

Deleted: Isotopic Evidence for Ice Growth by Riming in Precipitation¶

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

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Abstract

In this study, we investigate the impact of riming on the ~~deuterium excess~~ (d-excess = $\delta^2\text{H} - 8 \cdot \delta^{18}\text{O}$) of precipitation. In mixed-phase clouds, precipitation forms by vapor deposition — where supercooled liquid droplets do not come in contact with ice particles — and by riming, where droplets freeze directly on particle surfaces. While vapor deposited ice generally has higher d-excess than liquid, riming is assumed to occur without isotopic fractionation. We correlated mean Doppler velocity (MDV), an independent indicator of riming, with precipitation d-excess (–23 to +45‰) at polar (Summit, Greenland; Ny-Ålesund and Andenes, Norway, Dumont d’Urville, Antarctica), mid-latitude (Cazadero, California) and tropical (Rio Claro, Brazil) sites. The d-excess decreases with increasing MDV (or riming intensity) at all locations, except for winter precipitation at Summit. The low d-excess of rimed ice is consistent with the evaporation of accreted liquid on particle surface before freezing is complete. For Summit winter, cold temperatures and low $\delta^{18}\text{O}$ of vapor suppress d-excess independently of riming. Calculations show that mixtures of ice growing by riming and vapor deposition (including diamond dust) can produce the observed range of precipitation d-excess in this study. We conclude that riming-driven lowering of d-excess can explain, at least in part, the spatial gradients in surface snow d-excess across Greenland and Antarctica previously attributed to changes in source moisture origin, and the very low d-excess of rainfall attributed solely to sub-cloud evaporation. Precipitation d-excess can be used to estimate the rimed mass fraction, providing observational constraints for improving microphysics schemes in climate models.

75 **1. Introduction**

76 Polar ice cores have long been known to record temperature changes in past climates based on their oxygen ($\delta^{18}\text{O}$) and
77 hydrogen ($\delta^2\text{H}$) isotope compositions (e.g., Dansgaard et al., 1969; Jouzel et al., 2013). The relative composition of these
78 isotopes, namely d-excess (defined as $\delta^2\text{H} - 8 \cdot \delta^{18}\text{O}$), is believed to provide insights into the climatic conditions
79 (temperature, relative humidity, windspeed) of source moisture origin (Dansgaard, 1964; Masson-Delmotte et al., 2005;
80 Jouzel et al., 2013). Lower d-excess of Greenland and Antarctic ice cores from the Last Glacial Maximum (LGM) compared
81 to the Holocene, or in the warming phase compared to the cooling phase of abrupt climate change events in Greenland
82 (Jouzel et al., 2007), has been suggested to indicate substantial and rapid reorganization of the high-latitude hydrological
83 cycles that changed the geographical region of source moisture origin (Johnsen et al., 1989; Jouzel et al., 2007). In addition
84 to temporal differences, the northward increase in d-excess of surface snow across Greenland is attributed to differences in
85 source moisture origin (Johnsen et al., 1989; Masson-Delmotte et al., 2005).

86
87 The interpretation of the d-excess of precipitation and ice cores is guided by two primary constraints (Fig. S1,
88 Supplementary Material). The first constraint is that d-excess is inherited from the oceanic evaporation source moisture. The
89 global average precipitation d-excess of $\sim +10\text{‰}$ (Dansgaard, 1964) reflects the differential kinetic fractionation of ^2H
90 relative to ^{18}O under mean oceanic evaporation conditions (Merlivat and Jouzel, 1979). During evaporation, H_2O diffuses
91 faster than both H_2^{18}O and HDO across the vapor–liquid interface, so the vapor is depleted in ^{18}O and ^2H relative to the
92 liquid. However, because HDO diffuses faster than H_2^{18}O (Merlivat, 1978), the resulting d-excess of the vapor is higher than
93 that of the liquid. The kinetic increase in vapor d-excess is greater when evaporation occurs at lower relative humidity or
94 warmer sea surface temperatures (Dansgaard, 1964; Merlivat and Jouzel, 1979). In the current climate, oceanic vapor is
95 estimated to have a d-excess range of $\sim +8$ to $+13\text{‰}$ (northern hemisphere) and $\sim +9$ to $+12\text{‰}$ (southern hemisphere),
96 suggesting a seasonal d-excess variability of ~ 3 to 5‰ (Pfahl and Sodemann, 2014). Ocean evaporation in dry conditions
97 ($\sim 30\%$ relative humidity) may result in vapor with d-excess of about $+25\text{‰}$ (Pfahl and Sodemann, 2014). However,
98 precipitation d-excess in the adjacent land areas generally does not correspond to such higher d-excess values because
99 precipitation results from a mixture of moisture sources with different evaporation histories (Pfahl and Sodemann, 2014).

100
101 The second constraint is related to ice growth in mixed phase clouds, where a supercooled liquid phase co-exists with ice and
102 vapor at temperatures above approximately -38°C (Pruppacher and Klett, 2010; Houze, 2014); there, when ice crystals grow
103 by vapor deposition (i.e. the Wegener-Bergeron-Findeisen [WBF] process), the d-excess of ice is higher than the co-existing
104 liquid or vapor. In the WBF process, liquid droplets do not come in direct contact with the ice particles, but instead evaporate
105 owing to a higher saturation vapor pressure over liquid than ice. The vapor then diffuses and is deposited on ice particles
106 (Houze, 2014; Pruppacher and Klett, 2010). As HDO diffuses faster than H_2^{18}O , the d-excess of ice may be $\sim 30\text{‰}$ higher
107 than that of the liquid, depending upon the level of supersaturation in the vapor with respect to ice (Jouzel and Merlivat,
108 1984; Uemura et al., 2005; Casado et al., 2016).

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45 Because of this large fractionation during ice growth by vapor deposition, source moisture d-excess may be more directly
46 reflected in “warm rain” that does not involve an ice phase. However, precipitation over land and oceans forms dominantly
47 by ice-phase processes (Mülmenstädt et al., 2015). Even in the tropics where warm rain is most prevalent, clouds producing
48 only warm rain are largely confined to shallow isolated oceanic convective cells and onshore tropical flow over land
49 (Schumacher and Houze, 2003; Mülmenstädt et al., 2015). This implies that the lower d-excess commonly observed in
50 precipitation cannot be explained by source moisture conditions alone and must reflect an additional in-cloud process that
51 reduces d-excess.

52

53 In addition to the WBF process, ice growth in mixed phase clouds can occur by riming. Unlike the WBF process,
54 riming involves the collision of ice particles and liquid droplets, leading to the direct freezing of
55 droplets on particle surfaces (Pruppacher and Klett, 2010; Houze, 2014). Collisions between ice particles form
56 aggregates or snow-flakes (Pruppacher and Klett, 2010); the colliding ice particles themselves may have
57 formed by the WBF or riming process.

58

59 A fundamental assumption in existing interpretations of precipitation d-excess is that riming occurs without an isotopic
60 fractionation (Federer et al., 1982; Jouzel and Merlivat, 1984; Ciais and Jouzel, 1994; Bailey et al., 2025). That is, the rimed
61 ice has the same isotopic composition and d-excess as the liquid. To the best of our knowledge, this assumption has not been
62 thoroughly explored in the literature.

63

64 Evidence from several field, laboratory and modeling studies suggests that isotopic fractionation may
65 indeed be involved in riming. For example, Warburton and deFelice (1986) and Demoz et al. (1991)
66 analyzed freshly fallen snow in the Sierra Nevada mountains and observed that the $\delta^{18}\text{O}$ of snow was
67 lower when ice crystal habits were typical of vapor deposition (dendrites, columns and plates) compared
68 to graupel and other rimed particles. Lowenthal et al. (2011) used sulfate concentrations in cloud water
69 and Rocky Mountain snow to estimate the fraction of ice that formed by riming and observed that the
70 $\delta^{18}\text{O}$ of snow increased as the rimed fraction increased. Bailey et al. (1969) conducted laboratory
71 experiments in an icing tunnel (air temperature = -20°C) to study isotopic fractionation during the
72 freezing of supercooled droplets on a cold surface (riming). The experiments were conducted at liquid
73 water concentrations of 1 or 5 g m⁻³. At the lower water content, the droplets froze completely and
74 rapidly, while at the higher water content, the supercooled liquid droplets were considered to have

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10 partially evaporated before freezing was complete. In both experiments, the $\delta^{18}\text{O}$ and $\delta^2\text{H}$ values of the
11 frozen material were slightly higher than those of the supercooled liquid. A modeling study by Jouzel et
12 al. (1985) corroborated the laboratory results of Bailey et al. (1969) and suggested that the higher δ values of
13 the rimed ice likely result from the evaporation of the accreted liquid and/or its partial freezing.

14
15 Studies of the mechanism of riming (e.g., Macklin and Payne, 1967; Pruppacher and Klett, 2010, and references therein)
16 suggest that when a liquid droplet collides with an ice particle, it may lose some water by splashing, depending on the impact
17 velocity. The drop initially freezes with a thin frozen shell as the latent heat of fusion released in the process warms the inner
18 part of the droplet. The colliding droplet may also spread to form a thin film on the particle surface. Heat dissipation then
19 occurs by conduction — raising the particle surface temperature above that of the surrounding air — and by evaporation
20 when the air is subsaturated with water vapor. If the latent heat is dissipated fast enough to keep the particle surface
21 temperature a few degrees below 0°C , the accreted water freezes in a "dry" growth regime, with or without the loss of some
22 accreted water by shedding. Conversely, if dissipation is slower — for example, due to high droplet concentration or
23 increased frequency of droplet-particle collisions — the particle surface temperature may rise to 0°C . In that case, the
24 accreted water freezes in a "wet" growth regime and some water may be lost by evaporation before freezing is complete. The
25 water loss by splashing, shedding, and evaporation may be as much as 30% or higher, depending upon the size and structure
26 of the ice crystals, the impact velocity of droplets, the liquid water content, and other physical parameters (Pruppacher and
27 Klett, 2010).

28
29 More significantly than the slightly higher δ values, the d-excess of rimed ice in the laboratory and modeling studies was
30 lower than that of the supercooled liquid. Natural hailstones analyzed by Jouzel et al. (1985) had low ($<+10\%$) d-excess
31 values. Increased riming of snow from the Rocky Mountains (Lowenthal et al., 2011) also appears to correspond to lower d-
32 excess values reported in Lowenthal et al. (2016). Studies of isotope fractionation during riming were conducted in the
33 context of the growth of hailstones, which are an extreme case of riming distinguished only by their much larger size (>5
34 mm) compared to rimed particles (Pruppacher and Klett, 2010). A continuum of ice particles from unrimed to graupel and
35 hail has been shown based on their mass-size relationships (Lin and Colle, 2011) and the results of hailstone studies are
36 equally applicable to the riming of smaller particles in mixed phase clouds. Wet-growth (particle surface temperature, $T_s =$
37 0°C) with partial evaporation of the accreted liquid requires that the supercooled water collection rate exceed the rate at
38 which it can be frozen. Calculations using a heat balance equation (Pruppacher and Klett, 2010) show that for small particles
39 of ~ 2 mm or less, and the relatively low liquid water contents (LWC) that are generally observed in stratiform clouds, wet-
40 growth may occur at air temperatures greater than about -10 to -15°C . Below that temperature, riming would occur by a
41 dry-growth process.

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Ice growth by riming frequently occurs in precipitation at all latitudes and across various meteorological conditions (Korolev et al., 2017). It is estimated that in mixed phase regions, the riming process is active in about half of the stratiform clouds, and almost all of the convective clouds. For example, riming is estimated to contribute about 30 to 50% of the surface snowfall mass in the Sierra Nevada Mountains (Mitchell et al., 1990; Lowenthal et al., 2011) and near Sapporo, Japan (Harimaya and Sato, 1989). Arctic precipitation at Hyytiälä, Finland and Ny-Ålesund, Svalbard includes up to 40% of the snow mass from riming (Moisseev et al., 2017; Chellini and Kneifel, 2024). At Oliktok Point, Alaska, USA, about two thirds of the Arctic precipitation forms by riming (Fitch and Garrett, 2022). Riming has been observed in snowfall on the South Pole (Ohtake, 1978). In eastern Antarctic coastal precipitation at Dumont d'Urville, a mean riming growth of about 30% was estimated with ~11% of the rimed particles being fully developed graupel (Grazioli et al., 2017).

