and colleagues</u>

Verdict

This paper is lucid and intriguing. Minor modifications to the text are needed before it is published.

Major comments

It would be a good idea to provide a precise definition of the meaning, values and units of all symbols in a table. This would clarify things when the text does not specify all the needed details.

Regarding the treatment of Mode 1 of raindrop-freezing fragmentation by Phillips et al. (2018), it should be emphasized in the introduction of the present paper by James et al. that numbers of secondary ice fragments were based only on lab observations of freezing drops in free-fall. No observations of electrodynamically levitated drops were used. Thus, Phillips et al. (2018) were vindicated, since it has since been shown that free-fall is crucial for fragmentation. Regarding the treatment of Mode 2, I think there is still wide experimental uncertainty introduced by the treatment of numbers of secondary drops emitted per impact with a larger ice surface. Sensitivity tests should be done to explore the realm of uncertainty. Anyway, raindrop-freezing fragmentation does not appear to be very important in most recent studies using the Phillips et al. (2018) scheme.

I think a more serious issue of the present paper is the lack of dependency on mean droplet size of the H-M process of rime-splintering. This was seen in lab studies by Hallett and Mossop, and was also consistent with aircraft observations of convective clouds by Harris-Hobbs and Cooper (1987) and Blyth et al. (1993). Yes, those studies involved artificial shattering biases on optical probes, but the correction merely introduces a systematic bias and the qualitative correlation with H-M conditions still likely applies. Can the simulations be re-done with a factor depending on mean droplet size somehow, including this in Eq (2), to account for the numbers of cloud-droplets > 24 microns ?

Thus, in view of the empirical uncertainty about splashing in Mode 2 of randrop-freezing fragmentation, it would be prudent for James et al. to perform sensitivity tests of their cloud simulations by raising the onset threshold of DE (0.2 currently) by a factor of 10 or 30. Also, sensitivity tests on reducing the constant of proportionality by an order of magnitude would also explore the range of experiment uncertainty. Error-bars reflecting this uncertainty from Mode 2 are needed on the control simulation plots of IE ratio.

Comments about the Review by Kiselev about treatment of Mode 2

I am grateful for the lucid comments about raindrop-freezing fragmentation during drop collisions with a larger ice particle (Mode 2) from the reviewer, Kiselev. It is illuminating to see papers published in colloidal science and fluid dynamics journals of relevance to splashing.

Curiously, in a sense, the two papers cited by the review by Kiselev qualitatively confirm the validity of the general theoretical approach for the treatment of Mode 2 by Phillips et al. (2018) because they show that the splashing onset condition is related to the Weber number. This Weber number is identical to the dimensionless energy ("DE") of the 2018 scheme, except for a factor of 12. So, the main import of Kiselev's criticism of the treatment of Mode 2 concerns uncertainty in the empirical estimation of parameters by Phillips et al. as applied in the present paper by James et al.

I would like to clarify one or two details about Kiselev's critique of the treatment of Mode 2 of raindrop-freezing fragmentation by Phillips et al. (2018).

First, observations of drop-drop collisions were not the only lab observations used to constrain the splashing scheme in the treatment of Mode 2. There was also comparison with the observations by Levin and Hobbs (1971) of drops (2.5 mm, 4.2 m/s) falling on a rough copper hemisphere. Levin and Hobbs observed about 150 splash-drops produced per collision. They reported that there was onset of splashing at about 0.5 m/s impact speed. Thus, the dimensionless energy (DE = initial collision kinetic energy divided by drop surface energy) per drop impact was $500\ D\ v^2\ /(6 \times 0.073\ )$ = 50 (for about 150 splash-droplets per collision) and 0.7 (for onset of secondary droplet emission or splashing). This critical onset value of DE (0.7) is between one and two orders of magnitude lower than the threshold claimed by Kiselev in the review, who analyzed the Sykes et al. and Charalampous data.

Moreover, Levin and Hobbs observed that target roughness is crucial for the production of secondary drops, and presumably must have selected a rough copper sphere with the aim of better representing the actual roughness of ice precipitation particles, in the quest to study charge separation in clouds. Inspection of the results from Levin and Hobbs seem to suggest an approximate proportionality between number of secondary droplets and DE. Intriguingly, Levin and Hobbs report observing that a "crown" of fluid would be produced on impact, with the secondary droplets being emitted from the crown (e.g. by jets). On very smooth surfaces, they observed that there would be no crown, and presumably little or no secondary droplet emission (no splashing).

Thus, although Phillips et al. (2018) in the text cited the drop-drop collisions as the source of the critical DE value (0.2), their simultaneous use of the Levin and Hobbs observations of drops on a rough copper hemisphere also approximately support this value (0.7, the same order of magnitude as 0.2). In fact, Phillips et al. used chiefly the Levin and Hobbs results for the most important parameter of the scheme, namely the coefficient of proportionality between drop number and excess DE, rather than the drop-drop collision data.

Secondly, the review by Kiselev claims that factors of surface roughness, water adsorbed onto the surface and curvature of the surface were not accounted for in the treatment by Phillips et al. (2018). Is this claim true ? Yes and no. Naturally, these factors are greatly sensitive, as observed by Levin and Hobbs for roughness, as noted above. Yet, Levin and Hobbs knew that these factors were influential and designed their copper sphere experiment so as to be representative for collisions with ice precipitation particles in natural clouds (their copper sphere was dry, as with ice during dry growth by riming, and rough, as is true of most riming; their ratio of the radii of curvature between the incident drop and target was about 10%, a plausible order of magnitude for Mode 2 collisions in natural clouds). Their focus was on charge separation in electrified clouds. So the average conditions of these three factors may be implicitly factored into the Levin and Hobbs results and hence into the Phillips et al. (2018) scheme.

