

Report #1

Suggestions for revision or reasons for rejection

(visible to the public if the article is accepted and published)

Review of "Isotopic Evidence for Ice Growth by Riming in Precipitation" by Aggarwal and co-authors, Manuscript egusphere-2026-687.

Assessment: Minor revisions

I thank the authors for their extensive efforts to rework the paper in response to the suggestions of the reviewers and for making explicit their hypothesis about the potential impact of riming on ice composition. I find the paper improved but have a few minor concerns as outlines below.

Response: We thank the reviewer for a careful and thorough review of the manuscript once again and appreciate the reviewer's support for the revisions.

Minor Comments (5/112 means page 5, line 112):

4/127-129: This point seems really crucial for the notion that wet growth by riming is ubiquitous in mixed phase clouds with a variety of ice crystal sizes, not just for systems that produce hailstone-sized ice particles. Since many isotope scientists may not have a deep knowledge of how droplets freeze when accreted by an ice particle, a couple of extra sentences (or even a paragraph and figure in the supplement) that expand on this point and give some intuition for why this process applies for a wide range of precipitation (not just hail) might be helpful. Alternately, if there's a reference that talks about this, that could be cited.

Response: We appreciate the comment and agree that this is an important issue. The paragraphs immediately preceding and following the text highlighted in the review comment cite many references that describe in detail the riming process and its occurrence in different meteorological conditions. We believe that the concern raised by the reviewer may be addressed by additionally citing Korolev et al. (2017) in the preceding paragraph.

I was curious about this, so I found Figure 16-6 in Pruppacher and Klett, which depicts the Schumann-Ludlam limit, the liquid water content threshold (w_{l_crit}) that divides wet and dry growth regimes. Notes by Flossman (p. 104 in <https://andreaeflossmann.wordpress.com/wp-content/uploads/2017/02/evolution-of-ice-in-clouds.pdf>) provide a derivation of w_{l_crit} that suggests $w_{l_crit} \sim 1/(\text{ice_radius} \cdot v_t) = \text{ice_radius}^{-(1+b)}$ if the terminal velocity, $v_t = a \cdot r^b$. With an estimate of $b=0.6$ (Table 2 in Milbrandt and Morrison, 2013, <https://doi.org/10.1175/JAS-D-12-0204.1>), $w_{l_crit} \sim \text{ice_radius}^{-1.6}$.

The Schumann-Ludlam limit in Pruppacher and Klett's Figure 16-6 seems to be from experiments with ice diameter of 1-2 cm (Lesins and List, 1986, [https://doi.org/10.1175/1520-0469\(1986\)043%3C2813:SADSOG%3E2.0.CO;2](https://doi.org/10.1175/1520-0469(1986)043%3C2813:SADSOG%3E2.0.CO;2)). For 2 mm ice particles, w_{l_crit} would be $(2 \text{ mm}/1 \text{ cm})^{-(1.6)} \sim 13$ times larger than shown in Figure 16-6 of Pruppacher and Klett, which would seem to rule out wet growth for many (but not all) smaller liquid water contents. Changes in particle density and collection efficiency between larger and smaller ice could have some impact here, but the calculation referenced in the paper seems different from my (admittedly quick and back-of-the-envelope) estimate here. If my calculations or approach is mistaken, the authors could correct me in their response.

Response: We appreciate the comment and agree with the reviewer that the Schumann-Ludlam threshold scales with particle size. But the estimated scaling factor of 13 is too large because changes in the relative importance of diffusive and convective heat transfer occurring with a change in particle size were not accounted for. When these are included, the Schumann-Ludlam limit for a 2mm particle is about 3.6x compared to that for a 1 cm particle.

The Schumann-Ludlam limit defines the liquid water content (LWC) above which a persistent liquid film is present during riming and this film evaporates before freezing is complete. However, evaporation of the accreted liquid on a particle surface (and associated isotopic fractionation) may occur even when a liquid film is not persistent. Mossop (1976; doi:10.1002/qj.49710243104) has shown (his Fig. 8) that for LWC below the Schumann-Ludlam limit, accreted liquid on small particles (0.5 to 2 mm) is evaporated as long as the latent heat released during riming raises the particle surface temperature above the ambient ($T_a = -5^\circ\text{C}$) by more than 0.6°C . This transitional regime provides a physical mechanism consistent with the observed MDV–d-excess correlation across a range of cloud conditions, including stratiform clouds where LWC may fall below the Schumann-Ludlam threshold.

