

Author response to review comments on manuscript EGUSPHERE-2026-687:

“Isotopic Evidence for Ice Growth by Riming in Precipitation” by Aggarwal et al.

Major changes to the manuscript include the following:

1. Added new text based on the analysis of cloud microphysical processes using radiosonde data from Summit, Ny-Ålesund, and Dumont d’Urville.
2. Expanded the Ny-Ålesund dataset with Doppler velocity and d-excess values for 2014-2017.
3. Added new text based on extensive calculations of the isotopic composition of precipitation in different growth environments
4. Re-arranged data and methods section.
5. Revised discussion section with significant changes to existing text and added new text

We have also reviewed and revised the text to ensure consistency and factual accuracy. We would like to propose a revised manuscript title that we believe is more informative about the physical process being studied and its observable consequence:

“Isotopic fractionation during ice growth by riming and its effect on the d-excess of precipitation”

Response to Reviewer Comments

We thank the two anonymous reviewers and Dr. Sodemann for their constructive criticism and helpful suggestions for strengthening the manuscript. We first outline our response and changes made to address the major issues that are common to all reviews. Subsequently, responses are provided for additional comments specific to each of the three reviews (RC1, RC2, CC1).

Common issues: we believe that all reviews raise concerns that can be summarized as follows:

1. Feasibility of riming in non-hailstone environments and evaporation of accreted liquid
2. Low d-excess of winter precipitation at Summit

3. Riming being an additional process instead of the only process for d-excess variations
4. Unclear (inaccurate) text related to d-excess of ocean evaporation

Response

1. Feasibility of riming in non-hailstone environments

Text has been revised to include specific field observations indicating isotopic fractionation during riming and to provide more details on why hailstone studies are relevant.

Warburton and deFelice (1986) and Demoz et al. (1991) analyzed freshly fallen snow in the Sierra Nevada mountains. The $\delta^{18}\text{O}$ values were higher when ice crystals included graupel and other rimed particles compared to those of snow with ice crystals consisting of unrimed particles such as dendrites, columns and plates. Lowenthal et al. (2011) used sulfate concentrations in cloud water and snow to estimate the fraction of ice that formed by riming and observed that the $\delta^{18}\text{O}$ of snow increased with the rimed fraction.

Bailey et al.'s (1969) experiments involved the direct freezing of supercooled water droplets on a cold surface, similar to the riming growth of small particles and graupel. Jouzel et al. (1985) built their model using the conceptual framework derived from experimental studies of droplet freezing (and wet growth involving evaporation) along with the physical and isotopic fractionation constants during the evaporation of water.

The latent heat of fusion released from the accreted liquid can be dissipated by conduction (warming the particle surface to 0°C) and after that, by evaporation. Because of the higher temperature on the particle surface, the vapor pressure over the accreted liquid water is higher compared to that in the bulk air. This vapor pressure gradient allows for the evaporation of the accreted liquid before freezing is complete.

The key parameters that determine whether wet growth occurs during riming to form hailstones or graupel are ambient temperature, particle size and the availability of sufficient liquid water. Using established equations of heat balance (Pruppacher and Klett, 2010), it can be shown that evaporation during wet growth (particle surface temperature = 0°C) requires that the supercooled water collection rate exceed the rate at which it can be frozen. For small

particles of ~2mm, and the relatively low liquid water contents (LWC) that are generally observed in stratiform clouds, calculations show that wet growth may occur at air temperatures greater than about –10 to –15 °C. Below that temperature, riming would occur by a dry growth process where isotope fractionation is determined by the liquid-solid fractionation coefficient that has only a small temperature dependence.

Using vertical profiles of temperature (T) and relative humidity (RH) derived from radiosondes at Summit, Ny-Ålesund, and Dumont d’Urville, the revised version shows that temperatures were conducive for riming during summer and winter precipitation at Ny-Ålesund and Dumont d’Urville, and during the summer at Summit. At the mid-latitude (Cazadero) and tropical (Rio Claro) locations, riming conditions commonly are known to occur, particularly within about a kilometer above the melting layer (Houze, 2014).

2. Low d-excess of winter precipitation at Summit

Winter precipitation at Summit has low d-excess, but also low MDV. This introduced a contradiction for the proposed riming-driven lowering of d-excess of precipitation. We have investigated this issue in more detail, using vertical profiles of T and RH derived from radiosondes, and revised the text to exclude riming as a potential cause of low d-excess for winter precipitation at Summit.