Riming increases the particle mass and density and the particle becomes more rounded in shape (Pruppacher and Klett, 2010; Houze, 2014). These changes result in a generally higher terminal fall velocity of rimed particles compared to unrimed particles, which grow by vapor deposition (Locatelli and Hobbs, 1974; Weiss et al., 1977; Heymsfield and Kajikawa, 1987; Pruppacher and Klett, 2010; Heymsfield et al., 2013; Garrett and Yuter, 2014; Houze, 2014; Kneifel and Moisseev, 2020; Matrosov, 2023). A lightly rimed snowflake might fall at 0.5–1 m s⁻¹ while a dense graupel particle falls at >1.5 m s⁻¹ (Locatelli and Hobbs, 1974). So, terminal velocity is essentially a proxy for the degree of riming.

While the fall velocity of rimed or unrimed particles is dependent on physical characteristics (size, shape, mass and density), the d-excess would respond to the microphysical process (WBF or riming) responsible for the phase change from vapor or liquid to ice. Consequently, it may be possible to distinguish d-excess change resulting from ice growth purely by the WBF process (unrimed) from that by riming based on the higher fall velocities (>1 ms⁻¹) of rimed particles.

In this study, we use the terminal fall velocity as an independent parameter to explore the impact of riming on d-excess in polar to tropical precipitation (observed d-excess range ~−23 to +45‰). We have also used photographic images of precipitating snowflakes to document riming and vertical profiles of temperature and humidity derived from radiosonde soundings to characterize cloud processes at the polar locations in our study. Our findings reveal that precipitation d-excess values decrease with increasing terminal fall velocity or the fraction of particle mass attributed to riming. We conclude that d-excess is a powerful indicator of riming or vapor deposition growth during precipitation formation and may be used to characterize changes in cloud microphysical processes at a variety of spatial or temporal scales, with implications for the interpretation of d-excess records in ice cores and other high-resolution paleoclimate archives.

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A lower d-excess of rimed ice is consistent with the mechanism of riming growth that involves evaporation and partial loss of the accreted water (e.g., Macklin and Payne, 1967; Pruppacher and Klett, 2010 and references therein). Briefly, when a liquid droplet collides with an ice particle, it may lose some of the water by splashing, depending on the impact velocity.

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51 2 Data and Methods

52 2.1 Study Locations

53 To test our hypothesis that d-excess values in precipitation reflect microphysical processes associated with riming, it is
54 important to consider concurrent data for isotope compositions and terminal fall velocities on a daily or shorter time scale;
55 this approach minimizes data averaging much beyond the precipitation events where isotope compositions are acquired.
56 Likewise, because the nature of cloud processes is essentially the same on the global scale (Pruppacher and Klett, 2010;
57 Houze, 2014), data from the polar to the tropical regions would be important. Table 1 lists the coordinates and Fig. 1 shows
58 the locations where we have successfully compiled concurrent isotope and terminal fall velocity data. In addition, vertical
59 profiles of temperature and relative humidity were obtained for the polar locations to characterize the cloud microphysical
60 processes.

62 Table 1. Coordinates and surface elevations (meters above sea level) of study locations.

63

Location	Latitude	Longitude	Elevation (masl)
Polar			
Summit, Greenland	72° 36' N	38° 30' W	3250
Ny-Ålesund, Svalbard, Norway	78° 55' N	11° 56' E	11
Andenes, Norway	69° 14' N	15° 41' E	10
Dumont d'Urville, Antarctica	66° 39' S	140° 0' E	41
Mid-latitudes			
Cazadero, California	38° 36' N	123° 13' W	475
Tropics			
Rio Claro, Brazil	22° 39' S	47° 54' W	670



66 Figure 1. Map showing the study locations (base map downloaded from <https://www.freeworldmaps.net/printable/hammer-worldmap-hd.jpg>).

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2.2 Terminal Fall Velocity

The terminal fall velocity of ice particles is commonly determined using vertically-pointing ground-based Doppler radars (Houze, 2014) or particle imaging devices such as a multi-angle snowflake camera, MASC (Garrett and Yuter, 2014; Praz et al., 2017). Doppler velocities from profiling radars have been reported at each of the study locations for all or part of the same time period as the isotope samples. In addition, a limited amount of concurrent terminal fall velocity data acquired with a MASC were retrieved for Summit, Greenland.

2.2.1 Doppler velocity and terminal fall speed in stratiform clouds

The use of Doppler radars for measuring the terminal fall velocities of hydrometeors has been extensively described in the literature (e.g., Weiss and Hobbs, 1975; Orr and Kropfli, 1999; Protat and Williams, 2011; Houze, 2014). Briefly, Doppler radars used for cloud and precipitation studies emit microwave radiation that is reflected by various hydrometeors (snow, ice or rain drops) and the travel time, strength, and phase of the returned signal are used to determine the altitude (or range), reflectivity, and Doppler velocity of the reflecting objects (Houze, 2014). The reflectivity values are reported as the “effective reflectivity” (Z_e) in units of $\text{mm}^6 \text{m}^{-3}$ (or in decibels, $\text{dBZ} = 10 \log_{10} Z_e$). The Doppler velocity is denoted as the mean Doppler velocity (MDV) with units of ms^{-1} and is the reflectivity-weighted mean of the measured Doppler spectrum. An altitude correction is applied to Doppler velocities owing to the lower air density (pressure) at higher elevations (Heymsfield et al., 2013): $\text{MDV}_o = \text{MDV}_h \left(\frac{\rho_o}{\rho_h} \right)^{-0.4} \approx \text{MDV}_h \left(\frac{1000}{P_h} \right)^{-0.4}$. Here, ρ is air density, P is atmospheric pressure, and the subscripts o and h , respectively, are mean sea level or the height where the measurement is made. Note that we are using the convention that positive MDV is towards the radar such that lower MDV values indicate weaker downward motion.

The mean Doppler velocity consists of two components: the terminal fall velocity of the hydrometeor (V_t) and the vertical air motion (V_a): $\text{MDV} = V_t + V_a$ (Orr and Kropfli, 1999; Houze, 2014). In stratiform clouds, small-scale vertical air motions are both upwards and downwards (Orr and Kropfli, 1999; Houze, 2014). It has been shown that these small-scale air motions cancel out when MDV is averaged over a period of at least 20–30 min in stratiform or anvil cloud regions (Mosimann, 1995; Orr and Kropfli, 1999; Protat and Williams, 2011). Then, $\text{MDV} \approx V_t$. The assumption of cancelling vertical air motions is not valid for convective clouds where downdrafts and updrafts are significantly higher and persistent, such that the use of

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76 MDV for characterizing the rimed nature of snow particles is only feasible for stratiform precipitation (Mosimann, 1995; Orr
77 and Kropfli, 1999).

78
79 Terminal velocities and particle size distributions from field campaigns conducted over a wide temperature range (−86° to
80 0°C) were evaluated by Heymsfield et al. (2013). All else being equal (e.g., particle shape, degree of riming), the MDV (and
81 reflectivity) is lower when precipitation consists of smaller particles, which in general are produced in colder clouds. As a
82 result, seasonal differences in particle size may **potentially** mask the increase in MDV due to riming (Heymsfield et al.,
83 2013; Chellini and Kneifel, 2024).

84
85 **2.2.2 Doppler velocity and precipitation phase**

86
87 While d-excess is measured in precipitation samples collected on the ground, the altitude at which the MDV values can be
88 used to characterize riming depends on the phase (snow or rain) of precipitation. For snowfall, MDV close to the ground at a
89 height free of ground interference can be used (Weiss and Hobbs, 1975; Orr and Kropfli, 1999). In the case of rainfall,
90 however, the MDV aloft in the snow region above the melting level is used (Fig. S2).

91
92 Stratiform rain results from the melting of snow particles in an ~500 m thick layer **called the melting layer** (Fig. S2;
93 Pruppacher and Klett, 2010; Houze, 2014). The height of the melting layer above ground is **typically** less than about 3 km in
94 mid- to high-latitudes and **between 4 and 5 km** in the tropics. Within the melting layer, the snowflakes generally melt
95 without further growth or breaking into smaller drops (Ohtake, 1969; Karrer et al., 2022). Weiss et al. (1977) investigated the
96 use of MDV during rainfall to determine the rimed nature of snow particles above the melting layer. They used the fall
97 velocity of rimed and unrimed snow particles of known mass measured in situ by Locatelli **and Hobbs** (1974) as the fall
98 velocity of snow (V_s) above the melting layer. The relationship between raindrop mass and fall velocity (Atlas et al., 1973)
99 was used to obtain the fall velocity of raindrops just below the melting layer (V_r). Based on a correlation of V_s versus V_r ,
00 graupel and heavily rimed particles could be differentiated from unrimed or lightly rimed aggregates, as validated against
01 field measurements (Weiss et al., 1977). Mosimann (1995) and Zawadzki et al. (2005) have also suggested that riming
02 during rainfall can be characterized **using particle fall velocities in the snow region**.

03
04 The melting layer is identified by a distinct peak in **the profile of** radar reflectivity or “bright band” (Fig. S2; Pruppacher and
05 Klett, 2010; Houze, 2014), **which occurs when ice particles become coated in water and appear brighter to the radar because**
06 **of the dielectric difference between water and ice. Following** White et al. (2002), the top of the melting layer **is identified at**
07 the inflection point in the MDV **vertical gradient just above the bright band, where the profile transitions** from a nearly
08 **constant MDV in the snow region above to a rapidly increasing MDV within the melting layer** (Fig. S2). **The bottom of the**

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21 melting layer is similarly identified at the inflection point just below the bright band, when all the ice particles have fully
22 melted into smaller rain drops, and the MDV transitions from its maximum within the melting layer to the more gradual
23 increase characteristic of the rain region below. We used this approach to retrieve the MDV above the melting layer at
24 Cazadero, California and Rio Claro, Brazil where all surface precipitation was in the form of rainfall. The same approach
25 was used for Ny-Ålesund and Andenes, Norway when rainfall at the surface was indicated by a bright band aloft in the
26 reflectivity profiles.

27
28 **2.2.3 MASC fall velocity and snowflake photographs at Summit**

29 Particle fall velocities for some of the days of isotope sampling at Summit were available from a Multi-Angle Snowflake
30 Camera (MASC). A MASC captures photographs of snow particles in free fall from multiple angles while simultaneously
31 measuring their fall velocity (Garrett and Yuter, 2014). The cameras are automatically triggered by near-infrared detectors
32 that are vertically offset by 3.2 cm. The minimum particle size detected by the MASC is 0.1 mm. The fall velocity is
33 calculated by the time difference when the upper and lower detectors are triggered successively within 1 s, equivalent to a
34 minimum fall speed of 0.03 m s⁻¹.

35
36 In addition to the photographs captured by the MASC, an ice particle imaging camera (IcePIC), which consisted of a
37 manually operated camera attached to a microscope, is a part of the instrument cluster of the ICECAPS observatory at
38 Summit (Shupe et al., 2013). The device is housed in a wooden shed that prevents contamination from blowing snow during
39 the collection of images. Falling snow was collected on glass slides that were cleaned with isopropyl/glycol mixture, dried,
40 and placed on a wooden table. This table, however, was in an open area where blowing snow contamination on the slide was
41 possible. To minimize this effect, the slides were placed on either side of a vertical barrier to be
42 differentiated as upwind or downwind locations. Sample accumulation times ranged from a few minutes
43 to several hours depending upon precipitation rate. The slides were then observed with the IcePIC system.
44 The microscope stage and air around the device were at or near ambient temperature. The IcePIC photographs and
45 accompanying notes from the microscopic examination were retrieved from the ICECAPS database
46 (<https://downloads.psl.noaa.gov/psd3/arctic/summit/icepic>) on 31 July 2023. The photographs may be
47 used to qualitatively characterize riming on ice crystals during precipitation events, but may not be
48 representative of particle habits in daily precipitation.

