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

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

RC: Verdict

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

Major comments

RC: 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.
AC: We have put the list of symbols in Appendix B.

Table B1: List of symbols

Symbol	Description	Value and units
A	Number density of the breakable asperities in the region of contact	-
c_w	Specific heat capacity of liquid water	4200 J kg^{-1} K^{-1}
C	Asperity fragility coefficient	-
D_a	Diameter of aerosol particle	m
D_d	Diameter of drop in mode 2	m
$D_{a,m}$	Median aerosol particle diameter of mode	(m)
$D_{i,m}$	Median aerosol particle diameter of mode A, B or C	nm
D_s	Diameter of the smaller colliding particle in ice-ice collisional breakup	m
DE	Dimensionless energy	-
DE_{crit}	Critical value of dimensionless energy for onset of splashing	0.2, unless otherwise stated
f	Mass fraction of a drop frozen by the end of stage 1 freezing	-
f_{RS}	Function of rime splintering	-
F	Interpolation function for the onset of fragmentation	-
$K_{0(CB)}$	Collisional kinetic energy at impact	J
L_f	Specific latent heat of freezing	3.3×10^5 J kg^{-1}
m_r	Mass of rime	kg
m_1, m_2	Mass of colliding ice particles	kg
m_d	Mass of drop in mode 2	kg
m_i	Mass of ice particle in mode 2	kg
N	Number density of aerosol particles	kg^{-1}
N_{CB}	Number of ice particles due to ice-ice collisional breakup	-
N_i	Total number density of aerosol particles of mode A, B or C	cm^{-3}
N_{M1T}	Total number of ice particles due to mode 1	-
N_{M1L}	Total number of large ice particles due to mode 1	-
N_{M2}	Number of ice particles per drop accreted due to mode 2	-
N_{RS}	Number of ice particles due to rime splintering	-
N_T	Total number of aerosol particles of mode	kg^{-1}
R_{FL}	Rime fraction of the larger colliding particle	-
R_{FS}	Rime fraction of the smaller colliding particle	-
t	Time	s
T	Freezing temperature of water drop	°C
T_0	Value of T at maximum of Lorentzian for Eq.5	°C
T_{B0}	Value of T at maximum of Lorentzian for Eq.6	°C
v_1, v_2	Fall speed of colliding ice particles	m s^{-1}
v_d	Fall speed of drop in mode 2	m s^{-1}
v_i	Fall speed of ice particle in mode 2	m s^{-1}
w	Updraft speed	m s^{-1}
α	Equivalent spherical surface area of the smaller colliding particle	m^2
β	Parameter in Eq. 5	K^{-1}
β_B	Parameter in Eq. 6	K^{-1}
γ	Parameter related to riming intensity	-
γ_l	Surface tension of liquid water	0.073 J m^{-2}
ζ	Intensity of Lorentzian in Eq.	-
ζ_B	Intensity of Lorentzian in Eq.	-
η	Half width of Lorentzian in Eq. 5	°C
η_B	Half width of Lorentzian in Eq. 6	°C
κ	Hygroscopicity parameter	0.61
ρ_L	Density of the larger colliding particle	kg m^{-3}
σ_g	Standard geometric deviation of the logarithmic distribution	-
σ_i	Standard geometric deviation of the logarithmic distribution of mode A, B or C	-
Φ	Probability of any drop in the mode 2 splash containing ice	0.3, unless otherwise stated
Φ_s	Aspect ratio of the smaller colliding particle	-
Ω	Interpolating function for the onset of fragmentation	-

RC: 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.

AC: Regarding the Mode 1 parameterisation being based on drops in free fall only, we have added *'Based on the available laboratory literature of drops in free fall only…'*

Regarding Mode 2, we have added the following sentence in the introduction *'However, large experimental uncertainties exist around the treatment of the number of secondary drops emitted per impact with a more massive ice surface.'*

RC: 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 ?

AC: We acknowledge that there is a dependence of the rime-splintering process on the mean drop size. This has been communicated by Harris-Hobbs and Cooper (1987), in addition to Mossop (1976). The mean drop size dependence is not included here. The justification is twofold:

(1) often there are processes that lead to broadening on the drop size distribution that parcel models do not typically capture (e.g. entrainment / mixing)
(2) The case where the rime-splintering process had an appreciable effect was the warmer / deeper cloud simulation with natural aerosol (Fig. 10a). As shown in the supplementary material (Fig. S19a) the droplet sizes are > 24 microns in this case.