The radiosonde data show that on isotope sampling days, winter temperatures at Summit were below –38°C at heights greater than ~2400 m above ground level (AGL), indicating that mixed-phase conditions did not exist in most of the precipitating column. However, a deep layer with ice supersaturation was present from near the surface to ~6700 m AGL with a mean ice supersaturation (SI) of ~11% (range ~0 to 37%) and mean T at the maximum supersaturation level ranging from –27°C near surface to –50°C at higher elevations. Ice growth in that layer would occur by vapor deposition (diamond dust). Calculations using Jouzel and Merlivat (1984) show that with high levels of ice supersaturation at very low temperatures, the d-excess of vapor deposited ice would be low (near zero or negative) when the $\delta^{18}\text{O}$ of the vapor also is very low, as expected at Summit. This result is consistent with calculations of d-excess using a GCM where supersaturation with ice is parameterized as $S_i = a + b \cdot T$ with $a=1$ and $b = -0.002$ to -0.006 (Jouzel and Merlivat, 1984; Dütsch et al., 2019). A parameter value of $b = -0.002$ is commonly used to reproduce the relatively higher d-excess of ice cores (Dütsch et al., 2019). However, a value of $b = -0.006$ with low $\delta^{18}\text{O}$ of vapor results in low or negative d-excess of ice

(Fig. 1 of Dütsch et al.). With $b = -0.006$ and $T = -50^{\circ}\text{C}$, $S_i = 1.3$ ($SI = 30\%$); at $T = -30^{\circ}\text{C}$, $S_i = 1.18$, similar to the observed temperature and RH values for Summit winter. Thus, our calculations are consistent with those of Dütsch et al. and provide an explanation for low d-excess and low MDV in the absence of riming.

In addition to vapor deposition, the revised version includes calculations of the expected d-excess of rimed ice using the equations of Jouzel et al. (1985). Sensitivity analysis is presented and discussed showing the influence of various input parameters. Finally, the end-member d-excess (of vapor deposited and rimed ice) is used to calculate a “process-weighted” d-excess of precipitation. These process-weighted calculations show that ice growth by vapor deposition (including diamond dust) at various ice supersaturation and temperature combinations can result in d-excess values consistent with the observed low d-excess of Summit winter precipitation. Varying proportions of vapor deposited ice and rimed ice (wet and dry growth) can result in a range of d-excess values similar to the observed range for summer precipitation at Summit and for the rest of the study locations.

3. Riming being an additional process instead of the only process for d-excess variations

With the above explanation for winter Summit precipitation where low d-excess may occur without riming, and the influence of the isotopic composition of vapor on the calculated d-excess, the conclusions have been revised as follows:

“A process-weighted framework was developed to compute the d-excess of precipitation as a mixture of ice forming by vapor deposition (including diamond-dust) and riming (wet or dry growth) to show that the d-excess variability resulting from in-cloud processes can explain the observed d-excess variations at the study locations with diverse climatic conditions from the tropical to the polar regions. Our results suggest that low d-excess values in precipitation and ice cores, that have been traditionally attributed to changes in source moisture origin or sub-cloud evaporation, warrant re-examination. This does not imply, however, that source moisture characteristics at the precipitation site are irrelevant to understanding the d-excess of precipitation. The source vapor $\delta^{18}\text{O}$ and d-excess are explicit inputs to the fractionation calculations and their seasonal and spatial variability contributes to d-excess variability. Rather, we suggest that the in-cloud fractionation component — controlled by growth temperature, ice supersaturation, and riming intensity — is of comparable or greater magnitude than the source signal in many precipitation regimes. Separating the two contributions requires independent constraints on in-cloud conditions such as those provided by the radar-based riming proxy introduced in this study. The conventional attribution of d-excess variability to conditions at the

oceanic evaporation source (sea surface temperature and relative humidity) requires additional caution, because the source evaporation signal is substantially modified by Rayleigh distillation and mixing during transport before reaching the precipitation site."

4. Unclear (incorrect) text related to d-excess of ocean evaporation and differential diffusion of isotopes

The text in the introduction has been revised to address these comments:

"The interpretation of the d-excess of precipitation and ice cores is guided by two primary constraints. The first constraint is that d-excess is inherited from the oceanic evaporation source moisture. The global average precipitation d-excess of ~10‰ (Dansgaard, 1964) reflects kinetic isotope fractionation under mean oceanic evaporation conditions (Merlivat and Jouzel, 1979). During evaporation, H₂O diffuses faster than both H₂¹⁸O and HDO across the vapor–liquid interface, so the vapor is depleted in ¹⁸O and ²H relative to the liquid. However, because HDO diffuses faster than H₂¹⁸O (Merlivat, 1978), the resulting d-excess of the vapor is higher than that of the liquid. The kinetic increase in vapor d-excess is greater when evaporation occurs under lower relative humidity or warmer sea surface temperature conditions (Dansgaard, 1964; Merlivat and Jouzel, 1979). In the current climate, oceanic vapor is estimated to have a d-excess range of ~8–13‰ (northern hemisphere) and ~9–12‰ (southern hemisphere) with a spatial and seasonal d-excess variability of ~3 to 5‰ (Pfahl and Sodemann, 2014). Ocean evaporation under dry conditions (~30% relative humidity) can produce vapor with d-excess values of about 25‰ (Pfahl and Sodemann, 2014). However, precipitation d-excess in adjacent land areas generally does not correspond to such elevated d-excess values because precipitation results from a mixture of moisture sources with different evaporation histories (Pfahl and Sodemann, 2014)."

Reviewer #1

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

This manuscript speculates that riming in precipitating, mixed-phase clouds has a strong control on the deuterium excess (d-excess) of precipitation and could potentially explain spatial and temporal variations in paleoclimate records in ice cores, in addition to process-driven short term variability in d-excess in the present day. Relationships are first established between the deuterium excess of precipitation and fall speeds of ice-phase precipitation using both in situ measurements (with MASC) and radar retrievals of doppler velocity, with lower deuterium excess in precipitation with greater riming. These relationships seem to hold more broadly across regions, and the relationship between the doppler velocities above the melting layer with riming makes the connection between riming and d-excess. A potential mechanism for evaporative fractionation of partially-frozen

droplets during the riming process is sketched out with some support from past literature. The manuscript claims that the riming effect on d-excess explains many of the d-excess signals seen in paleoclimate datasets.