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51 **2.3 Doppler velocity and isotope data at the study locations**

52 Table 2 shows the details of the vertically-pointing Doppler radars deployed at the locations in this study
53 along with the sources of isotope and radar data. Isotopic compositions of precipitation were obtained from
54 published studies. The oxygen and hydrogen isotope analyses were conducted by standard methods using mass-
55 spectrometers or laser analyzers and the analytical details are available in the original publications. As noted previously, d-
56 excess is defined as: $d(\text{‰}) = \delta^2\text{H} - 8 \cdot \delta^{18}\text{O}$, where $\delta = (R_{\text{sample}} / R_{\text{standard}} - 1) \cdot 1000$, and R is the isotope ratio ($^{18}\text{O}/^{16}\text{O}$) or
57 ($^2\text{H}/^1\text{H}$) in a sample or the VSMOW (Vienna Standard Mean Ocean Water) isotope standard.
58
59 The dates and times of data averaging for reflectivity and MDV are provided in Table S1. At the polar locations,
60 precipitation samples were collected on a daily (~24 hr) time scale while those at the mid-latitude and tropical locations were
61 collected at a higher frequency (~30 min or less). We note that daily isotope sampling at the polar locations includes parts of
62 two calendar days. To be precise, we would average the MDV over the same 24-hr period. However, radar data were not
63 always available for two consecutive days and in some cases, there was no or little precipitation on the first day. Sensitivity
64 testing with available data for 24-hr average MDV over consecutive days of precipitation indicated a negligible difference
65 from the daily average for one calendar day.

66
67
68 **Table 2. Characteristics of Doppler radar and sources of isotope or radar data at the study locations**
69

Location	Frequency (GHz)	Resolution Height (m)	Time (s)	Range (m)	Data Sources
Summit	35	45	2	6000	Kopec et al., 2019; ARM, 2010
Ny-Ålesund	24.1	30	60	930/1500	Mellat et al., 2021; Leroy-Dos Santos et al., 2020; Seidl et al., 2024; Ebell et al., 2023; NCPOR, 2026
Andenes	35	30	2	6000	Seidl et al., 2024; ARM, 2019
Dumont d'Urville	24.1	100	60	3100	Leroy-Dos Santos et al., 2023; Wiener et al. (2024)
Cazadero	2.875	60	30	10162	Coplen et al., 2015
Rio Claro	24.1	150/300	60	4650/9300	Santos et al., 2024

70
71
72 **2.3.1 Summit**

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78 Kopeck et al. (2019) reported the isotopic compositions of daily precipitation between July 2011 – September 2014. The d-
79 excess values range from +6.4 to +34.9‰ in the summer (June, July, August or JJA) and from –10.6 to +15.9‰ in the winter
80 (December, January, February or DJF). During the other seasons, d-excess values range from –9.3 to +25.4‰.

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81
82 Snowfall at Summit is generally weak (Shupe et al., 2013) and average daily values of reflectivity and MDV were calculated
83 for the days concurrent with isotope sampling. The reflectivity and terminal fall velocity at Summit increase as snow
84 particles fall toward the ground surface, indicating continuous particle growth (Castellani et al., 2015; Pettersen et al., 2018).
85 Therefore, the lowest radar level (~200 m) was used that is free of ground clutter (Shupe et al., 2013; Castellani et al., 2015).
86 An air density correction was applied to MDV values as the ground elevation at Summit station is 3250 m.

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87
88 **2.3.2 Ny-Ålesund and Andenes**

89 Leroy-Dos Santos et al. (2020) reported the isotopic compositions of daily precipitation during 2014 – 2018 from Ny-
90 Ålesund, located on the west coast of Svalbard, Norway. The samples consisted of snow, rain and ‘melt’, likely referring to a
91 mixture of rain and snow. Smaller datasets of rain and snow samples were reported for July-August 2018 (Mellat et al.,
92 2021) and February – March 2020 (Seidl et al., 2024). Isotopic composition of Andenes precipitation (rain and snow) for
93 February – March 2020 also was reported by Seidl et al. (2024). The 2020 dataset from Ny-Ålesund and Andenes included a
94 limited number of sub-daily samples. The d-excess values for the entire Ny-Ålesund dataset ranged from –55.8 to +37.6‰.
95 The lowest values were measured for snow samples collected on 2016-12-12 (–55.8‰) and 2018-04-14 (–51.7‰) at Ny-
96 Ålesund. Samples selected for this study (based on radar data availability) have a d-excess range from –8.2 to +45.2‰ at
97 Ny-Ålesund and from –1.0 to +30.2‰ at Andenes.

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98
99 Doppler radar data at Ny-Ålesund were obtained from two co-located MRR-2 instruments deployed by research groups from
00 Germany (for 2017-2020) and India (2014-2017). The results from the two instruments were compared on overlapping
01 measurement days; the inter-instrument mean MDV difference (0.023 m s⁻¹) was negligible and no offset correction was
02 applied.

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Table 2. Characteristics of Doppler radar and sources of data at the

03 Daily or sub-daily averages of reflectivity and MDV were obtained for the days concurrent with isotope sampling. The
04 precipitation phase was identified from the presence or absence of a bright band in vertical profiles of reflectivity. When
05 present, the top of the bright band generally was at about 600 – 900 m above ground. As both snow and rain at Ny-Ålesund
06 and Andenes may occur within a single day, we also evaluated sub-daily vertical profiles of reflectivity and Doppler velocity
07 in the 0 – 12 hr and 12 – 24 hr time intervals. In many cases, the bright band occurred only in one interval and the radar
08 reflectivity and MDV profiles generally were not usable for correlation with the daily values of d-excess, except for some of
09 the 2020 samples that were collected at sub-daily intervals (Table S1). These constraints on radar data did not allow for the
10 use of the lowest d-excess samples (< –10‰) in this study. For snow precipitation, the 150-m level above ground was used
11 to avoid ground interference with the radar measurements at lower levels.

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25

26 **2.3.3 Dumont d'Urville,**

27 The isotopic compositions of daily precipitation from January – July 2019 and January – December 2020 were reported by
28 Leroy-Dos Santos et al. (2023). Wiener et al. (2024) reported the corresponding radar reflectivity and Doppler velocity. As
29 precipitation at Dumont d'Urville is stratiform (Wiener et al., 2024), daily averages of reflectivity and MDV were calculated.
30 These values increase with decreasing altitude up to about 1 km above ground, consistent with snow particles growing as
31 they fall towards the ground surface. At lower heights, sub-cloud sublimation of snow may be significant due to strong
32 katabatic winds blowing off of the Antarctic continent (Grazioli et al., 2017; Wiener et al., 2024).

33
34 Sublimation decreases the reflectivity of snow at lower altitudes (Duran-Alarcon et al., 2019). Katabatic winds may
35 consistently increase the Doppler velocity (Wiener et al., 2024) and it may no longer be equated with the terminal fall
36 velocity. We used the vertical profiles of reflectivity below 2.5 km to exclude sublimation-affected data (Duran-Alarcon et
37 al., 2019). The profiles could be classified into two groups. One group shows a substantial decrease below ~1 km that
38 indicates sublimation (Fig. S3). In the second group, reflectivity below ~1 km did not change significantly and the MDV
39 values were retrieved for the 300-m level, which is the lowest level at this location free of ground interference with radar
40 measurements (Wiener et al., 2024), and avoids the katabatic layer that affects the lowest ~200–500m at this location
41 (Vignon et al., 2019). The d-excess of the selected samples of daily precipitation ranges from –5.1 to +14.0‰.

42
43 **2.3.4 Cazadero,**

44 Coplen et al. (2015) provided the isotopic compositions of precipitation from multiple events between January to March over
45 the 2005 – 2010 period and radar reflectivity and Doppler velocity data for several of these events were available. The
46 precipitation samples were collected with an automated sampling device at ~30-min intervals. We selected the time intervals
47 where a bright band in radar reflectivity was clearly present and estimated the top of the melting layer using the vertical
48 gradients of MDV and reflectivity (White et al., 2002). Average MDV values just above this height were calculated for 30-
49 min intervals concurrent with isotope samples. The top of the melting layer for periods of interest in this study was ~2 to 3
50 km above ground and the MDV values were adjusted for air density at that altitude. The d-excess of samples selected for this
51 study ranged from –22.9 to +20.5‰.

52
53 **2.3.5 Rio Claro**

54 The isotopic composition of tropical rainfall at Rio Claro, Brazil was reported by Santos et al. (2024). Rainfall was sampled
55 at 5-10 min intervals during precipitation events lasting for about an hour to several hours. Three events (8 October 2019, 10
56 December 2019 and 5 January 2020) had a sufficiently long stratiform period to allow averaging over 25- to 30-minute
57 intervals. The top of the melting layer, determined using the same approach outlined above for Cazadero, ranged from ~4 to

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69 4.5 km and an altitude correction was applied to average MDV values. The d-excess values of samples from the selected
70 events ranged from +4.5 to +21.9‰.
71
72 **2.4 Temperature and Relative Humidity**
73 Vertical profiles of temperature (T) and relative humidity (RH) from radiosonde soundings were retrieved for Summit
74 (Shupe et al., 2013), Ny-Ålesund (Maturilli and Kayser, 2016, 2017; Maturilli and Dünschede, 2023) and Dumont d'Urville
75 (Météo-France: <https://donneespubliques.meteofrance.fr/>; accessed on 12 June 2026). Height-resolved ice supersaturation
76 index (SI) profiles were computed from daily radiosonde soundings interpolated to a uniform 100 m vertical grid and
77 matched to precipitation isotope sample dates. SI was defined as $(e_{\text{actual}}/e_{\text{ice(T)}} - 1) \cdot 100$, where $e_{\text{actual}} = (RH/100) \cdot e_{\text{w(T)}}$ is the
78 vapor pressure of water vapor in the air computed from the radiosonde-measured RH and temperature, $e_{\text{ice(T)}}$ and $e_{\text{w(T)}}$ are,
79 respectively, the saturation vapor pressure over ice and over liquid water at the given temperature, both computed following
80 Murphy and Koop (2005). Ice supersaturation is expressed in two equivalent forms in this paper: as a percentage (SI) for
81 radiosonde profile analysis, and as a dimensionless ratio $S_i = 1 + SI/100$ in the isotope fractionation equations (Jouzel and
82 Merlivat, 1984). We also define SI_w as the value of SI at liquid water saturation: $SI_w = (e_{\text{w(T)}/e_{\text{ice(T)}}} - 1) \cdot 100$. Because e_{actual}
83 $= (RH/100) \cdot e_{\text{w(T)}}$, it follows that $SI/SI_w = RH/100$ — that is, the ratio SI/SI_w is numerically equal to the fractional relative
84 humidity with respect to liquid water, reaching 1.0 when $RH = 100\%$. As SI/SI_w approaches 1, supercooled liquid droplets
85 can coexist with ice crystals and riming growth would be enabled (Korolev and Mazin, 2003). When $SI > 0$ but $SI/SI_w \ll 1$,
86 the air is supersaturated with ice but well below saturation with liquid water and ice growth occurs predominantly by vapor
87 deposition.

88 3 Results

89 We first examine the correlation of d-excess with riming intensity at Summit, Greenland using photographs, fall velocity
90 from the Multi-Angle Snowflake Camera (MASC), and MDV, from vertically pointing radar, followed by an examination of
91 cloud microphysical conditions using radiosonde soundings. We then present d-excess-MDV correlations at the remaining
92 sites and radiosonde-derived cloud profiles at Ny-Ålesund and Dumont d'Urville. Finally, we examine the effects of sub-
93 cloud rain evaporation and in-cloud processes on d-excess variability, including a process-weighted calculation of d-excess
94 based on the vapor deposition and riming growth frameworks of Jouzel and Merlivat (1984) and Jouzel et al. (1985).