RC: Thus, in view of the empirical uncertainty about splashing in Mode 2 of raindrop-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.

AC: We have completed sensitivity studies on DEcrit for a factor of 10 and 30. See response to Line 170 in the Specific Comments below. The constant of proportionality was reduced by an order of magnitude to 0.01, and lower, (see Figs. 4 & 8) and showed that very little ice enhancement occurred due to mode 2. Hence, showing that further experimental studies are required to help constrain this uncertainty.

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 dropdrop 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-glasstarget 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 raindropfreezing fragmentation in a representative manner.

Specific comments

RC: 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 ?
AC: We have added further details highlighted in yellow below.

2.1 Model description

All simulations in this paper used the bin microphysics model (BMM, https://github.com/UoM-maul1609/bin-microphysics-model an adapted version of the control model described in Fowler et al. (2020), developed at the University of Manchester. The bin microphysics model includes activation of cloud droplets and condensation / deposition from water vapour, collision and coalescence of water drops, inertial impaction of aerosol particles, ice-ice aggregation, riming and secondary ice processes.

Aerosol particles are represented as multiple log-normal modes of different chemical compositions (externally-mixed modes). Each externally-mixed mode is described by an internal mixture that has the same chemical composition across all sizes. The BMM is initialised by summing multiple log-normal size distributions:

$$\frac{dN}{d\ln(D_a)} = \frac{N_T}{\sqrt{2\pi}\ln\sigma_g}\exp\left[-\frac{\ln^2\left(\frac{D_a}{D_{a,m}}\right)}{2\ln^2\sigma_g}\right] \quad (1)$$

where N is the number density of aerosol particles, D_a is the aerosol particle diameter for the mode, N_T is the total number of aerosol particles in the mode, D_a, m is the median aerosol particle diameter for the mode and σ_g is the geometric standard deviation of the logarithmic distribution.

The activation of cloud condensation nuclei is calculated from condensation of liquid water onto the aerosol particles with the equilibrium vapour pressure described by κ-Kohler theory, where the size and hygroscopicity of an aerosol particle is related by a single parameter, κ (Petters and Kreidenweis, 2007). The rate of drop growth via diffusion takes into account mass accommodation through modified diffusivity and conductivity terms (Jacobson, 2005; H.R. Pruppacher, 2010). While initial growth of cloud drops occurs via the diffusional growth equation, later growth to raindrops occurs via the collision-coalescence process. Collision-coalescence growth is described by the stochastic collection equation which is solved using the method of Bott (1998), and collisional efficiencies are calculated according to the Long (1974) kernel. The model treats binned distribution for liquid particles and a separate binned distribution for ice particles.

Homogeneous freezing from a supercooled water drop follows the method described in Koop et al. (2000). Heterogeneous freezing occurring via ice nucleating particles is calculated using the DeMott et al. (2010) ice nucleation parameterisation, which requires knowledge of the aerosol particle number density with diameter $\geq$ 0.5 μm. The same parameterisation is used to describe the freezing of rain drops. We first determine the number of aerosol particles with diameter $\geq$ 0.5 μm contained within a rain drop and multiply this by the number concentration of particles within the same category. The number of ice nucleating particles that are active is then calculated using the DeMott et al. (2010) parameterisation. This is scaled by how many aerosol particles are contained within the drop giving the number of frozen drops in that category.

Ice particle growth from the vapour is described using a growth rate which takes into account mass accommodation through modified diffusivity and conductivity terms (Jacobson, 2005). Once formed, ice particles grow according to the model described in Chen and Lamb (1994). The ice particle bins carry properties that are averaged within a mass bin. These properties are the aspect ratio of the ice crystals; the volume of the crystals; the rime mass; and the number of ice crystal 'monomers' per ice particle. Ice-ice aggregation and riming are also calculated using the method of Bott (1998), which is modified to transport the extra properties discussed above. The terminal velocity of ice particles is determined from Heymsfield and Westbrook (2010) based on the mass and shape of the ice particles.