Assessment: Major revisions

I find the manuscript provocative and interesting if speculative. This study presents empirical evidence for the fractionation effect of riming but spends little time enumerating/exploring other explanations for the trend, aside from rain evaporation. How much of the relationship between riming and precipitation d-excess comes from the differing isotopic composition of cloud liquid and precipitating ice? Riming presumably increases the precipitation efficiency so that more cloud liquid falls out as precipitation than in the absence of riming. How much would such an effect contribute to the riming d-excess relationship independent of any hypothesized evaporative fractionation of partially-frozen droplets? How much of the riming d-excess relationship might be explained by changes in the condensation temperature, relative humidity over ice during deposition, subcloud relative humidity during rain evaporation and/or changes in water vapor isotopic composition during the storms studied here?

In my view, a revised version of this manuscript should ideally do two things: 1) explore other potential controls on the d-excess (condensation temperature, relative humidity over ice above the melting layer, a fuller accounting for potential changes due to subcloud rain evaporation/equilibration, changes in the water vapor isotopic composition) to make a more compelling case for isotopic fractionation during riming, and 2) explain more clearly the distinction between isotopic changes due to fractionation during the riming process and the isotopic impact of having more cloud liquid included in precipitation. If the authors view item (1) above as beyond the scope of the manuscript, I would at least ask them to characterize the riming-d-excess relationship as a hypothesis and note that the observational analysis is primarily based on correlation.

Response: We thank the reviewer for a careful review of the manuscript. As noted in the response to common issues above, detailed calculations of the d-excess resulting from other growth conditions along with a discussion of sensitivity to various input parameters have been included in the revised version.

The revised version also includes specific consideration of sub-cloud evaporation that is most relevant for Cazadero, California. There, we show that at best, only a small part (<5‰) of the observed low d-excess values (up to -22‰) may result from sub-cloud evaporation. The d-excess change from sub-cloud evaporation would not affect our arguments on riming-driven lowering of d-excess.

An increase in the proportion of cloud liquid in precipitation may affect the proportion of rimed ice in the precipitation. The revised version now includes a process-weighted calculation of precipitation d-excess with end-members forming by vapor deposition, diamond-dust and riming. This shows that a change in the proportions of these end-members can results in a range of precipitation d-excess similar to that observed at the study locations.

Major comments:

1. For me, the most obvious impact of riming on precipitation isotopic composition is simply that cloud liquid might have a different isotopic composition than precipitating ice, and precipitation with different fractions of cloud liquid might have different d-excess. Why isn't this emphasized in the paper, or at least explored and discarded? Why do we need the hypothesized, ephemeral liquid layers to do so much work isotopically when this simpler explanation exists? Why would those liquid layers experience such strong fractionation when the surrounding air is close to liquid saturation (since cloud liquid droplets are present)? How could those liquid layers induced by riming have such a strong impact on the overall isotopic composition of precipitation at the ground when only a small fraction of the precipitation mass is in the liquid phase during the hypothesized fractionation?

Response: Please see the common responses #1 and #2 above.

2. While the precipitation fall speed above the melting layer is hypothesized to be the main control on precipitation d-excess here, the potential for meteorology or changes in the water vapor isotopic composition to affect the d-excess signal seems to have been mostly ignored. Could correlations between the presence of riming and other factors explain some of the riming-d-excess relationship in many of the figures? The condensation temperature, supersaturation during vapor deposition, subcloud relative humidity during rain evaporation and possibly the isotopic composition of water vapor may be available from radiosonde data, reanalysis or vapor isotope observations. Why not explore such alternative explanations for the signals seen here?

Response: As noted above, we have revised to present a process-weighted calculation for precipitation d-excess. The particle fall velocity can increase as particle size increases, or the particles become dense and rounded as a consequence of riming, but the fall velocity does not control the d-excess of precipitation.

3. The deuterium excess of ice formed by vapor deposition varies nonlinearly with increasing distillation, even for an equilibrium process (e.g., Figure 7 of Kopec et al, 2018, <https://doi.org/10.1029/2018JD028750>). The main point here is that other processes (e.g., vapor deposition onto ice) can cause precipitation d-excess to fall without a riming signal. Such behavior is also seen when vapor deposition occurs under ice supersaturated conditions (e.g., Dutsch et al, 2019, <https://doi.org/10.1029/2019MS001764>, Figure 1).

Response: This is an important comment and we have responded to it in the common issues above. Additional analysis for the revised version led us to conclude that riming is not relevant for winter Summit precipitation.

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

2/39: Newer reconstructions of local and remote temperatures from ice core data (Markle and Steig, 2022, *Clim. Past*, <https://doi.org/10.5194/cp-18-1321-2022>) seem to show somewhat more modest changes in ocean evaporation temperature of ~3 K. Their model relies on a nonlinear definition of the deuterium excess, which may be important when interpreting d-excess signals in polar regions.