96 3.1 Riming and d-excess at Summit

97 3.1.1 Snow particle photographs

98 We reviewed hundreds of IcePIC photos for the 2011-2014 period that were concurrent with the isotope samples. Selected
99 images corresponding to a range of d-excess values in daily precipitation are shown in Table 3. High d-excess ($\sim +22\%$)
00 occurs in the summer (JJA) when snowfall contains unrimed single crystals or aggregates of various shapes, including

Moved up [12]: The cameras are automatically triggered by near-infrared detectors that are vertically offset by 3.2 cm. The minimum particle size detected by the MASC is 0.1 mm. The fall velocity is calculated by the time difference when the upper and lower detectors are triggered successively within 1 s, equivalent to a minimum fall speed of 0.03

Moved up [13]: ice particle imaging camera (IcePIC), which consisted of a manually operated camera attached to a microscope, is a part of the instrument cluster of the ICECAPS observatory at Summit (Shupe et al., 2013). The device is housed in a wooden shed that prevents contamination from blowing snow during the collection of images. Falling snow was collected on glass slides that were cleaned with isopropyl/glycol mixture, dried, and placed on a wooden table. This table, however, was in an open area where blowing snow contamination on the slide was possible. To minimize this effect, the slides were placed on either side of a vertical barrier to be differentiated as upwind or downwind locations. Sample accumulation times ranged from a few minutes to several hours depending upon precipitation rate. The slides were then observed with the IcePIC system. The microscope stage and air around the device were at or near ambient temperature. The IcePIC photographs and accompanying notes from the microscopic examination were retrieved from the ICECAPS database (<https://downloads.psl.noaa.gov/psd3/arctic/summit/icepic/>) on 31 July 2023. The photographs may be used to qualitatively characterize riming on ice crystals during precipitation events, but may not be representative of particle habits in

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The Multi-Angle Snowflake Camera (MASC) captures photographs of snow particles in free fall from multiple angles while simultaneously measuring their fall velocity (Garret and Yuter, 2014).

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At Summit, the MASC has operated since 2014 as a part of the ICECAPS observatory (Shupe et al., 2013). Measurements of ... [6]

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65 columns, bullets, stellar plates or dendrites. Lower summer d-excess (~+14‰) is associated with snow particles showing
66 heavier riming. The original crystal shapes of stellar plates and dendrites are preserved but riming growth covers almost the
67 entire particle surface.

68
69 Lowest d-excess of -2.5 to +3.8‰ corresponds to winter snowfall (DJF) with relatively small particles of ~20 to ~700 µm.

70 As the IcePic photographs show, snow particles collected on a slide kept in open air, blowing snow may have been captured
71 and all of the small particles may not have fallen directly as precipitation.

72
73 3.1.2 MASC fall velocity

74 The MASC at Summit operated periodically during 2014 and measurements of fall velocity for several days in June, July
75 and August 2014 were concurrent with isotope samples. Photographs of snow particles on which the fall velocity was
76 measured provide visual evidence of the degree of riming corresponding to measured particle fall velocity.

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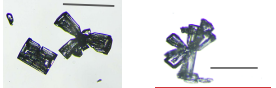
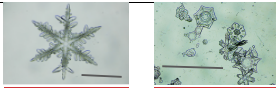
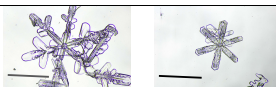
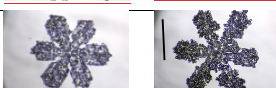
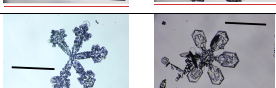
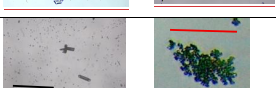
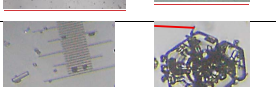
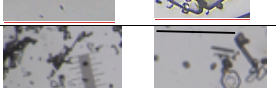
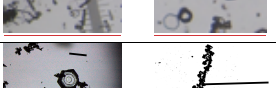
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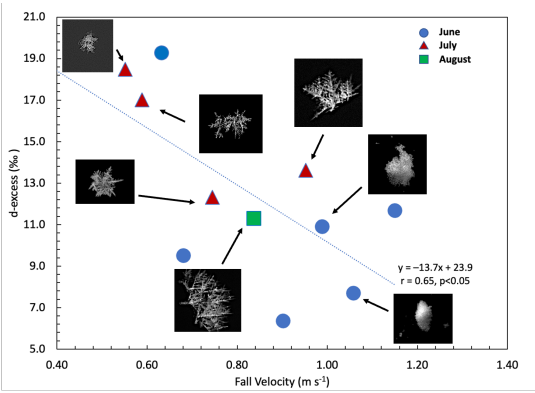
82 Table 3. Photographs of snowflakes during precipitation events at Summit, Greenland captured with the IcePIC system. The
83 scale bar in each photograph is ~500 μ m (black) or ~100 μ m (red). Photographs may not be representative of daily
84 precipitation for which the d-excess values are listed here.

		<u>Date/Collector Notes</u>	<u>d-excess (‰)</u>
<u>A</u>		<u>2012-07-03</u> <u>Small columns, bullets and plates (not visibly rimed).</u>	<u>+22.4</u>
<u>B</u>		<u>2012-07-24</u> <u>Stellar crystals, small plates, dendrites (not visibly rimed).</u>	<u>+22.7</u>
<u>C</u>		<u>2012-06-25</u> <u>Mostly aggregates. Stellar crystals, sector plates (not visibly rimed).</u>	<u>+21.4</u>
<u>D</u>		<u>2012-08-06</u> <u>Moderately to heavily rimed stellar plates and dendrites.</u>	<u>+13.8</u>
<u>E</u>		<u>2012-05-27</u> <u>Stellar dendrites and plates. Much riming, some very heavy. Unrimed particles (right) observed in the afternoon.</u>	<u>+13.5</u>
<u>F</u>		<u>2014-01-06</u> <u>Very small particles, columns, bullets and rosettes. (possible blowing snow contamination)</u>	<u>+3.8</u>
<u>G</u>		<u>2014-02-05</u> <u>Small needles, prisms, rime clumps, few plates with rime.</u> <u>(scale graduations: 0.01mm)</u>	<u>+3.0</u>
<u>H</u>		<u>2014-01-29</u> <u>Small columns, needles, prisms, large amount of rime pellets. (possible blowing snow contamination)</u> <u>(scale graduations: 0.01mm)</u>	<u>-2.0</u>
<u>I</u>		<u>2014-02-28</u> <u>Rimed crystals and smaller irregular crystals.</u> <u>Rime clump on right is magnified from a portion of the image on left.</u>	<u>-2.5</u>

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87 The MASC-measured average fall velocity of snow particles and the d-excess of corresponding daily samples of summer
88 precipitation (JJA) are shown in Fig. 2. Representative photographs of snow particles, on which the fall velocity was
89 measured, are also shown. Note that precipitation samples for d-excess were collected over a ~24 hr period while the MASC
90 fall velocities were measured for shorter durations so that the MASC measurements may not be representative of daily
91 precipitation. The average fall velocity was calculated from 3 to 93 measurements on individual particles (Table S2), except
92 for one day that had a single measurement.

94 Although the MASC data are for the summer months (June – August), they correspond with a significant range of d-excess
95 from +6.4 to +19.3%. A strong, inverse correlation is seen between the d-excess of daily precipitation and the average fall
96 velocity of snow particles ($r = 0.65$; $p < 0.05$). The particle shapes and riming in MASC photographs are consistent with those
97 from the IcePIC photographs (Table 3). MASC photographs from two events in July (red triangles with d-excess of ~18‰;
98 Fig. 2) show snow particles that are lightly rimed and the ice crystal shape is clearly evident. Following Locatelli and Hobbs
99 (1974), these particles would be classified as lightly rimed assemblages of dendrites. The higher fall velocities (~1 m s⁻¹) and
00 lower d-excess (~+10‰) correspond to June events (blue circles) where heavily rimed particles have a more rounded shape
01 with the underlying ice crystal outline no longer apparent. These particles would be classified as lump graupel or graupel-
02 like snow (Locatelli and Hobbs, 1974). For intermediate d-excess and fall velocities in July and August events, the particles
03 are similar to lightly rimed aggregates of dendrites (Locatelli and Hobbs, 1974).



09 Figure 2. d-excess versus average fall velocity measured with a MASC at Summit. Selected photographs of particles for
10 which the fall velocity was measured are also shown. The dotted line shows the best fit linear regression line.

Moved up [15]: snow particles collected on a slide kept in open air, blowing snow may have been captured and all of the small particles may not have fallen directly as precipitation.

3.1.2 MASC fall velocity

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Moved up [16]: Table 3. Photographs of snowflakes during precipitation events at Summit, Greenland captured with the IcePIC system. The scale bar in each photograph is ~500 μm (black) or ~100 μm (red).

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3.1.3 Doppler velocity

Fig. 3 shows a scatter plot of d-excess versus MDV in daily precipitation at Summit. The data are categorized as summer (JJA) and winter (DJF). For clarity, Summit data for the other seasons are not shown as they lie within the bounds of the summer and winter categories (Fig. S4).

The variation of d-excess with MDV for the summer precipitation (brown circles in Fig. 3) shows essentially the same inverse relationship as that indicated by the MASC fall velocities (Fig. 2) also obtained in the summer. Samples with higher d-excess ($>+20\%$) have a range of MDV values that are mostly less than $\sim 1.0 \text{ m s}^{-1}$, indicating unrimed to moderately rimed particles (Locatelli and Hobbs, 1974). Lower d-excess of ~ 12 to 14% is associated with higher MDV ($>1 \text{ m s}^{-1}$) that is indicative of heavier riming (Kneifel and Moisseev, 2020; Matrosov, 2023). Table 3 shows a moderate to heavily rimed particle collected on 2012-08-06, consistent with a relatively lower d-excess of daily precipitation ($+13.8\%$) on that day.

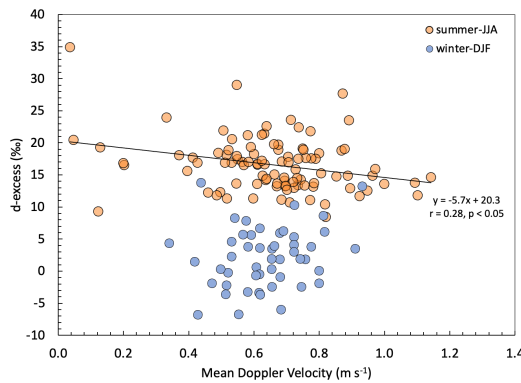


Figure 3. d-excess and MDV in daily precipitation at Summit. The best fit line for linear regression of summer data is shown (solid black).

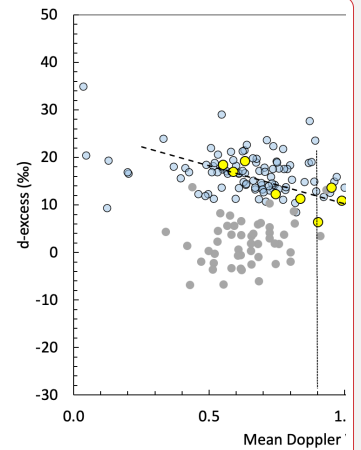
Winter precipitation at Summit (blue circles in Fig. 3) mostly has lower d-excess than the summer but with comparable MDVs, albeit with a slightly narrower MDV range than in the summer. Notably, the maximum MDV value in the winter is $<1 \text{ m s}^{-1}$ and an inverse correlation with d-excess is absent. The winter MDV values likely reflect a smaller particle size at colder temperatures (Takahashi and Fukuta,

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1988; Heymsfield et al., 2013) while the lower d-excess may result from an alternative mechanism than riming as discussed below.

Summit precipitation is attributed to deep (~4–6 km above ground level [AGL]) or shallow (<3 km AGL) clouds with varying proportions in different months (Shupe et al., 2013; Miller et al., 2015; Pettersen et al., 2018). Pettersen et al. (2018) used differences in the microwave absorption and scattering properties of cloud liquid water and ice to separate the majority of the precipitation events (both by accumulation and occurrence frequency) into three categories: snow originating from fully glaciated ice clouds (IC), snow where cloud liquid water was measurable in the column (CLW), and where the IC or CLW cloud types could not be differentiated based on their microwave signals (indeterminate). The CLW type clouds were single- or multi-layer, Arctic mixed-phase clouds while the IC type were similar to deep, nimbostratus-like clouds. Snowfall mass accumulating at Summit from IC and CLW events was seasonally variable with CLW events more prevalent in the summer and the IC events in winter (Pettersen et al., 2018). Back-trajectory analysis indicated that CLW clouds frequently originated from the west-southwest of Greenland and IC clouds primarily from the southeast, but with no seasonal differences in air mass trajectory for each cloud type, suggesting that moisture source origin alone cannot explain the seasonal d-excess variability. A further study (Kopeck et al., 2019) suggested that summer precipitation originated from vapor sublimated from surface snow, resulting in higher d-excess; however, this is inconsistent with the back-trajectory analysis of Pettersen et al. (2018). The lower d-excess in winter at Summit compared to summer is therefore unlikely to reflect seasonal differences in the origin of source moisture. Alternatively, riming may be important only in the summer while ice growth in the colder and drier winter conditions may occur mostly by vapor deposition, but with lower d-excess.