To address reviewer concerns and to clarify, we have re-arranged the text in preceding paragraphs and revised the paragraph marked by the reviewer:

“Bailey et al. (1969) conducted laboratory experiments in an icing tunnel (air temperature = -20°C) to study isotopic fractionation during the freezing of supercooled droplets on a cold surface (riming). The experiments were conducted at liquid water concentrations (LWC) of 1 or 5 g m^{-3} . In both experiments, the $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values of the frozen material were slightly higher than those of the supercooled liquid. A modeling study by Jouzel et al. (1985) corroborated the laboratory results of Bailey et al. (1969). At lower LWC, the droplets froze completely and rapidly (“dry-growth” regime), while at higher LWC, the supercooled liquid was considered to have partially evaporated before freezing was complete (“wet-growth” regime). These studies were conducted in the context of the hailstone growth. Hailstones are an extreme case of riming distinguished by their larger size ($>5\text{ mm}$) compared to rimed particles (Pruppacher and Klett, 2010). A continuum of ice particles from unrimed to graupel and hail has been shown based on their mass-size relationships (Lin and Colle, 2011) and the physics of isotopic fractionation in hailstones is equally applicable to the riming of smaller particles in mixed phase clouds.

More significantly than the slightly higher δ values, the d-excess of rimed ice in the laboratory and modeling studies was lower than that of the supercooled liquid. Natural hailstones analyzed by Jouzel et al. (1985) had low ($<+10\text{‰}$) d-excess values. Increased riming of snow from the Rocky Mountains (Lowenthal et al., 2011) also appears to correspond to lower d-excess values reported in Lowenthal et al. (2016).

Studies of the mechanism of riming (e.g., Macklin and Payne, 1967; Pruppacher and Klett, 2010, Korolev et al., 2017, and references therein) suggest that when a liquid droplet collides with an ice particle, it may lose some water by splashing, depending on the impact velocity. The drop initially freezes with a thin frozen shell as the latent heat of fusion released in the process

warms the inner part of the droplet. If the latent heat is dissipated fast enough to keep the particle surface temperature (T_s) a few degrees below 0°C , the accreted water freezes in the dry-growth regime, with or without the loss of some accreted water by shedding. Conversely, if heat dissipation is slower — for example, due to high droplet concentration or increased frequency of droplet-particle collisions — T_s rises and the accreted water spreads to form a thin film on the particle surface. In that case, the accreted water freezes in the wet-growth regime where the liquid film may be partially evaporated before freezing is complete. This evaporation is driven by the vapor pressure difference between the warmer particle surface and the ambient air.

The liquid film during wet-growth may be persistent ($T_s = 0^\circ\text{C}$) or transient, depending on the particle size, LWC, and ambient air temperature (T_a). The minimum LWC required to sustain a persistent liquid film is the Schumann-Ludlam limit (Pruppacher and Klett, 2010). Calculations using a heat balance equation (Pruppacher and Klett, 2010) show that for small particles of ~ 2 mm, this limit may be reached in stratiform clouds at $T_a > -10$ to -5°C , particularly when LWC increases due to embedded convection or enhanced turbulence. Even below this limit, Mossop (1976) showed that a transient liquid film is sufficient for wet-growth (with evaporation of the liquid) on small particles (0.5 to 2 mm) at $T_a = -5^\circ\text{C}$ when T_s rises above the ambient by more than 0.6°C . Wet growth — whether from a persistent or transient liquid film — is therefore expected in stratiform clouds at air temperatures warmer than approximately -10°C , consistent with the Hallett-Mossop riming zone (~ -8 to -3°C) within which supercooled liquid water is most abundant (Mossop, 1976). Below about -10°C , riming would occur predominantly by dry-growth process.

5/158: Specify where in the vertical (or at what temperatures) increases in terminal fall velocity are related to precipitation d-excess, something like: "increasing terminal fall velocity above the 0°C isotherm"

Response: We appreciate the suggestion and have made an attempt to revise the text. The simple " 0°C isotherm" does not apply to the case of snowfall and since the melting layer has not been introduced yet in the abstract or the introduction, the change becomes complicated. We have therefore revised the sentence to remove MDV here and refer only to rimed mass:

"Our findings reveal that precipitation d-excess values decrease with an increasing fraction of particle mass attributed to riming."

24/646-648 and Fig 8: It would have been helpful to have these MDV and d-excess values included in the figure itself or in the caption. Another option would be a short legend: "low MDV/high d_p " green, etc.

Response: OK; revised.

27/731: My reading of figure 10c is that SI increases to $\sim 20\%$ at 3900m, not " $\sim 10\%$ " as specified in the text.

Response: Thanks for pointing out the discrepancy. Text revised.

Fig 10: Please specify the MDV/d-excess contrast between these two cases as suggested for Figure 8 above.

Response: OK.

29/799-800: Could the authors explain the reasoning behind "we would expect lower d-excess samples to lie in the unrimed or lightly rimed categories" (even with a short phrase)?

Response: The logic is that sub-cloud evaporation only affects d-excess, not the snow or rain velocities measured within the cloud layer. We have revised the text for clarity:

"Because V_r is measured immediately below the melting layer, before rain has fallen far enough to be subjected to sub-cloud evaporation, the $V_s - V_r$ classification reflects in-cloud microphysics rather than sub-cloud processes. Consequently, if the low d-excess of a sample was caused by sub-cloud evaporation rather than riming, we would expect that sample to lie in the unrimed or lightly rimed fields."