Response: Thanks for the comment. The Markle and Steig analysis attempts to use a logarithmic definition of d-excess to arrive at a better explanation of ice core data. But the analysis is still predicated on precipitation d-excess being governed only by source moisture origin and vapor deposition. In the revised version, we argue that ice growth at low temperatures, low $\delta^{18}\text{O}$ of vapor and higher ice supersaturations may result in low d-excess of winter precipitation at Summit, while riming may be important for Summit summer precipitation, as well as at the other polar locations.

2/49: When I look at plots of ocean evaporation d excess in Pfahl and Sodemann (2014), I find ranges of about 8-13 per mil (Figure 1b) and about 0-25 (Figure 2), a wider range than stated in the present manuscript. Why do those ranges differ from the one included in the present manuscript?

Response: Text revised; please see common response #4.

2/57-59: During the WBF process, the liquid saturation ratio is unlikely to vary substantially from one. As a result, one can make a clear estimate of the supersaturation over ice (as the ratio of $e_{\text{sat_liquid}}(T)$ over $e_{\text{sat_ice}}(T)$) which varies from 0 at 0 degC to ~50% at -40 degC. If isotopic equilibrium between vapor and liquid is assumed and one chooses values for the isotopic composition of vapor, the deuterium excess difference between the ice and liquid phase can be computed explicitly, which results from both kinetic fractionation over ice and from the equilibrium fractionation factor differences between liquid and ice.

Response: Thanks for the comment. We have revised the manuscript with observed vertical profiles of temperature and relative humidity at Summit and Ny-Ålesund, and calculated the supersaturation with respect to ice. Precipitation d-excess has been calculated for a range of temperature and supersaturation values and used to calculate a process-weighted d-excess value of precipitation.

Looking at supplemental figure S1, point C with 200% supersaturation over ice seems unrepresentative of conditions in earth's atmosphere, where the threshold for homogeneous nucleation of ice is roughly 100% at the tropical tropopause. Emphasizing the extreme supersaturations possible at colder temperatures is a bit misleading because the deuterium excess of precipitation depends on the mass-weighted isotopic composition integrated across all hydrometeors that contribute to that precipitation. Relatively little deposition is possible at low temperatures where the saturation vapor pressure over ice is small.

Response: Thanks for pointing out the discrepancy. The 200% number in Fig. S1 was a typographical error; the figure has been revised to 100% as the correct number.

2/63: "Most precipitation _events_ over land and oceans, ..." Those papers are focused on precipitation events rather than accumulated precipitation.

Response: The observation is correct for Muelmenstadt et al. However, the Field and Heymsfield paper cited in the initial manuscript considered accumulated precipitation. On reflection, we have revised the text for clarity: "*However, precipitation over land and oceans forms dominantly by ice-phase processes (Muelmenstadt et al., 2015). Even in the tropics where warm rain is most prevalent, clouds producing only warm rain are largely confined to shallow*

isolated oceanic convective cells and onshore tropical flow over land (Schumacher and Houze, 2003; Mulmenstädt et al., 2015). This implies that the lower d-excess commonly observed in precipitation cannot be explained by source moisture conditions alone and must reflect an additional in-cloud process that reduces d-excess."

sec. 2.3.3 (Doppler velocity at the study locations): I wish the list in this section and section 2.2.X could be merged. This would help the reader stay engaged with the material rather than having to wade through a second set of details about the various datasets. This could probably be accomplished by labeling section 2.2 as "Study data" and have that include both "isotopic data" and the contents of sections 2.3, 2.3.1 and 2.3.2. Then, section 2.3 could detail the data available from the various sites.

Alternatively, some of this could also be moved to an appendix or the supplement, with a smaller summary remaining in this section.

Response: That is a good suggestion. The presentation has been re-arranged in the revised version.

16/figure 3: Both the mean doppler velocity and d-excess are uncertain, so total least squares is more appropriate here than linear regression. The same is true for the plots later in the paper. Arguably the mean doppler velocity and rime fraction are more uncertain than the d-excess.

Response: We agree with this observation. Total least squares regression was carried out and the parameters are now shown in Fig. 6 (previously Fig. 3).

16/413-417: It's difficult for the reader to assess this argument without more information.

Response: Thanks for the comment; the revisions no longer include that argument.

20/sec 3.3 on subcloud evaporation: Echoing major comment 2, why isn't subcloud relative humidity considered in the discussion of subcloud evaporation? Presumably such information is readily available from radiosondes adjacent to the radar sites, in addition to reanalysis.

Response: Radiosonde soundings were not available for Cazadero or Rio Claro. We have revised the text for clarification

"For most of the precipitation events at Cazadero, the vertical profiles of reflectivity below the melting layer either did not decrease or decreased only for a brief interval at the beginning or the end of precipitation when rain rates generally were minimal. In those cases, we discarded the d-excess and MDV data for ~30 min at the start or end of each event to avoid the potential effect of sub-cloud evaporation on our analysis. This approach of partially discarding the early or late portions of an event was also used for Rio Claro."