3.1.4 Radiosonde profiles at Summit

To investigate this further, profiles of temperature (T), relative humidity (RH), and ice supersaturation (SI) derived from radiosonde soundings at Summit were evaluated for the winter (blue curves in Fig. 4) and summer (brown curves in Fig. 4). Mean temperatures at the surface (Fig. 4A) are 18°C colder in winter (−30.9°C) than in summer (−12.5°C). With increasing height, the seasonal difference in mean temperature remains nearly the same throughout the column. The RH at the surface is lower in the winter (~73%) than in the summer (~85%) and decreases with height such that the RH is nearly the same (~50%) at ~3500 m AGL in both seasons (Fig. 4B). At higher altitudes, the RH is slightly higher in the winter than in the summer. Most of the precipitating column in the winter is beyond the mixed-phase regime ($T < -38^{\circ}\text{C}$ above ~2400 m AGL) where ice growth would occur by vapor deposition as diamond dust. In the summer, $T < -38^{\circ}\text{C}$ is above ~4600 m AGL, indicating that riming can occur at lower levels if RH approaches liquid saturation.

RH in the summer approaches liquid water saturation at some levels near the column base at temperatures between −4 and −15°C (Fig. 4A–B), where near-zero SI (Fig. 4C) indicates conditions favorable for riming growth. Higher in the column, SI is positive and RH is below liquid saturation, favoring ice growth by vapor deposition. About 20% of summer soundings

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Deleted: (Kneifel and Moiseev, 2020; Matrosov, 2023). We can assess the particle size effect on MDV from differences in average Z_e for the summer (−5.7 dBZ) and winter (−14.3 dBZ). The Z_e and MDV values in our study are described by a power law with an exponent of 0.09, which is within the range of exponent values found for Arctic

Deleted: (Matrosov, 2023). These results suggest that, for similar levels of riming, the winter MDV values may be smaller by ~20% on average compared to the summer.

Alternatively, riming growth in the winter may occur on a nucleus consisting of a frozen drop, as opposed to a larger particle that formed by vapor deposition, resulting in a lower d-excess of the rimed ice. Experimental studies indicate that riming at low temperatures (−22 °C) can be initiated on particles as small as 30 to 60 μm (Avila et al., 2009). Clumps of frozen drops may be present in winter precipitation at Summit (e.g., Row 1, Table 3), assuming that the image represents falling snow particles and not blowing snow. A proportionally smaller fraction of ice growth by vapor deposition in the winter compared to the summer may therefore result in much lower winter d-excess values. Additionally, Souchez and Jouzel (1984) found that ice formed by partial freezing had a lower d-excess as the initial $\delta^{18}\text{O}$ and $\delta^2\text{H}$ of the water decreased. Winter precipitation in general has lower $\delta^{18}\text{O}$ and $\delta^2\text{H}$ and the differences are starker in the polar regions. At Summit, the average winter ^{18}O and $\delta^2\text{H}$ are, respectively, −40‰ and −318‰, compared to −29‰ and −216‰ in the summer (Kopeck et al., 2019). These large differences in seasonal isotopic compositions may result in a lower d-excess of rimed ice in the winter compared to the summer. We note that these explanations are somewhat speculative, although they are consistent with the generally lower particle size, reflectivity and MDV in the winter. A more detailed discussion of riming growth in the winter precipitation at Summit and its effect on d-excess is beyond the scope of this work.

Precipitation at Summit

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Deleted: Analysis of 36-hr back trajectories indicated that the CLW clouds frequently originated from the west-southwest of Greenland and the IC clouds primarily came from the southeast, but there were no seasonal differences in air mass trajectory for each of the cloud types.

Deleted: the winter (Pettersen et al., 2018). However, the indeterminate cloud type (that includes IC and CLW) had the same or higher accumulation in the winter as the IC type.

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show near-liquid-saturation conditions at the column base, consistent with the mixed-phase CLW cloud type identified by Pettersen et al. (2018) as more prevalent in summer, and with the higher MDV values (~1 to 1.2 m s⁻¹; Fig. 3) indicating riming contribution to some lower d-excess summer events. The contrasting roles of vapor deposition and riming in controlling summer d-excess are illustrated by the two case studies discussed below.

In the winter, RH never approaches liquid water saturation at any level throughout the precipitating column (Fig. 4B). At the low temperatures characteristic of the Summit winter column (−30 to −65°C), liquid water cannot be sustained even when ice supersaturation is high (Korolev and Mazin, 2003; Murphy and Koop, 2005). Ice-supersaturated conditions (SI > 0) occur in 60–68% of winter soundings near the surface, declining to ~25% at 4000–6000 m AGL (Fig. 4D), confirming that vapor deposition prevails throughout on most winter isotope sampling days. When SI > 0, the mean SI is ~10% throughout the column in both seasons (not shown). Despite the ice-supersaturated conditions, the lower d-excess of winter precipitation (−6.8 to +13.8‰) is counterintuitive because vapor deposition at higher SI is generally thought to produce higher d-excess at a given temperature (Jouzel and Merlivat, 1984). However, vapor deposition at very low temperatures from low δ¹⁸O vapor can produce condensate with low or even negative d-excess (Dütsch et al., 2019; their Fig. 1). The roles of SI, temperature and vapor δ¹⁸O in reconciling the low winter d-excess at Summit are explored further in Sect. 3.5.

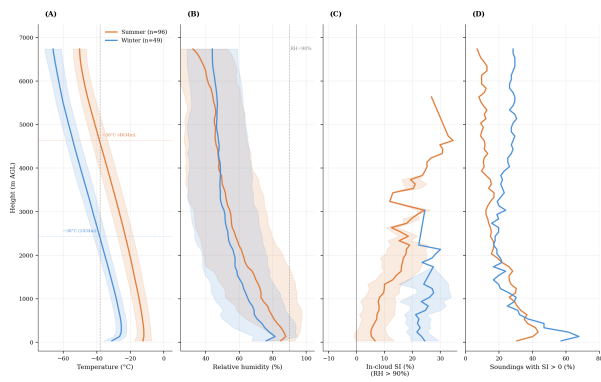


Figure 4. Mean vertical profiles of air temperature (A), relative humidity (B), and in-cloud ice supersaturation (C) from radiosonde measurements at Summit during summer (brown) and winter (blue). Panel D shows percentage of soundings when SI>0 for at least one radiosonde level.

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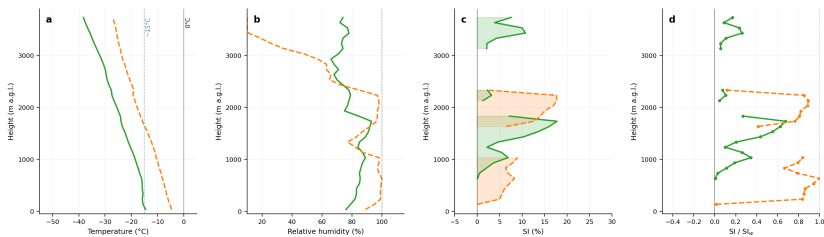
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01 To illustrate the influence of vapor deposition and riming growth on d-excess variability in summer precipitation, we
 02 examine radiosonde profiles from two contrasting events on 2 July 2012 (d-excess = +23.6‰; MDV = 0.89 m s⁻¹) and 26
 03 July 2013 (d-excess = +14.7‰; MDV = 1.14 m s⁻¹). We use the ratio SI/SI_w (Sect. 2.4) as a diagnostic of proximity to liquid
 04 water saturation where wet- or dry-growth riming would be favored depending upon the ambient temperature.

05 The precipitation column on 2 July 2012 (green curves in Fig. 5) was cold and deep, with temperatures ranging from -16°C
 06 near the surface to -30°C at ~3000 m AGL (Fig. 5a). Relative humidity exceeded 80% through a deep cloud layer from near
 07 the surface to ~2500 m AGL, within which SI rose from near zero at cloud base to ~18% (Figs. 5b-c). Throughout this layer,
 08 SI/SI_w remained well below 1.0 (Fig. 5d), indicating that the air was substantially undersaturated with respect to liquid water
 09 despite the high ice supersaturation. Under these conditions, which are characteristic of cold ice clouds well below the
 10 mixed-phase temperature range, ice growth would occur entirely by vapor deposition at variable ice supersaturation,
 11 consistent with the high d-excess of +23.6‰.

12 A contrasting two-layer cloud structure existed on 26 July 2013 (orange curves in Fig. 5). In the lower layer (surface to
 13 ~1000 m AGL, T = -5 to -11°C), RH approached liquid water saturation and SI was near zero or slightly negative (Figs. 5a-
 14 c), with SI/SI_w approaching 1.0 (Fig. 5d). These conditions are favorable for riming growth, with wet-growth more likely
 15 given the warmer temperatures and near-liquid-saturation environment. An upper cloud layer was present from ~1600 to
 16 2300 m AGL (T = -15 to -19°C) where SI was positive and SI/SI_w was lower, indicating ice supersaturation but less
 17 proximity to liquid saturation. At these lower temperatures, any riming would occur by the dry-growth process where
 18 accreted liquid freezes on contact rather than sustaining a liquid film, producing only a modest d-excess reduction (~3‰;
 19 Sect. 3.5). The combination of near-liquid-saturation conditions in the lower layer, vapor deposition in the upper layer, and
 20 the transition between the two growth regimes is consistent with the intermediate d-excess of +14.7‰, which is lower than
 21 that during the depositional 2 July event.

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26 Figure 5. Height-resolved profiles of T, RH, SI and SI/SI_w during Summit precipitation events on 2 Jul 2012 (green curves)
27 and 26 Jul 2013 (orange curves).

30 3.2 Riming and d-excess at other study locations

31 We now discuss the correlation of d-excess and MDV at the remaining polar and non-polar locations in this study, as well as
32 cloud profiles derived from radiosonde soundings at Ny-Ålesund and Dumont d'Urville in order to characterize the effect
33 of cloud processes on d-excess.

35 3.2.1 MDV and d-excess correlation

37 The d-excess and MDV at Ny-Ålesund, Andenes, Dumont d'Urville, Cazadero and Rio Claro (Fig. 6) are inversely
38 correlated. This correlation was quantified using total least squares (TLS) regression across all locations (n=211), which
39 accounts for uncertainties in both variables. The regression yielded a slope of -59.7 and intercept of 83.4 with $r = -0.67$
40 ($p < 0.001$). The slope is negative at each individual site (range: -45 to -78), but the correlation is weaker at Dumont
41 d'Urville (slope = -45, $r = -0.43$). The two best-sampled sites (Cazadero, n=57; Ny-Ålesund, n=112) show tightly
42 constrained, statistically robust slopes (-62.4 and -63.5, respectively, both $p < 0.001$). This consistency across sites,
43 spanning mid-latitude, polar, and tropical climates, supports a common physical mechanism — riming-driven lowering of d-
44 excess — rather than a coincidental cross-site correlation.

46 Ny-Ålesund precipitation has a wide range of d-excess from -8.2 to +45.2‰ (green circles in Fig. 6). Rain has lower values
47 (-8.2 to +14.9‰; median = +3.1‰) compared to snow (+3.9 to 45.2‰; median = +19.2‰). Correspondingly, the MDV is
48 higher on rain days (0.8 to 1.9 m s⁻¹; median = 1.2 m s⁻¹) compared to snow days (0.6 to 1.7 m s⁻¹; median = 1.0 m s⁻¹).
49 Andenes precipitation (dark grey circles in Fig. 6), consisting mostly of snow samples, has a d-excess range of -1 to +33.4‰
50 (median = +28.2‰) with MDV ranging from 0.4 to 1.1 m s⁻¹ (median = 0.7 m s⁻¹).