By contrast, the two laboratory studies cited by Kiselev involve observations of extremely smooth spherical surfaces. Sykes et al. (2022) used "smooth untreated borosilicate-glass substrates" while Charalampous and Hardalupas (2017) used similarly smooth glass (smoothness of 35 nm). While such studies are theoretically illuminating (they nicely demonstrate that the ratio of drop to target radius of curvature is a sensitive parameter rather than the target radius per se), the observations of Levin and Hobbs about the crucial role of target roughness for the splashing process indicates that they cannot be used quantitatively for constraining Mode 2.

Finally, Schremb et al. (2017) observed splash-droplets from drops of 3 mm in diameter falling at about 2 m/sec on a larger ice target, with a reported Weber number of about 200 (DE value of about 15). Although they did not report splash-drop numbers, analysis of the published photos in their paper and my personal communication with Schremb shows that dozens of splash-drops were emitted per collision. This suggests that for ice, the critical DE value for onset of splashing must be much less than about 15, contrary to the lab results cited by the review. Perhaps onset of splashing in Schremb's experiment would be expected to have occurred at a critical DE value of about unity, given the proportionality noted above inferred from the Levin and Hobbs results.

On the one hand, the Mode 2 treatment by Phillips et al. might have been an over-estimate, since Levin and Hobbs (1971) observed 10 times more splash-drops for a rough curved copper surface than for similar values of drop size, DE and Weber number compared to Schremb et al. with a larger ice surface (a vertical columnar shape). Alternatively, one could argue that the roughness of the surface is more realistic in the case of Levin and Hobbs than for Schremb et al., such that Schremb et al. may have observed too few (unreported) splash-droplets per collision if their ice surface was smoother than for ice precipitation in natural clouds. There is much uncertainty still, but roughness appears to be crucial.

In summary, there seems to be variability of the splashing among drops among different types of larger solid surface for different lab experiments, even for similar macroscopic dynamics of collision. The observations by Levin and Hobbs of drops falling on a rough copper sphere were designed for representativeness when studying charging in collision of ice particles in natural clouds. They found target roughness is crucial for secondary droplet emission (and for splashing in Mode 2). Ice precipitation in natural clouds is almost instantly roughened by riming. Hence, the smooth-glass-target observations cited in the review by Kiselev, though useful theoretically for the physics of splashing, must greatly under-estimate the real splashing in natural clouds during Mode 2 by ice precipitation. The smooth-glass-target observations cannot be supposed to be more reliable than observations by Levin and Hobbs.

Of course, future lab experiments on this topic will be invaluable to elucidate Mode 2 of raindrop-freezing fragmentation in a representative manner.

Specific comments

Line 90: More details of the model description are needed in the text. What are the microphysical species represented ? What processes of growth are treated ? Is raindrop-freezing treated ?

Line 120: Why is the observed dependency on cloud-droplet size not represented in Eq (2) ? Drops larger than 24 microns are needed for rime-splintering, as shown by Hallett and Mossop's later work.

One can simply multiply the formula shown in Eq (2) by an extra factor that increases with mean droplet size from zero to unity over a certain range.

Line 148: It might be good to clarify that Mode 2 is for raindrops.

Line 155: Need to say that NM2 is the number of fragments per drop accreted.

Line 167: It is unclear what is meant here: "We ran all possible combinations using the methodology described by Montgomery (2013); Teller and Levin (2008). For k factors there are 2^k different combinations". What is k ? What factors are being mentioned ? What combinations are being mentioned ?

Line 170: as noted above, there is a need to perform sensitivity tests with respect to the number of splash droplets per collision predicted for Mode 2 of raindrop-freezing fragmentation. This is due to the experimental uncertainty.

Line 180: There is over-loading of the terms "Mode 1" and "Mode 2" in Table 3 to refer to aerosol modes. Can a different label be used for aerosol modes (e.g., Modes A, B, C) ?

Line 248: A problem with the DeMott (2010) parameterization is that there is no dependency on aerosol chemistry and that on size is only represented in a basic way, using a threshold. There is no direct dependency on dust surface area. If the aerosol chemical composition was observed for the simulated cases, then other schemes are possible (e.g. Phillips et al. 2008, 2013).

Line 298: the plural of "maximum" is "maxima", as it is a Latin word.

Figure 5: I think it would be wise to include an error-bar in the simulations arising from errors in the parameters of Mode 2, in view of the discussions above and by Kiselev's review. Also, a logarithmic y-axis would be more appropriate in view of the uncertainty.

Line 568: the sentence does not make grammatical sense. Did Sotiropolou et al. explicitly treat Mode 2 ?

Line 578: Need to explain the mechanism for how high IN concentrations in the model suppress SIP and coalescence. Do they cause subsaturation with respect to liquid, from vapour growth of ice, and is this what curbs the droplet growth, inhibiting raindrop-freezing fragmentation ? Or is the liquid depleted by riming of the primary ice ?

As argued by Yano and Phillips (2011), there is an upper limit to the ice concentration that depends on vertical velocity and temperature, corresponding to the onset of subsaturation with respect to liquid. When the cloud-liquid evaporates and the cloud becomes ice-only, all SIP ceases. Waman et al. (2021) nicely showed this happening, so that paper needs to be cited. Could the lack of SIP when INPs are numerous be due to this upper ceiling being reached by the primary ice, or even approached ?