Subsequent text was revised based on sub-cloud evaporation calculations:

"We estimated the magnitude of d-excess by sub-cloud evaporation using the Stewart (1975) model, which calculates isotopic fractionation during raindrop evaporation as a function of drop

size, fall distance, ambient temperature and relative humidity. Graf et al. (2019) applied a similar approach. They assumed a cloud bottom at 1 km and surface relative humidity and temperature, respectively, of 75% and 12°C. In that scenario, a small raindrop (0.5 mm) may lose ~28% of its mass by sub-cloud evaporation that would lower its d-excess by ~10‰. For a larger drop (1 mm), ~7% of the mass may be lost with a 5‰ decrease in d-excess. For the 1 March 2009 Cazadero event, with a sub-cloud region of about 1 km and particle size mostly greater than 0.5 mm (Fig. S7), our Stewart model calculations using the observed surface conditions ($T = 11^{\circ}\text{C}$, $RH = 63\%$) show a d-excess decrease of ~2‰, less than the ~5‰ decrease in Graf et al. (2019). Sub-cloud evaporation at Cazadero on 1 March 2009 may therefore have reduced the d-excess by a maximum of 5‰.

Surface relative humidity at Rio Claro exceeded 90% during the portions of the stratiform precipitation events used in this study (Santos et al., 2024), indicating near-saturation conditions in the sub-cloud layer and consistent with the absence of decreasing reflectivity profiles in the sub-cloud region. The Stewart model calculations confirm that raindrop evaporation at $RH > 90\%$ (and a sub-cloud region of ~3 km) would result in a d-excess change of less than 1‰ regardless of drop size.

At Ny-Ålesund, the sub-cloud layer was typically less than ~500m, where rain drop evaporation potentially could occur. However, the relatively colder temperatures and higher RH documented in the radiosonde profiles (mean values of -2°C and 78–91%) would further limit any evaporative loss in the shallow sub-cloud region."

22/Fig 5 caption: Please define V_r and V_s here.

Response: OK.

24/632-633: "This implies that the isotopic analysis of precipitation sampled on an hourly or daily scale would essentially provide a 'process-weighted' value of d-excess." I agree with this sentence. Such an approach might be used to identify the potential role of fractionation by riming.

Response: We agree. We have presented process-weighted calculations but have refrained from calculating a "riming fractionation". Instead, we have presented a correlation of d-excess with the rimed mass fraction. We believe that a more comprehensive dataset of d-excess and the degree of riming in mid-latitude and tropical precipitation would be needed to better define the riming fractionation.

29/Data availability: Gathering the references in a single place is useful for the reader who would like to take advantage of those data. Please do that.

Response: The reference list identifies the data sources as [dataset].

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

9/255: "... at the inflection point _where the MDV begins to increase from its more nearly uniform values aloft (Fig. S2)." I find the language about the "vertical" gradient a bit confusing.

We have revised the text for clarity: *“Following White et al. (2002), the top of the melting layer is identified at the inflection point in the MDV vertical gradient just above the bright band, where the profile transitions from a nearly constant MDV in the snow region above to a rapidly increasing MDV within the melting layer (Fig. S2). The bottom of the melting layer is similarly identified at the inflection point just below the bright band, when all the ice particles have fully melted into smaller rain drops, and the MDV transitions from its maximum within the melting layer to the more gradual increase characteristic of the rain region below.”*

18/483: Suggested re-wording: "The average MDV in the snow region above the melting layer and the d-excess values in surface precipitation ..."

We have revised the text: *“In the snow region above the melting layer at Cazadero, the average MDV values ranged from 0.9 to 1.9 m s⁻¹ with d-excess values of -22.9 to +20.6‰ measured in surface precipitation (open squares in Fig. 5). At Rio Claro, the MDV values ranged from 1.2 to 1.6 m s⁻¹ and the d-excess from +4.5 to +21.9‰ (blue squares in Fig. 5).”*

28/725: "While riming _is more common_ in warmer conditions, ..."

Thanks. Text revised.

Reviewer #2

Isotopic Evidence for Ice Growth by Riming in Precipitation

This paper compares isotope ratios in precipitation with doppler velocity measurements and photographs of snow and precipitating ice. An anti-correlation between doppler velocity and the precipitation deuterium excess (d-excess) is presented. The paper argues that this anti-correlation indicates that d-excess in precipitation can be thought of as primarily reflecting the extent to which precipitation particles are rimed, and that any effects of moisture source region are essentially overwritten.

While I find the correlations presented fascinating, I do not feel that they substantiate the claim that d-excess is primarily an indicator of riming. I recommend reconsidering how the correlation is presented and using it as an opportunity to suggest future research steps. Furthermore, I strongly recommend that the paper cites works that have already used numerical modeling approaches to quantify the contributions of moisture source and microphysical processes to the d-excess signal. Finally, I suggest revisiting the introduction, as some of the descriptions of d-excess sensitivity do not match the literature consensus. I also strongly feel that one must be cautious in drawing parallels between hailstones over the US Great Plains and the smaller rimed particles likely to be encountered in Antarctica and Greenland.

Response: Thanks for the review and comments. The objective of our study is to address the issue of fractionation during riming. We believe that an exhaustive review of the literature on precipitation d-excess would distract from the main objective and therefore we have summarized by noting that all studies in the published literature are based on two common assumptions: that precipitation d-excess is influenced by conditions at source moisture origin

and during snow formation by vapor deposition. These assumptions do not include riming and that is the point of departure for the present study.