52 Precipitation at Ny-Ålesund and Andenes is produced in low-level mixed-phase clouds within cyclonic, atmospheric river
53 and frontal systems (Lauer et al., 2023; Chellini and Kneifel, 2024; Ebell et al., 2024). Heavily rimed particles with higher
54 MDVs are commonly observed at Ny-Ålesund (Chellini and Kneifel, 2024; Mahrendl et al., 2024). Riming is attributed to
55 increased turbulence, indicated by a higher eddy dissipation rate (EDR), which also is higher at cloud top temperatures
56 warmer than about -10°C (Chellini and Kneifel, 2024), consistent with the generally higher MDV and lower d-excess of rain
57 compared to snow in our study.

Deleted: We used the results of Pettersen et al.'s classification, which was conducted at one minute time resolution, to calculate the mode of the cloud type at 12-hr intervals in daily precipitation. The dominant precipitating cloud type for low d-excess (-6.9 to 13.8‰) winter precipitation in this study is the indeterminate type (consisting of IC or CLW clouds) followed by the CLW type (Table S2). An examination of selected cloud radar images shows that supercooled liquid generally was present on days when d-excess was low and the indeterminate cloud type was dominant. This indicates that the indeterminate clouds for the winter days in our study likely were of CLW type, which would allow for the riming growth of ice.

3.2 MDV

Deleted: polar and non-polar

Deleted: Andenes (Norway),

Deleted: (Antarctica),

Deleted: (California),

Deleted: Brazil). Data for these locations are shown on Fig. 4 where we have also shown the Summit data (from Fig. 3) for reference. An inverse correlation of d-excess and MDV can be seen in Fig. 4. A linear

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Deleted: locations. The correlation for individual locations also is fairly strong ($R^2 = 0.43$ to 0.78) except for Dumont d'Urville ($R^2 = 0.18$)....

Deleted: samples (green circles in Fig. 4) consist both of snow and rain with

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Deleted: and above the melting layer for rain, ranges from 0.6 to 1.8 m s⁻¹. The d-excess (-1 to 20.3‰) and MDV

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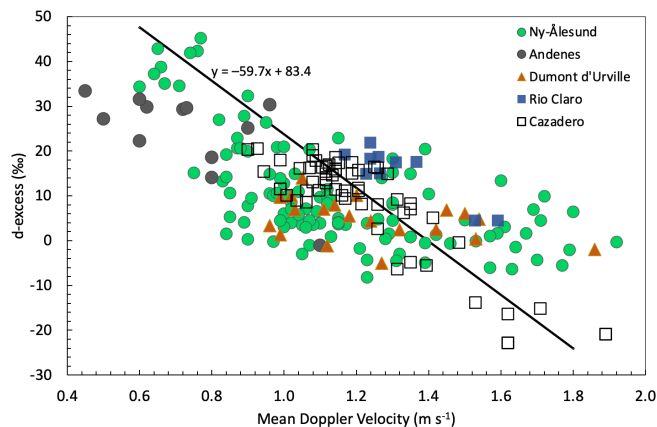


Figure 6. d-excess versus MDV in polar and non-polar precipitation at all study locations except Summit. The total least squares best-fit line is shown ($r = -0.67$; $p < 0.001$).

Daily average MDV at Dumont d'Urville ranges from ~ 1 to 1.9 m s^{-1} , indicating significant riming growth, with the corresponding d-excess of -5 to $+14\%$ (brown triangles in Fig. 6). Antarctic precipitation at this coastal location is produced mostly in warm fronts of extratropical cyclones (Jullien et al., 2020) with frequent riming and graupel formation (Grazioli et al., 2017). A strong katabatic layer at ~ 200 to 500 m AGL results in extremely dry conditions near the surface such that over one-third of the time when it is precipitating, snowfall is fully sublimated (virga) before reaching the ground surface (Jullien et al., 2020). For several samples in our limited dataset of 20 samples, MDV values of 1.5 to 1.9 m s^{-1} are higher than the maximum monthly average ($\sim 1.5 \text{ m s}^{-1}$) in a much larger, seven-year dataset of MDVs with 5500 measurements per month (Wiener et al., 2024). The higher MDV of our samples may be an artefact of stronger turbulence in low-level katabatic winds so that the upward and downward air motions did not completely cancel out for all samples in our dataset (Sect. 2.3.3).

At Cazadero and Rio Claro, precipitation was sampled only as rainfall. Average MDVs were obtained over ~ 30 min intervals during stratiform events characterized by the presence of a reflectivity bright band. The top of the melting layer was at a height of $\sim 2 \text{ km}$ at Cazadero and $\sim 4 \text{ km}$ at Rio Claro. In the snow region above the melting layer, the average MDV and d-excess values, respectively, ranged from 0.9 to 1.9 m s^{-1} and -22.9 to $+20.6\%$ at Cazadero (open squares in Fig. 6) and from 1.2 to 1.6 m s^{-1} and $+4.5$ to $+21.9\%$ at Rio Claro (blue squares in Fig. 6).

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Deleted: -2 to $+14\%$ are consistent with the common occurrence of graupel and other rimed particles in precipitation from synoptic, extra-tropical

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45 Stratiform precipitation at Cazadero is produced mostly in landfalling, extra-tropical cyclones and atmospheric rivers (White
46 et al., 2003). Precipitation d-excess is nearly the same in both regimes (Coplen et al., 2015). Orographically forced, relatively
47 deeper clouds at Cazadero seed the ice particles that fall through the lower, feeder clouds where supercooled water droplets
48 are accreted and ice grows by riming (White et al., 2003). At Rio Claro, precipitation is associated with mesoscale
49 convective systems (Santos et al., 2024) where the seeder–feeder process is commonly observed during stratiform events
50 (Houze, 2014).

51

52

53 **3.2.2 Radiosonde profiles**

54 **3.2.2.1 Nv-Ålesund**

55 Height-resolved profiles of T, RH and in-cloud SI (RH > 90%) on rain days (brown curves) and snow days (blue curves) are
56 shown in Fig. 7. Mean temperature decreases from ~5°C near the surface to about -15°C at 4000 m on rain days (Fig. 7A),
57 and from -5°C near the surface to about -25°C at 4000 m on snow days. Mean RH profiles are nearly the same on rain and
58 snow days (Fig. 7B), increasing from near-surface values of ~80% to near or above 90% between ~800 and 1500 m, and
59 then decreasing to ~65–70% at 4000 m.

60

61 The 0°C isotherm on rain days ranged widely from near the surface to ~3150 m AGL, with a mean of 914 ± 688 m and
62 median of 700 m, reflecting the large variability in synoptic conditions across the 2014–2018 sampling period. The majority
63 of rain events (59%) had freezing levels between 500 and 1500 m, with 25% below 500 m and 11% above 2000 m. This
64 variability in freezing level height directly determines the depth of the ice-phase column available for riming and vapor
65 deposition growth, contributing to the wide range of d-excess values observed in surface rainfall.

66

67 The fraction of radiosonde levels within clouds (RH > 90%) increased from ~10% near the surface to a maximum of 74% at
68 800–1050 m (Fig. 7B). Within this cloud layer, RH approached liquid water saturation (mean RH = 91–97%), indicating
69 mixed-phase conditions where riming would be the dominant growth process at warmer temperatures near the base of the ice
70 column.

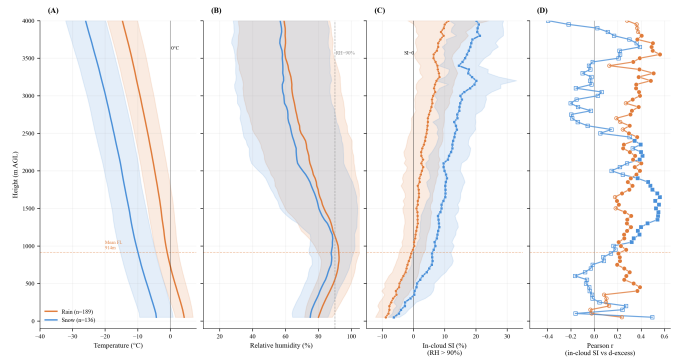
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72 Mean in-cloud SI on rain days is negative (-8%) near the surface and becomes positive above ~1000 m (near the mean 0°C
73 level), increasing to ~10% at 4000 m (Fig. 7C). These vertical transitions in T and RH define a two-layer growth structure
74 above the 0°C level on rain days: a lower mixed-phase riming layer from ~1000 – 2750 m AGL where SI is weakly positive
75 (0 to 5%) but the cloud is near water saturation, and an upper depositional layer (~2750–4000 m) where SI increases from 5
76 to 10% as temperatures decrease from -8 to -13°C. Ice crystals nucleate and grow by deposition in the upper layer before
77 falling into the riming zone below, so the isotopic composition of surface precipitation integrates contributions from both
78 growth regimes. This is consistent with the mixed-phase cloud structure described by Korolev et al. (2017).

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82 Figure 7. Mean vertical profiles of (A) air temperature, (B) relative humidity, (C) in-cloud SI, and (D) Pearson correlation
83 coefficient (r) for the linear regression of d-excess and height-resolved mean in-cloud SI from radiosonde measurements at
84 Ny-Ålesund during rain (red) or snow (blue) precipitation days. Open symbols in D indicate $p > 0.05$ (statistically
85 insignificant) and solid symbols indicate $p < 0.05$ (statistically significant).
86

87 Snow days show positive SI throughout the column, increasing from near zero at ~300 m to ~20% at 4000 m (Fig. 7C),
88 reflecting consistently colder temperatures at all levels and the absence of liquid saturation conditions. Under these
89 conditions vapor deposition is the dominant growth process, with riming occurring episodically where turbulent vertical
90 motions activate supercooled liquid water (Korolev et al., 2017; Chellini and Kneifel, 2024).

91 A significant positive correlation exists between d-excess and height-resolved in-cloud SI across most of the ice-phase
92 column on rain days (brown curve in Fig. 7D), with r values ranging from 0.18 to 0.50 ($p < 0.05$) between 400 and ~3500 m
93 and showing considerable variability with height. The correlation extending below the mean 0°C isotherm to ~400 m likely
94 reflects variability in freezing level height across rain events, such that on days with lower freezing levels the ice-phase
95 column extends to those lower altitudes and the SI–d-excess relationship is maintained. Precipitation events with higher SI in
96 the upper depositional layer are associated with relatively higher d-excess in surface rainfall, consistent with the seeder-
97 feeder growth model in which stronger vapor deposition aloft partially offsets the d-excess reduction from riming below.

98 On snow days, the SI – d-excess correlation (blue curve in Fig. 7D) is significant between 1050 to ~2000 m ($r=0.32$ to 0.56 ,
99 $p<0.001$), consistent with vapor deposition being the dominant growth process in that layer and indicating that it represents
00 the primary zone of isotopically significant crystal growth on snow days. The radiosonde profiles therefore characterize the
01 vapor deposition component of snow day growth, while the MDV independently captures the riming component — the two

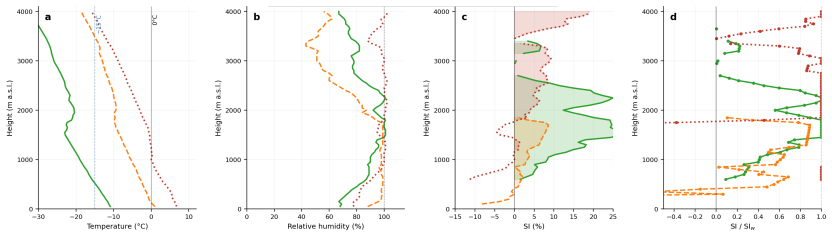
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02 measurements being complementary rather than redundant. In contrast to rain days, where the melting layer
03 thermodynamically anchors the riming zone as a persistent day-scale feature, transient mixed-phase conditions on snow days
04 may not be captured in a once-daily radiosonde profile but are evident in higher MDV values (up to 1.7 m s^{-1}) that directly
05 reflect riming intensity regardless of when and where in the column it occurred.

07 Three radiosonde profiles for 12 February 2016 (d-excess = +34.5‰; MDV = 0.71 m s^{-1}), 31 August 2014 (d-excess =
08 +13.9‰; MDV = 0.99 m s^{-1}) and 19 July 2014 (d-excess = -4.6‰; MDV = 1.31 m s^{-1}) illustrate the effect of ice growth
09 regimes on d-excess variability at Ny-Ålesund.