RC: 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.
AC: See above.

RC: Line 148: It might be good to clarify that Mode 2 is for raindrops.
AC: We have added the following sentence 'For fragmentation to occur, the drop diameter must be greater than 0.15 mm and the mass of the drop must be less massive than the ice particle.'

RC: Line 155: Need to say that NM2 is the number of fragments per drop accreted.
AC: Added. Sentence now says: 'Then the number of fragments per drop accreted due to M2 (N_{M2})...'

RC: 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 ?
AC: We have rephrased the sentence 'We ran all possible combinations of SIP mechanisms which gave a total of 15 simulations for each sensitivity investigated in addition to a control simulation with no SIP mechanisms. The combinations are given in Table A1 of the Appendix.'

RC: 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.
AC: We have added M2 DE_{crit} as a sensitivity study to Table 2 and added the following section:

3.2.4 M2 Φ and DE_{crit}

The M2 parameterisation depends on both the probability of the splash containing ice (Φ) and the onset of splashing (DE_{crit}) as shown in Eq. 9. Both of these parameters are determined on experimental studies and present a source of uncertainty. Therefore, we individually investigated the M2 Φ and DE_{crit} for shallower clouds with an updraft speed of 2 m s^{-1} for both natural and near-city aerosol size distributions and warmer and colder cloud bases.

The results are given in Sections 4.1.4 and 4.2.4 for a natural and near-city aerosol size distribution respectively.

4.1.4 Sensitivity test: M2 DE_{Crit}

The M2 parameterisation given in Eq. 9 requires the onset of splashing, denoted as DE_{crit}. We used the value of 0.2 given in Phillips et al. (2018) based on laboratory data of drops colliding on roughened copper hemispheres from Levin et al. (1971). To test the sensitivity of ice enhancement to DE_{crit} during M2 simulations, we ran simulations for warmer and colder cloud bases with natural aerosol size distribution and updraft speeds of 2 m s^{-1} using the following DE_{crit} values: 0.2, 3 & 6. The results are plotted in Figure 5.

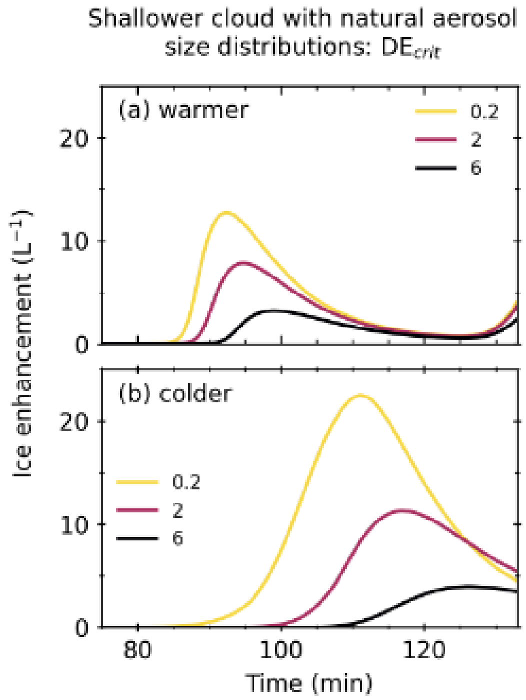


Figure 5. M2 ice enhancement against simulation time for three DE$_{crit}$ values (0.2, 3 and 6) for a shallower (1.3 km) cloud with natural aerosol size distributions and updraft speed of 2 m s^{-1}. Warmer refers to cloud base temperatures of 7 °C, and colder refers to cloud base temperatures of 0 °C.

4.2.4 Sensitivity test: M2 DE$_{Crit}$

Figure 9 shows the M2 ice enhancement against simulation time for three DE$_{crit}$ values, 0.2, 3 and 6, for shallower clouds with a near-city aerosol size distribution. In general, lower values of DE$_{crit}$ had earlier maximum ice enhancement peaks compared to higher values of DE$_{crit}$. The maximum ice enhancement peaks were greater for lower DE$_{crit}$ values.