Please also see the common responses above for the relevance of hailstone growth studies to riming and the process-weighted change in precipitation d-excess resulting from varying proportions of vapor deposition and riming.

Specific major comments

A. d-excess sensitivity. The first part of the introduction discusses the response of d-excess to evaporation and depositional growth. Many statements in this section appear to contain errors or lack specificity.

1. For example, evaporation increases (it does not lower) the d-excess in the vapor compared to the liquid.

Response: Text revised; please see common responses.

2. Also, when describing ice crystal growth by WBF, it would be helpful to specify with respect to what is the d-excess higher (particles grown by other processes? The vapor?).

Response: Text revised; please see common responses.

3. Starting on line 61, the paper claims that the increase in d-excess growth during vapor deposition is much larger than the range of values resulting from moisture source variability; thus moisture source may only have a limited influence. However, there is no citation backing this.
4. I strongly recommend looking at papers by Zhengyu Xia (<https://doi.org/10.1029/2021GB007245>, <https://doi.org/10.1029/2022GL101901>), which attribute variations in d-excess explicitly to moisture source variations and microphysical processes.
5. Another possibly helpful resource is Gao et al. 2025 (<https://doi.org/10.1029/2024JD043047>).

Response: Thanks. We agree that moisture source origin and vapor deposition as controls on precipitation d-excess have been extensively discussed in the literature, including in the sources cited in the comment. For our study, we have cited the relevant references of Pfahl and Sodemann (2014) for present-day variability of ocean evaporation and Jouzel and Merlivat (1984) for d-excess of vapor deposited ice. The study by Xia et al. (2021) concluded that d-excess of low- and mid-latitude precipitation was non-conservative and may not simply be used as a tracer of oceanic moisture sources, consistent with the premise of our study. As we have noted, we believe that an exhaustive review of the literature would distract from the objective of this study.

B. Hailstones vs. rime. The second part of the introduction attempts to link higher delta values and lower d-excess values to riming conditions. Much of this background is pulled from older experimental studies modeling isotopic variations in hailstones, and I worry that much of it is irrelevant to the study at hand.

1. For example, I'm concerned that the conditions in which hailstones form are quite distinct from the Antarctic and Arctic conditions in which most riming discussed in this paper occurs. Specifically, I would imagine that the extent of latent heating, splashing, and evaporation of liquid water during hailstone production is much larger.
2. Similarly, are wet and dry growth regimes as relevant for polar rime production?

Response: Text revised with more details; please common responses above.

3. Keep in mind that none of these early hailstone works were able to measure the supercooled liquid water in cloud for comparison. Lowenthal et al. 2016 did in fact measure both snow and cloud condensate, however, they mostly found insignificant differences between these two reservoirs. In fact, they assumed that snow would look more like cloudwater isotopically as a way to define the extent of riming.

Response: We agree that Lowenthal and co-workers have conducted some of the most detailed field studies to characterize the effect of riming on precipitation isotope compositions. However, we respectfully disagree with the interpretation of Lowenthal et al. (2016). They noted that cloud water, sampled on the surface, and snow, which formed at higher altitudes, had different isotopic compositions. They also assumed that riming did not occur with isotopic fractionation which is implicit in their statement that rimed ice would have the same composition as cloud water. The purpose of the present study is to test that assumption and our results show that a fractionation does occur in riming.

In an earlier study, Lowenthal et al. (2011) estimated the rimed mass fraction (RMF) of snow using sulfate concentrations in snow and the cloud water. This 2011 study was conducted at the same Colorado location and season (February) as the 2016 study. The 2011 results show a positive correlation between $\delta^{18}\text{O}$ and RMF; $\delta^2\text{H}$ was not measured in the 2011 study. Assuming that the $\delta^{18}\text{O}$ – RMF correlation seen in 2011 also existed in 2016, the $\delta^{18}\text{O}$ -RMF regression from 2011 can be used to estimate RMF during the 2016 study. Measured d-excess of snow in Lowenthal et al. (2016) is then found to be negatively correlated with the estimated RMF, consistent with the results of our study.

4. Perhaps more to the point, can the data evaluated here even distinguish whether rime has a lower d-excess than the cloud material from which it formed?

Response: The objective of the paper is to show that rimed ice has a lower d-excess than unrimed ice. We have used the MDV as an independent indicator of riming and its inverse

correlation with d-excess suggests a lower d-excess of rimed ice. Please note that in this study, we have not aimed for or conducted an isotope mass-balance during riming. However, as noted in the response to common issues, the revised version presents detailed calculations of d-excess resulting from riming and vapor deposition, as well a sensitivity analysis of the influence of various input parameters, including cloud water composition.

C. Correlation not causation. Figures 3 and 4 are the primary scatter plots arguing for a casual relationship between d-excess and riming (where doppler velocity is the proxy for riming). This relationship is fascinating; however, I am not convinced that these analyses alone allow us to claim that riming is the primary driver of differences in d-excess. For consideration....