11 On 12 February 2016 (green curves in Fig. 8), surface temperature was -10.5°C , decreasing to -20°C at 1000 m AGL and to
12 still lower values at higher levels (Fig. 8a). A deep cloud layer (RH>90%, SI > 0) extended from ~600 to 3400 m AGL (Fig.
13 8b), within which three distinct zones of ice crystal growth are apparent. Vapor deposition would be favored at cloud top
14 (~3000 to 3200 m AGL; $T \sim -24$ to -26°C) and at cloud base (600 to 1250 m AGL), where the cloud was mildly
15 supersaturated with ice (SI = ~0 to 8‰; Fig. 8c) but substantially undersaturated with respect to liquid water (SI/SI_w < 0.5;
16 Fig. 8d). A middle cloud layer between ~1250 and 2400 m AGL had higher SI values of 17–26‰ with SI/SI_w exceeding 0.6
17 and briefly approaching 1.0 at several levels, where riming would be favored. However, at the colder temperatures in this
18 layer (-20 to -23°C), riming would occur by the dry-growth process with only a modest lowering of d-excess (Sect. 3.5). As
19 a result, precipitation d-excess would be determined predominantly by the vapor-deposition signal, consistent with the
20 observed high d-excess of +34.5‰.

21



22
23
24 **Figure 8** Height-resolved profiles of T, RH, SI and SI/SI_w during Ny-Ålesund precipitation events on 12 Feb 2016 (green
25 curves), 31 Aug 2014 (orange curves) and 19 Jul 2014 (red curves).

28 On 19 July 2014 (red curves in Fig. 8), surface temperature was $+6.7^\circ\text{C}$, with the freezing level at ~1000 m AGL (Fig. 8a).
29 An unusually deep isothermal melting layer extended from ~1000 to ~1400 m AGL where temperatures remained at or

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40 within a fraction of a degree of 0°C. Immediately above, from ~1400 to 1800 m AGL, SI was slightly negative despite high
41 RH of 95–99% (Fig. 8b), reflecting the near-zero temperatures in this transitional layer where air approaches but does not
42 reach ice saturation. The primary ice-forming cloud extended from ~1800 to 3300 m AGL, where RH was ~100% and SI
43 ranged from near zero to ~9% (Fig. 8c). At the relatively warm temperatures in this layer ($T = -2$ to -11°C), SI/SI_w reached
44 water saturation intermittently (Fig. 8d), indicating conditions favorable for wet-growth riming as the dominant growth
45 process. A brief drier intrusion at 3300–3500 m AGL, where RH dropped to 88–91% and SI briefly became negative,
46 separated the riming zone from an upper cloud layer at 3500–4000 m AGL ($T = -12$ to -16°C). In this upper layer, SI
47 increased from ~2 to ~19% with RH ~100% near the top, and SI/SI_w of ~0.74 on average indicates predominantly
48 depositional growth with near-saturated conditions developing at the uppermost levels. The strongly rimed character of this
49 event, with wet-growth conditions dominating most of the ice-phase column at relatively warm temperatures, is consistent
50 with the low d-excess of ~4.6% observed at the surface.

51
52 Surface temperature on 31 August 2014 was +1.0°C, with the 0°C isotherm at ~200 m AGL (orange curve in Fig. 8a). RH
53 was 88% near the surface and rose sharply to ~98% by ~200 m, and then remained consistently high (97–99%) through
54 ~1700 m AGL (Fig. 8b). Ice supersaturation was intermittently positive from ~300 m upward and became consistently
55 positive from ~900 m to ~1800 m AGL (Fig. 8c), with temperatures ranging from ~0.6°C near the lower cloud margin to
56 ~9.8°C near the cloud top. SI/SI_w increased steadily with height through the cloud, approaching but not reaching water
57 saturation (0.84 to 0.88) between ~1300 and 1700 m AGL (Fig. 8d), before declining sharply toward the cloud top. Vapor
58 deposition would be favored near cloud top, while the near-water-saturation state in the 1300–1700 m layer at temperatures
59 of ~7.3 to ~9.5°C indicate wet-growth riming. The intermediate d-excess of +13.9% — lower than the vapor deposition
60 event on 12 February 2016 but higher than the more strongly rimed 19 July 2014 rainfall — is consistent with a combination
61 of vapor deposition near cloud top and wet-growth riming in the lower cloud layer, where riming remained intermittent
62 rather than fully established at water saturation.

63
64 **3.2.2.2 Dumont d’Urville**
65 Height-resolved profiles of T, RH and in-cloud SI (RH>90%) at Dumont d’Urville on isotope sampling days in the summer
66 (DJF; brown curves) and the winter (JJA; blue curves) are shown in Fig. 9. Although acceptable MDV values were available
67 for 20 of the 85 isotope sampling days, the radiosonde data are available for all days and this analysis is based on the full
68 dataset. Mean temperatures decrease from ~-2°C near the surface to -20°C at 4000m AGL in the summer (Fig. 9A) and
69 from -13°C to -26°C in the winter (Fig. 9A). Unlike Summit, temperatures throughout the 0–4000m column remain above
70 -38°C in both seasons, indicating that the entire precipitation-forming layer is within the mixed-phase regime where ice
71 growth can occur both by riming and vapor deposition.

72

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Mean RH near the surface is 65–70% in both seasons, decreases to about 55% between ~200 to 500m AGL, and shows a slight increase at higher levels before decreasing again above 2500m (Fig. 9B). In-cloud conditions ($RH > 90\%$) occur on 35–40% of isotope sampling days in summer and winter respectively (not shown), confirming that the $RH > 90\%$ criterion is appropriate for identifying cloud layers at this coastal Antarctic site. Mean in-cloud SI is ~8% in summer and ~6% in winter, with winter clouds shallower on average (~2400m vs ~3400m AGL cloud top in summer) and fewer in-cloud levels per day (4.5 in winter vs 8.6 in summer). Individual precipitation events frequently show a two-layer vertical structure — a lower cloud layer near water saturation where riming would be favored and an upper layer with $SI > 0$ but below water saturation where vapor deposition would dominate — as illustrated by the case studies below.

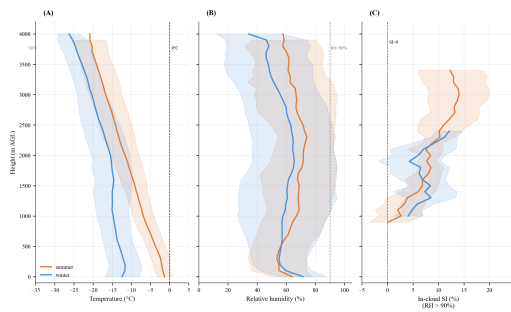


Figure 9. Mean vertical profiles of (A) air temperature, (B) relative humidity, and (C) in-cloud SI at Dumont d'Urville during summer (brown) or winter (blue) precipitation days.

The influence of vapor deposition and riming on d-excess can be illustrated with the radiosonde profiles for two events on 29 May 2019 (d-excess = +14.0; $MDV = 1.05 \text{ m s}^{-1}$) and 23 July 2019 (d-excess = +4.8; $MDV = 1.54 \text{ m s}^{-1}$).

On 29 May 2019, near surface temperature was approximately -16.4°C and decreased to about -33°C at 3300 m AGL (Fig. 10a). RH remained below 90% through most of the column, except for a narrow sub-layer (900–1300 m AGL, $T = -21$ to -22°C) with $RH > 90\%$. Ice-supersaturated conditions ($SI > 0$) extended from ~100 to 3300 m AGL, reaching up to ~15% at 3100 m AGL despite $RH < 90\%$ through most of this depth (Fig. 10b-c). The SI/SI_w ratio peaked at 0.64 within the higher-humidity sub-layer and was 0.4–0.5 through the deeper ice supersaturated layer above (Fig. 10d), placing the profile within the vapor deposition regime. This is consistent with the higher d-excess of +14.0‰ in surface precipitation.

The 23 July 2019 event corresponds to an intense synoptic scale intrusion of warm and moist air identified by Leroy-Dos Santos et al. (2023) as representative of the synoptic events that dominate precipitation variability at Dumont d'Urville.

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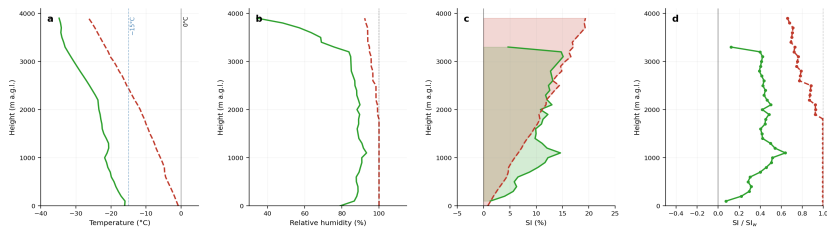


Figure 10. Height-resolved profiles of T, RH, SI and SI/SI_w during Dumont d'Urville precipitation events on 29 May 2019 (green curves) and 23 Jul 2019 (red curves).

3.3 Sub-cloud evaporation and d-excess

Sub-cloud evaporation of rain results in lower d-excess (Dansgaard, 1964) and could be responsible for the low d-excess observed at Cazadero, Rio Claro and Ny-Ålesund (Fig. 6). The sub-cloud layers at Cazadero (~1 km) and Rio Claro (~3 km) were sufficiently deep so that rainfall may be partially evaporated. Evaporation decreases the drop size across the particle size spectrum although smaller raindrops evaporate faster (Kumjian and Ryzhkov, 2010; Xie et al., 2016). The decreased particle size would result in a decreasing reflectivity profile in the sub-cloud region and could be used as an indicator of rainfall evaporation. 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 the end of each event to avoid the potential effect of sub-cloud evaporation on our analysis. This

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34 approach of partially discarding the early or late portions of an event was also used for Rio Claro. A decreasing reflectivity
35 profile was not observed for rain events at Ny-Ålesund where the sub-cloud layer generally is less than about 500 m deep.

36
37 However, decreasing reflectivity profiles in the sub-cloud region were present for several hours during one Cazadero
38 precipitation event on 1 March 2009 (Fig. S5). The time-height profiles of reflectivity and MDV for this event are shown in
39 Fig. S6. Precipitation began at about 03:00 UTC and continued through 24:00 UTC with several breaks in between. Lowest
40 d-excess values at Cazadero, ranging from -22.9 to -13.9‰ (Fig. 6), were measured in the first three hours (05:45 to 08:45
41 UTC) of this event when the rain rate mostly was $<1 \text{ mm hr}^{-1}$ (Coplen et al., 2015). Surface air temperature and RH averaged
42 11°C and 63%, respectively, between 05:00 and 09:00 UTC. Later in the day, the average temperature was slightly lower
43 (9°C) with high RH (91%), consistent with a lack of sub-cloud evaporation in reflectivity profiles. We examined the effect of
44 sub-cloud evaporation of rain on d-excess in our study by using calculations based on Stewart (1975) and by comparing the
45 fall velocities above and below the melting layer based on Weiss et al. (1977).

46 3.3.1 Calculation of d-excess change at study locations

47 We estimated the magnitude of d-excess by sub-cloud evaporation using the Stewart (1975) model, which
48 calculates isotopic fractionation during raindrop evaporation as a function of drop size, fall distance,
49 ambient temperature and RH. For the 1 March 2009 Cazadero event, with a sub-cloud region of about 1
50 km and particle size mostly greater than 0.5 mm recorded with a disdrometer (Fig. S7), our Stewart
51 model calculations using the observed surface conditions ($T = 11^\circ\text{C}$, $\text{RH} = 63\%$) show a d-excess
52 decrease of $\sim 2\%$.

53
54 Graf et al. (2019) applied a similar approach for characterizing below cloud changes in isotopic composition. They assumed
55 a cloud bottom at 1 km and a surface RH and temperature of 75% and 12°C , respectively, and calculated isotopic changes
56 resulting from evaporation and below cloud rain-vapor interaction. In this scenario, a small raindrop (0.5 mm) may lose
57 $\sim 28\%$ of its mass by sub-cloud evaporation, which would lower its d-excess by $\sim 10\%$. For a larger drop (1 mm), $\sim 7\%$ of the
58 mass may be lost with a 5‰ decrease in d-excess.