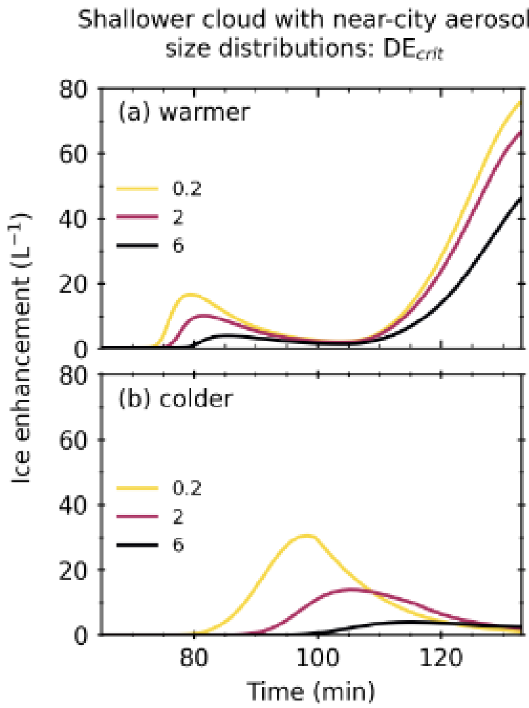


Figure 9. M2 ice enhancement against simulation time for three DE_{crit} values (0.2, 3 and 6) for a shallower (1.3 km) cloud with near-city aerosol size distributions and updraft speed of 2 m s^{-1}. Warmer refers to cloud base temperatures of 7 °C, and colder refers to cloud base temperatures of 0 °C.

We have also expanded the discussion of DE_{crit}.

Other uncertainties in the M2 parameterisation include the critical value for onset of splashing (DE_{cit}). For splashing to occur, the dimensionless energy must be greater than DE_{crit}. Phillips et al. (2018) estimated a value for DE_{crit} of ~0.2 based on room temperature experiments of colliding drops by Low and List (1982) and acknowledged that this was a source of uncertainty. The equation used to describe number of secondary liquid drops was further constrained using laboratory experiments by Levin et al. (1971). These experiments consisted of 2.5 mm drops impacting on a rough copper sphere at room temperature. The number of secondary drops formed in collision experiments is sensitive both to the geometry and impact material; hence the expression used to describe the number of liquid fragments presents another source of uncertainty. We increased the DE_{crit} by a factor of 10 and 30 to demonstrate the sensitivity of M2 ice enhancement on the DE_{crit}, as

shown in Figs. 5 and 9. When the DE_{crit} was increased by a factor of 10 or 30, the ice enhancement decreased compared to the stated DE_{crit} in Phillips et al. (2018), but ice enhancement still occurred. Despite the uncertainties within the M2 parameterisation, and in agreement with Phillips et al. (2018), it is more erroneous not to include M2 within simulations. Our laboratory experiments strongly suggest that the M2 SIP mechanism could be active (James et al., 2021), and further laboratory studies into this mechanism will help reduce the uncertainties listed.

RC: 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) ?
AC: Changed.

RC: 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).
AC: This is correct, but there are also often no measurements of the chemistry or dust surface area. Hence, we choose a simpler parameterisation to represent primary INP.

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

RC: 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 yaxis would be more appropriate in view of the uncertainty.
AC: It is not straightforward to do this due to the non-linearities involved and the structural uncertainty in the parameterisation (as discussed by the Reviewer 1 there are not only parametric uncertainties, but also structural). However, an idea of the uncertainties can be found looking at Figs. 4 & 8 (Mode 2 Φ) and Figs. 5 & 9 (Mode 2 DE_{crit}) as unknowns.

The purpose of the paper is to show the potential importance of M2 compared to the other mechanism, given our best guess.