1. *How does the height of particle growth change with riming?*
2. *Is riming more customary with warmer, wetter conditions? (See Wedum, Pettersen et al. 2026 as an example <https://doi.org/10.1029/2025JD044309>.)*
3. *What synoptic conditions favor riming? J-L Bonne et al. 2015 found that warmer and moister conditions at Greenland (not only promote riming but) can also accompany significant changes in moisture source.*
4. *Finally, to what extent does below cloud exchange matter?*
5. *I don't feel that the paper can rule out these other possible factors. I thus recommend that the paper revisit how it presents its results, backing off some of the bolder claims.*
6. *It also feels like the paper largely ignores the seasonal variation in Greenland: the winter values suggest no correlation (or maybe a slightly positive one?) between d-excess and doppler velocity. How does this finding influence confidence in the results?*

Response: The revised version has been prepared to clarify the text as much as possible. For the comment related to Greenland and winter precipitation at Summit, please see common responses above.

D. Photograph patterns. I appreciate that the paper looks for specific evidence of riming in precipitation photographs, but I have a hard time understanding how the table of photos supports the claim that d-excess decreases primarily with riming (MDV, or doppler velocity). Row B and F show two very different particle types with similar MDV but very different d-excess. Row D, the moderately to heavily rimed dendrites, have large MDV but medium d-excess. Based on the scale bars, is it possible that there is a stronger connection with particle size, with smaller particles resulting in lower d-excess?

Response: The revised version has addressed the low d-excess of winter Summit precipitation in detail. Ice formation at very low temperatures results in smaller particle size in the winter that is reflected in lower MDV. Please see common responses above for d-excess of winter Summit precipitation.

Comment from Sodemann

Comments on "Isotopic evidence of ice growth by riming in precipitation" submitted to ACP by Aggarwal et al.

The authors present work that proposes to revise the interpretation of the deuterium excess in precipitation and ice cores in terms of riming versus deposition processes in clouds. Based on analysis based on Greenland summer precipitation, a global relationship is constructed (using existing published datasets) between fall velocity and d-excess. The authors propose that the riming process dominates over other factors that may influence the d-excess, such as moisture sources and moisture transport history.

It is my impression that the authors bring a potentially important factor to light that could influence the d-excess in global precipitation. The manuscript also does a great job in compiling some of the literature to relate microphysical pathways to the isotopic composition of precipitation. However, I think that the evidence presented here is not convincing enough to dismiss or rule out that other factors than the one investigated here still are more important for the d-excess in precipitation.

In particular the proposed relevance for the interpretation of ice cores is in my view stretched too far, as many other factors, including post-depositional effects such as homogenisation of the top snow layers, vapour-snow exchange, and ablation effects can play into ice core signals, but none of these is discussed. In my view, the manuscript would benefit from not claiming that riming vs. deposition is the single most important factor, and rather presenting it as another factor that should be taken into account. This would open up an alley towards further quantification of the proposed effect from laboratory and field studies.

Response: we thank Dr. Sodemann for his comments on the manuscript and appreciate his insights and suggestions. We believe we have addressed most of his concerns in the revised version as outlined in the responses to common issues above. This includes revised text that does not exclude other potential effects on precipitation d-excess. As for the d-excess of snow and ice cores in polar areas, we agree that post-depositional processes noted in the comment may slightly modify the d-excess of surface snow or ice cores. However, we do not believe that those processes can produce the significant spatial variations that we have discussed here. Our reference to the lower d-excess of ice cores from the last glacial maximum (LGM) is only to note that moisture source origin has been invoked as the reason for those lower values. The revised version also provides a detailed treatment of d-excess calculated for vapor deposition and riming growth and should help to give a more comprehensive view of in-cloud processes other than riming affecting precipitation d-excess.

Specific comments

1. The cited evidence for isotope fractionation during riming appears to rely on one single laboratory study (Bailey et al., 1969). This study and the accompanying modelling study by Jouzel (1985) were done in the context of hail formation. While hail can be seen as an end member of the processes leading to graupel formation, there is thus no direct evidence for

isotope fractionation during riming for smaller hydrometeors. The proposed process that involves fractionation due to evaporation of accreted liquid before freezing is complete should thus be presented as hypothetical. It would make the manuscript overall stronger (more valuable) if this knowledge gap is pointed out throughout the interpretation, and in the conclusions.

Response: The revised version deals with this aspect in more detail as outlined in the response to the common issues. Detailed calculations of d-excess expected from vapor deposition and riming are now included. These calculations are based on established physics of isotope fractionation and sensitivity to various input parameters is discussed.

2. The analysis presented in Table 3 and Figure 2 is interesting, but appears difficult to reproduce. Some statistics should be given of how many snow flakes have been analysed and how much variation was observed. The selection of these few selected snow samples does also not rule out that other factors, such as the d-excess induced during equilibrium fractionation (Sodemann et al., 2008; Dütsch et al., 2017) or the moisture source conditions (Pfahl and Sodemann, 2014; Sodemann et al., 2024) influenced the d-excess if this has not been explicitly investigated.