32/881: " S_i " in Jouzel and Merlivat (1984) is not exactly the ice supersaturation ratio. It's the ice supersaturation modified by a temperature-dependent thermodynamic factor that matters more near 0C than at colder temperatures. See eqn 13 in Jouzel and Merlivat (1984).

Response: We agree with the comment. However, for practical purposes, there is a negligible effect on calculated d-excess in the temperature range of -50 to -20°C we have used. We have added new text:

"Jouzel et al. (1984) defined S_i as the effective ice supersaturation that accounts for the thermodynamic effects of latent heat at the particle surface. The effective S_i is equivalent to the ambient supersaturation ratio at temperatures below -35°C , and is lower at higher temperatures. The difference becomes most significant at temperatures near zero. Within the temperature range -35 to -20°C that we have used for calculations here, the effective S_i is only 1–5% lower than the ambient ratio. Therefore, we have simplified the calculation of vapor deposition and diamond dust d-excess (Table 4) by defining S_i as the ambient supersaturation ratio."

32/893 (Eqn 3): Why doesn't the freezing rate follow from the heat budget of the liquid and include the cooling due to evaporation?

Response: Eq. 3 follows Jouzel et al. (1985). We have added the following text after Eq. 3:

"Note that $d\mu_f$ is determined by mass balance ($d\mu_f = d\mu_c - d\mu_e$) rather than the heat budget directly, because both constraints are simultaneously satisfied in wet-growth; the heat budget determines the minimum LWC required, and once that threshold is exceeded, T_s is maintained at 0°C and the mass balance governs the freeze rate."

In addition, Eq. 4 was mislabeled: it defines $d\mu_c$, not $d\mu_f$, and has been corrected.

33/922: "almost instantaneous" freezing makes it sound like freezing may not actually occur under equilibrium conditions. If this fractionation occurs during dry growth, the isotopic mass balance

requires the loss of vapor that is, on average, more enriched than the liquid that was rimed. Jouzel et al (1985)'s appendix B suggests a way to calculate the integrated impact of this evaporation during dry growth.

Response: The reviewer is correct that Jouzel et al. (1985, Appendix B) provides a framework to calculate the isotopic effect of evaporation during dry growth. We have deliberately simplified our calculations by excluding this effect as it makes a negligible difference in calculated d-excess. Heat conduction into the ice particle is faster by almost two orders of magnitude than to the ambient air. As a result, the evaporated fraction of rapidly freezing cloud droplets is only about 0.3%, which gives a d-excess correction of approximately +0.06‰. We have added text acknowledging this simplification and its justification.

“Dry-growth occurs when the latent heat of fusion is dissipated without the particle surface warming to 0°C. In that case, a persistent liquid film does not form. The accreted droplets freeze rapidly, with heat conducted primarily into the ice particle rather than to the ambient air. A small fraction of each droplet may evaporate before complete freezing, but that would produce an isotopic correction of the order of 0.1‰ (following Jouzel et al. 1985, Appendix B). Therefore, the isotope ratio of the rimed ice is assumed to reflect only the equilibrium liquid-solid fractionation:”

40/1090-1092,1096: If we accept that riming does indeed have a strong impact on d-excess above the melting layer, will that d-excess survive exchange (evaporation/equilibration) with vapor below? In the deep tropics, the melting layer can be above 5km, leaving > 10 minutes of time for a 7 m/s raindrop to fall to the surface and interact with the vapor below.

Response: Yes. The d-excess signal from riming in the snow zone survives the sub-cloud layer at our tropical study location where the sub-cloud layer was ~4 km. This is because when rainfall occurs, the sub-cloud RH generally is high and even a 4 km sub-cloud layer does not result in a significant change in d-excess (Sect.3.3.2). Additional evidence is provided in Fig. 11 where Rio Claro samples with a low d-excess of ~5‰ plot in the graupel field in the $V_s - V_r$ space, consistent with their low d-excess resulting from riming.

41/1137: Since particular combinations of different adjustable parameters were chosen here, I would suggest softer language here, changing "was shown to result from" to "can be explained by".

Response: OK; revised

Typographical/wording Suggestions (in most cases, suggested changes are _between the underscores._):

7/200: "P is atmospheric pressure _in hPa_, ..."

Response: OK

8/230: "... without further growth or _by_ breaking into smaller drops ..."

Response: Thanks; we believe that the suggested change results in a different message so we have left it as is.

11/332: "... values increase with decreasing altitude _within the lowest_ 1km above ground, ..." I found "increasing with decreasing altitude up to about 1km" confusing.

Response:OK; text revised for clarity.

11/333: "_Close to the surface,_ sub-cloud sublimation of snow ..."

Response:OK; text revised.

11/338: "We used the vertical profiles of reflectivity below 2.5 km to _identify and_ exclude sublimation-affected data ..."

Response:OK; text revised.

11/339: "One group_, which is excluded from the analysis,_ shows a substantial decrease ..."

Response:OK; text revised.

12/371: Start a new paragraph after "Murhpy and Koop (2005)." since that next sentence is important for the reader's understanding of what follows.

Response:OK; text revised.