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

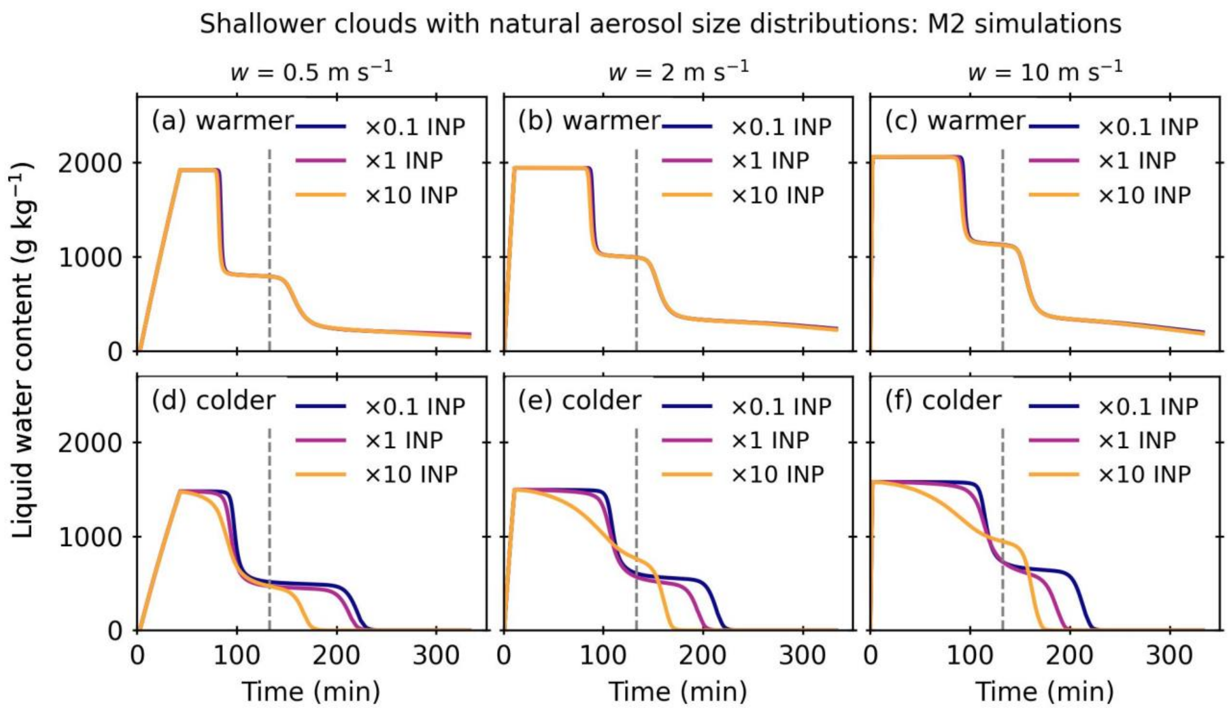
AC: We have rephrased the sentence to the following:

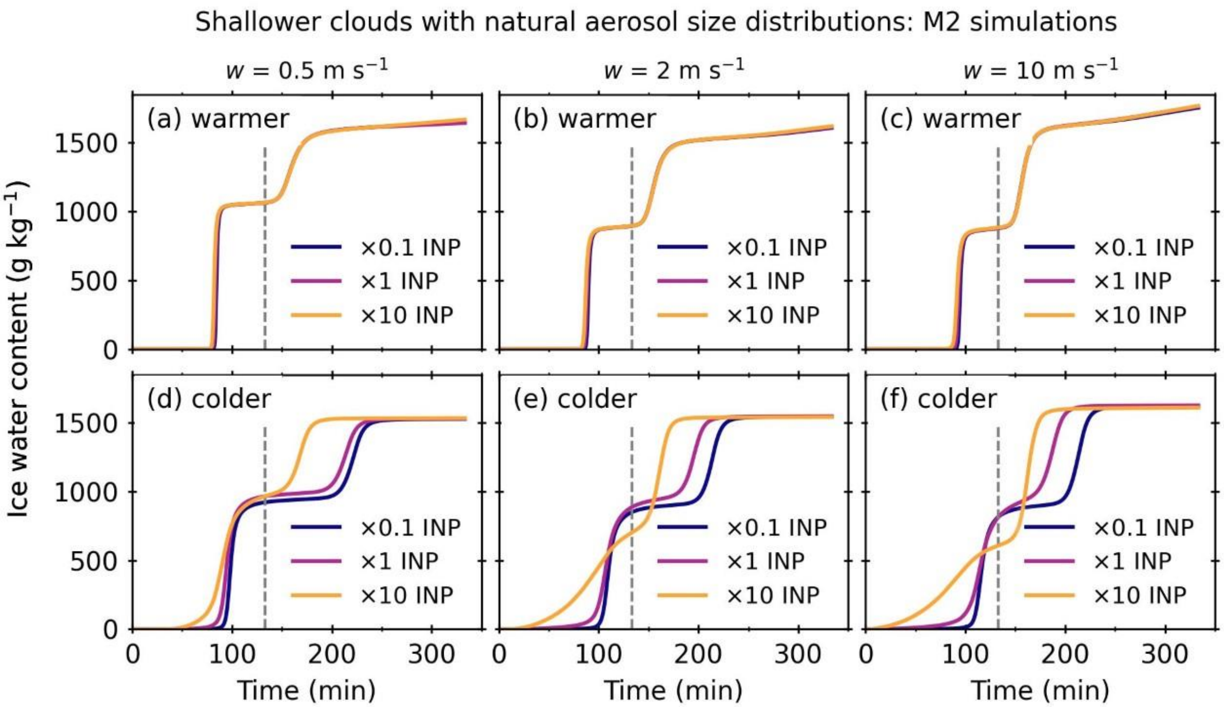
'Other studies which modelled the M2 SIP mechanism with the M1 SIP mechanism did not provide a breakdown of the contribution from M1 and M2 (Zhao et al., 2021, Zhao and Liu, 2022, Huang et al., 2022, Karalis et al., 2022). Two studies found that M2 combined with M1 was not an effective SIP mechanism in the simulated conditions (Sotiropoulou et al., 2020, Georgakaki et al., 2022). Sotiropoulou et al. (2020) stated that this was due to their thermodynamic conditions with relatively cold cloud base temperatures and Georgakaki et al. (2022) stated that this was due to the drops being too small to initiate M1 + M2 in their simulated conditions.'

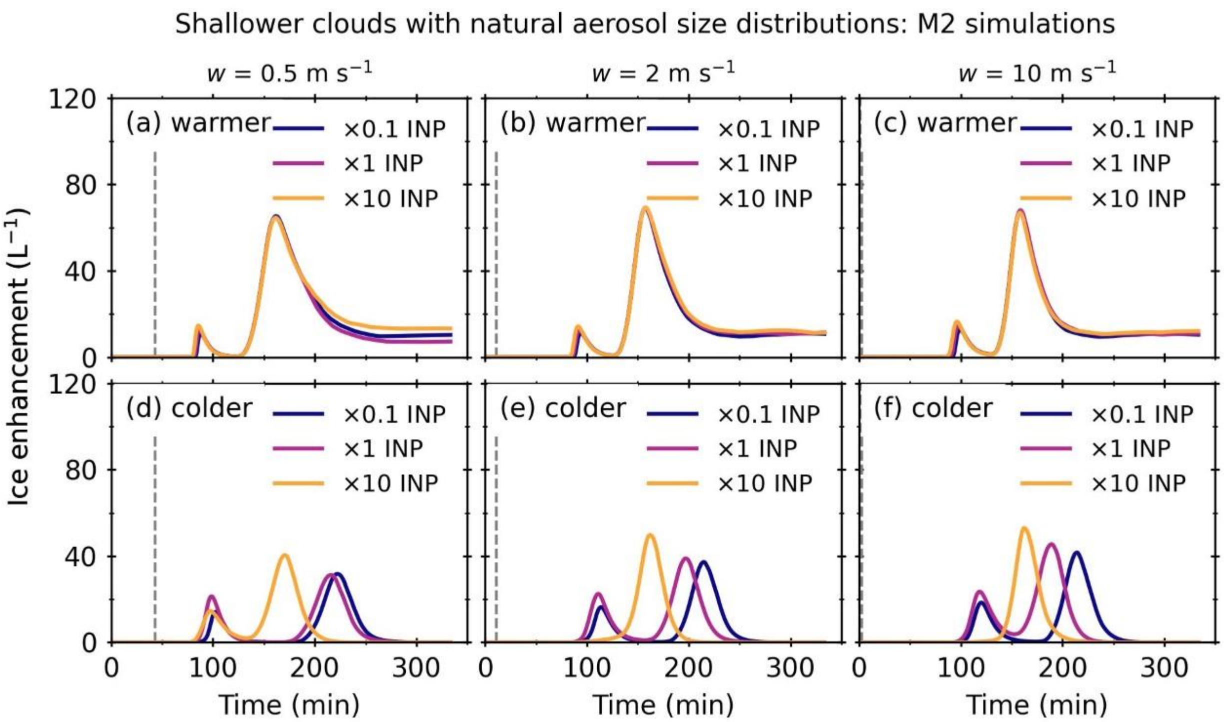
RC: Line 578: Need to explain the mechanism for how high IN concentrations in the model suppress SIP and coalescence. Do they cause subsaturation with respect to liquid, from vapour growth of ice, and is this what curbs the droplet growth, inhibiting raindrop-freezing fragmentation ? Or is the liquid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