Response: We appreciate the comment. The text has been revised to indicate that the fall velocity measurements in Fig. 2 are for 3 to 93 particles, except for one day where only one measurement was made. A summary of these measurements is now included in the supplementary material. The limitation of the measurements on individual particles or of photographs in Table 3, is noted with respect to d-excess values that are measured in daily precipitation, but the correlation — in Fig. 2 or in Table 3 showing higher d-excess when unrimed particles were observed in the summer — is consistent with that shown for d-excess and MDV on a daily time scale (Figure 4 in the original version).

We have also revised the conclusions to acknowledge other effects; please see response to common issues above.

3. In L. 61 and the paragraphs before, the authors claim that "the increase in d-excess during ice growth by vapour deposition is much larger than the range of lower values resulting from source moisture difference". However, this conclusion is made from an uneven paring between the global mean d-excess variation and local supersaturation near liquid droplets. Indeed, similar or even larger super- and sub-saturation regimes ($RH < 30\%$) can exist over oceans, for example when dry and cold arctic air is advected over the ice-free ocean and then warmed up in strong-wind conditions (Duscha et al., 2022; Sodemann et al., 2024). During such situations, the d-excess from water vapour evaporating under strong non-equilibrium conditions can reach 30 ‰ or more, quite comparable to the numbers cited for deposition growth.

Response: We have revised the text for clarification; please see common responses above.

4. The introduction recounts different interpretations of the d-excess, but lacks a clear description of how the d-excess arises from differences in diffusion speed between

isotopologues during supersaturated conditions. It would be useful to include such a description to facilitate the physical interpretation of the proposed fractionation due to riming. Note that also equilibrium fractionation can induce a d-excess signal, arising from the definition of the delta scale (Dütsch et al., 2017).

Response: Text revised; please see common responses above.

5. The evidence presented in Fig. 3 is puzzling. There appears to be a relation for the summer data, but not for the winter data. The regression line is not related to the depicted summer or winter data, but taken from Fig. 2. A regression line and confidence bounds for the winter and summer data would be instructive here. However, the fact that the winter data appear to show no or even an inverse relation substantially weakens the evidence for the proposed process put forward here. If there is no consistent relation across seasons at the same site, why should the signal translate to ice cores, or be dominating at other global sites? The explanation proposed in L. 410 does not really address or remedy such concerns.

Response: We agree that the explanation for winter Summit precipitation was not convincing. We have conducted substantial additional analysis and revised the discussion as outlined in the common responses above. Figure 3 (now Fig. 4) has been revised to remove the MASC data from Fig. 2 and the regression line is based only on the summer MDV data. Based on expanded analysis and calculations based on Jouzel and Merlivat (1984), the low d-excess of winter precipitation at Summit would result from vapor deposition growth at very low temperatures, high ice supersaturation, and low $\delta^{18}\text{O}$ of vapor. This conclusion is consistent with the results in Dütsch et al. (2019; their Fig. 1).

6. The evidence based on various datasets from different climate zones presented in Fig. 4 is intriguing. Importantly though, the figure leaves the questions of how important other factors could be in shaping this relation unaddressed. Each of the sites has different moisture sources, for example, and may be dominated by different transport pathways and fractionation conditions during transport. Accordingly, the contribution of such factors to the d-excess signal observed in the precipitation can not be ruled out, unless it has been investigated. Again, the Greenland data is evidence that other factors are important for the d-excess than fall velocity (as a proxy for riming). I don't think this is a problem in itself, but again the study would be significantly more impactful if such deficiencies are acknowledged, and the implications in the abstract and conclusions revised accordingly.

Response: We agree with the comment that the site independence of the inverse correlation of MDV with precipitation d-excess, except at Summit, is striking. The apparent contradiction evident in the Summit data has been addressed as noted previously and outlined in the response to common issues.

7. In L. 612, it is stated that the Greenland winter data is excluded from the regression. This choice seems to require a more rigid justification.

Response: We had in fact excluded all Summit data. However, in the revised version, a detailed explanation is provided for the different processes of precipitation formation at Summit (see above), justifying the exclusion of the Summit data from the regression.

8. The relative location of all data points in Fig. 5 and Fig. 6 appear identical, except for the horizontal axis having a different scale. Does it make sense that the RMF obtained from the method of Kneifel and Misseev (2020) is just a linear scaling of the MDV?

Response: We agree with this comment, but RMF is the relevant parameter to be used as an observational constraint and in process-weighted calculations of precipitation d-excess presented in the revised version. However, we have shortened the narrative related to RMF and moved it to the Results section.

9. My impression is that Sec. 4.2 is not needed and opens quite a lot of topics that would need to be assessed in more detail, also taking current interpretations into account. This includes the effect of moisture source and transport changes with different climates, and post-depositional effects in low-accumulation regions of Greenland and Antarctica. Given the weak links in some of the argumentation towards the importance and general validity of the proposed riming effect in the isotope composition, as indicated in the comments above, a more tentative interpretation, taking into account such uncertainties, would appear to make for a more impactful paper, as this could trigger new research to corroborate this new hypothesis.

Response: We have shortened the narrative substantially and moved the figures to supplementary material, along with the following statement in the “Discussion/Conclusion” sections: “The conventional attribution of spatial d-excess gradients to climatic conditions at source moisture origin implicitly assumes that in-cloud microphysical processes either do not vary systematically across the gradient or are negligible relative to the source signal. Our results suggest that this assumption warrants re-examination.”

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