AC: The high IN concentration in the model suppresses SIP and coalescence due to the Wegener-Bergeron Findeison process. Below are plots of liquid water content, ice water content and ice enhancement over the control simulation for a mode 2 simulation with a natural aerosol size distribution. These plots show an extended simulation run compared to the plots in the paper.




Shallower clouds with natural aerosol size distributions: M2 simulations
w = 0.5 m s^{-1}
w = 2 m s^{-1}
w = 10 m s^{-1}
(a) warmer
(b) warmer
(c) warmer
(d) colder
(e) colder
(f) colder
×0.1 INP
×1 INP
×10 INP
Ice water content (g kg^{-1})
Time (min)
0
500
1000
1500
100
200
300


Shallower clouds with natural aerosol size distributions: M2 simulations
w = 0.5 m s^{-1}
w = 2 m s^{-1}
w = 10 m s^{-1}
(a) warmer
(b) warmer
(c) warmer
(d) colder
(e) colder
(f) colder
×0.1 INP
×1 INP
×10 INP
Ice enhancement (L^{-1})
Time (min)
0
40
80
120
100
200
300

- When the simulations are run for longer, all INP concentrations in the colder simulations eventually deplete the LWC, but the x10 INP occurs earlier in the simulations and has a different profile.
- The IWC saturates around 1500 g/kg for all simulations with the x10 INP occurring earlier. The IWC is higher earlier in x10 INP simulations, corresponding to a lower LWC earlier in the simulations.
- Only one ice enhancement peak was observed for x10 INP, whereas for the lower INP concentrations there are two peaks, the first smaller and the second larger.

The Wegener-Bergeron Findeisen process occurs earlier in the x10 INP simulations as initially there is a higher IWC earlier in the simulations, which causes more vapour to condense onto the ice suppressing the coalescence process, hence reducing SIP. Whereas, for the x0.1 and x1 INP simulations, coalescence occurs earlier due to lower IWC allowing SIP formation, which increases ICNC.

We have added the liquid and ice water content figures to the supplement and added the following paragraph:

the RS mechanism was 'turned off' due to the Wegener-Bergeron Findeisen (WBF) process (Crawford et al., 2012). Figures S.25 & 26 of the Supplement show the liquid and ice water contents for M2 simulations with a natural aerosol size distribution, respectively, with extended simulation runtimes, demonstrating that the WBF process also occurred in our colder cloud base simulations. However, the WBF process occurred earlier, with a different profile, in the $\times 10$ INP concentration simulation compared to the $\times 1$ or $\times 0.1$ INP concentration simulations as initially there was a higher ice water content in the simulations, which allowed more water vapour to condense onto the ice suppressing the coalescence process, hence reducing SIP. Whereas, for the $\times 0.1$ and $\times 1$ INP simulations, coalescence occurred earlier due to the lower ice water content allowing SIP formation, which increased the ice enhancement.

References

Harris-Hobbs, R. L., and W. A. Cooper, 1987: Field Evidence Supporting Quantitative Predictions of Secondary Ice Production Rates. J. Atmos. Sci., 44, 1071–1082, https://doi.org/10.1175/1520-0469(1987)044<1071:FESQPO>2.0.CO;2.

Mossop, S.C. (1976), Production of secondary ice particles during the growth of graupel by riming. Q.J.R. Meteorol. Soc., 102: 45-57. https://doi.org/10.1002/qj.49710243104