59
60 We therefore consider a maximum of 5‰ lowering of d-excess by sub-cloud evaporation at Cazadero on 1 March 2009.
61 Sub-cloud evaporation, therefore, would not be the primary factor responsible for the low d-excess (about -22 to -13‰) of
62 Cazadero rain event on 1 March 2009 and its inverse correlation with MDV. Adjusting for this change would increase the d-
63 excess, but would not affect the MDV values that were obtained for the snow region above the melting layer, strengthening
64 the inverse correlation of d-excess with MDV in Fig. 6.

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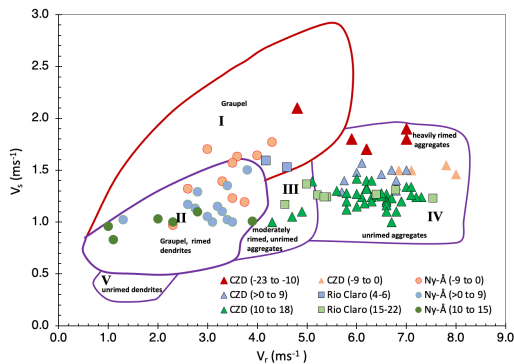
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93 Surface RH at Rio Claro exceeded 90% during the portions of the stratiform precipitation events used in this study
94 (Santos et al., 2024), indicating near-saturation conditions and consistent with the absence of decreasing
95 reflectivity profiles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

99 At Ny-Ålesund, the sub-cloud layer was typically less than ~500 m deep where rain drop evaporation potentially could
00 occur. However, the relatively colder temperatures and higher RH documented in the radiosonde profiles (mean near-surface
01 values of 5°C and 80%, respectively) further limit any evaporative loss in the shallow sub-cloud region.

03 **3.3.2 Fall velocity** above and below the melting layer
04 Independent confirmation that sub-cloud evaporation is not responsible for the low d-excess values comes from the
05 relationship between fall velocities in the snow region above the melting layer (V_s) and in the rain region immediately below
06 (V_r), following the classification of Weiss et al. (1977). The proposed $V_s - V_r$ categories for graupel, heavily rimed dendrites
07 and aggregates of dendrites, as well as lightly rimed or unrimed particles are shown in Fig. 11. The variability of fall
08 velocities within each category is quite large because of a range of particle size and density of the rimed and unrimed
09 particles. If sub-cloud evaporation were responsible for the lower d-excess of rain, we would expect lower d-excess samples
10 to lie in the unrimed or lightly rimed categories.



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24 **Figure 11.** Classification of rimed and unrimed particles based on fall velocities above (V_s) and below (V_r) the melting layer.
25 The boundaries for Categories I to V are as proposed by Weiss et al. (1977): **I.** Graupel (lump and conical,
26 hexagonal) and graupel-like snow; **II.** Graupel, graupel-like snow, moderately to densely rimed
27 dendrites, assemblages of dendrites, and columns, aggregates of unrimed side planes, bullets and
28 columns; **III.** Moderately rimed to unrimed aggregates of dendrites; unrimed aggregates of bullets, side
29 planes and columns; **IV.** Moderately rimed to unrimed aggregates and radiating assemblages of
30 dendrites; **V.** Unrimed dendrites. Graupel with varying density plots in multiple fields (highest density in category I). For
31 categories II, III and IV, unrimed particles plot towards the bottom and densely rimed towards the top. Also plotted are the
32 rainfall samples from Cazadero (CZD; triangles), Rio Claro (squares) and Ny-Ålesund (circles). Symbol colors correspond
33 to the range of d-excess values (red < -10‰, pink = -9 to 0‰; blue = >0 to +9‰; green > +10‰) as shown in the legend.
34

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The $V_s - V_r$ relationships in rainfall samples from Cazadero, Rio Claro and Ny-Ålesund also are shown on Fig. 11. Cazadero samples, for which the reflectivity profiles indicate potential sub-cloud evaporation, lie in the graupel or ‘heavily rimed’ aggregates fields (red triangles). This is consistent with their low d-excess (−22.9 to −13.9‰) and the effect of sub-cloud evaporation, if any, would not have been significant. The rest of the Cazadero samples lie in the heavily- or moderately-rimed aggregates fields, consistent with their increasingly higher d-excess (brown, blue and green triangles). Two of the Rio Claro samples with low d-excess of +4.5 to +5.6‰ lie in the graupel field (blue squares in Fig. 11). Other Rio Claro samples have much higher d-excess (green squares), consistent with their location in the light to moderately rimed aggregates fields. The d-excess of Ny-Ålesund samples is also consistent with their classification based on snow and rain region MDVs (Fig. 11). Lower d-excess samples (brown circles) lie in the graupel and heavily-rimed fields, consistent with a lack of sub-cloud evaporation as the bottom of the melting layer occurs within about 200 m above ground.

3.4 Rimed mass fraction

The MDV has been directly related to the rimed mass fraction (RMF), the ratio of particle mass acquired by riming to the total mass (Mosimann, 1995; Kneifel and Moisseev, 2020). This relationship was parameterized by using a large dataset of particle size and MDV in Hyytiälä, Finland precipitation (Kneifel and Moisseev, 2020): $RMF = -0.0528 MDV^4 + 0.2927 MDV^3 - 0.6125 MDV^2 + 1.0560 MDV - 0.4691$.

We used the Kneifel and Moisseev coefficients to calculate the RMF from MDV. Because RMF is derived from MDV via a non-linear equation, a separate total least squares regression with d-excess (excluding Summit) was conducted, yielding the following relationship: $d\text{-excess} = 46.5 - 124.2 \cdot RMF$ ($r = -0.67$). The regression equation indicates a low d-excess of about −78‰ for fully rimed particles (graupel at $RMF = 1$) and a high d-excess of +46.5‰ for unrimed particles growing by vapor deposition ($RMF = 0$).

Calculated RMF for Summit was excluded from the regression because riming at Summit may occur infrequently only in the summer (Sect. 3.1.3). Two Andenes samples result in negative RMF values. This arises because the generalized fall velocity–size relationship for unrimed particles in Kneifel and Moisseev (2020), derived from Hyytiälä precipitation, is not fully applicable to the unrimed particle population at Andenes, likely reflecting regional differences in particle size distributions. These two samples were also excluded from the regression.

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99 The inverse correlation of d-excess with RMF was essentially the same as that for MDV (Fig. S8), but RMF is used here
100 because it has a direct physical interpretation as the fraction of particle mass attributable to riming, allowing process-
101 weighted d-excess calculations to be directly linked to observable microphysical quantities (Sect. 3.5).
102

103 **3.5 In-cloud processes and precipitation d-excess**

104 The site-independent correlation of d-excess with MDV (Fig. 6) and the influence of vapor deposition versus riming growth
105 on d-excess of daily precipitation at Summit, Ny-Ålesund and Dumont d’Urville (Sect. 3.2) all indicate that variable in-cloud
106 processes modulate the d-excess of surface precipitation. The dynamic forcing controlling these processes changes rapidly
107 and frequently during a precipitation event (Korolev et al., 2017), such that precipitation collected for even a short interval of
108 10–20 minutes would contain hydrometeors that grew by vapor deposition at variable SI or by both vapor deposition and
109 riming. In addition, the lower d-excess of winter Summit precipitation without riming contributions indicates that vapor
110 deposition itself may result in variable d-excess. Isotopic analysis of precipitation therefore provides a process-weighted d-
111 excess value reflecting the integrated history of all growth modes experienced by the hydrometeor population. We now
112 explore the role of in-cloud processes in precipitation d-excess variability by calculating the d-excess of end-members
113 forming by vapor deposition (including diamond dust) and riming (wet and dry-growth) to evaluate a process-weighted
114 interpretation of observed d-excess at our study locations. A sensitivity analysis was conducted to evaluate the influence of
115 various input parameters on calculated d-excess. Finally, the end-member compositions were combined in different fractions
116 to show that the observed d-excess variations can be produced by in-cloud processes.
117
118

119 **3.5.1 Calculation of end-member d-excess**

120 The d-excess of vapor deposition and riming end-members in mixed-phase clouds were calculated following established
121 isotope fractionation frameworks (Jouzel and Merlivat, 1984; Jouzel et al., 1985). Calculations for diamond dust, which is
122 pure ice-phase precipitation forming at temperatures below −38°C, use the same kinetic fractionation as vapor deposition.
123

124 **3.5.1.1 Vapor deposition and diamond dust**

125 The isotope ratio during vapor deposition of ice, including diamond dust, is given as (Jouzel and Merlivat, 1984):
126

127
$$R_{ice} = \alpha_{sv} \cdot \alpha_k \cdot R_v \quad (1)$$

128

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where R_{ice} and R_v are the isotope ratios ($^{18}\text{O}/^{16}\text{O}$ or $^2\text{H}/\text{H}$), respectively, of vapor deposited ice and ambient vapor, α_{sv} is the equilibrium solid-vapor fractionation factor (Majoube, 1971 for ^{18}O ; Merlivat and Nief, 1967 for ^2H), and α_k is the kinetic fractionation factor (Jouzel and Merlivat, 1984):

$$\alpha_k = S_i / [\alpha_{sv} \cdot (D/D_{iso}) \cdot f \cdot (S_i - 1) + 1] \quad (2)$$

where S_i is the ice supersaturation ratio, D/D_{iso} is the ratio of diffusivities of the light to heavy isotopologue (1.0285 for H_2^{18}O ; 1.0251 for HDO ; Merlivat, 1978) reflecting the faster diffusion of the lighter molecule, and f is the ventilation factor. As noted previously, $S_i = 1 + SI/100$ and is parameterized as $S_i = a + b \cdot T$ with $a = 1$ and b generally in the range of -0.002 to -0.006 (Dütsch et al., 2019).

3.5.1.2 Wet- and dry-growth riming

Wet-growth occurs when the latent heat of fusion released by the freezing of the supercooled droplets raises the particle surface temperature to 0°C and the droplets spread to form a liquid film. This liquid film may partially evaporate before freezing is complete.

The isotopic composition of a liquid film on the growing particle evolves as follows (Jouzel et al., 1985):

$$dR_L/dt = [(R_c - R_L) \cdot (d\mu_c) + R_L \cdot (1 - \alpha_{ls}) \cdot d\mu_f + R_v \cdot d\mu_e - F] / \mu_L \quad (3)$$

where R_L is the isotope ratio of the liquid film, R_c is the cloud water isotope ratio, $d\mu_c$ is the total collection rate of supercooled liquid ($= d\mu_f + d\mu_e$), $d\mu_f$ is the freezing rate, $d\mu_e$ is the evaporation rate from the film surface, α_{ls} is the equilibrium liquid-solid fractionation factor, F is the isotope flux to the vapor phase, and μ_L is the film thickness. The freezing rate of the liquid is

$$d\mu_f = \pi \cdot r^2 \cdot V \cdot \text{LWC} \cdot E \quad (4)$$

where r is the particle radius, V is the relative fall speed of the particle with respect to air motion, LWC is the liquid water content, and E is the collection efficiency. The evaporation rate from the liquid film is governed by diffusive vapor transport away from the particle surface:

$$d\mu_e = 2\pi \cdot r \cdot D_{\text{H}_2\text{O}} \cdot \text{Sh} \cdot (\rho_s - \rho_e) \quad (5)$$

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58 where D_{H_2O} is the molecular diffusivity of water vapor ($= 2.4 \times 10^{-5} \text{ m}^2 \text{ s}^{-1}$; Pruppacher and Klett, 2010), Sh is the Sherwood
59 number ($Sh = 2 + 0.6 \cdot Sc^{1/3} \cdot Re^{1/2}$, computed from the same particle radius and fall speed as the freezing rate in Eq. 4), Sc is
60 the Schmidt number, Re is the Reynolds number, ρ_s is the saturation vapor density at the particle surface temperature, and ρ_e
61 is the ambient vapor density.

62 We note that evaporation of the liquid film on the particle surface is driven by the temperature difference between the
63 particle surface and ambient air. On the particle surface, the temperature (T_s) is 0°C regardless of the ambient air temperature
64 (T_a). Because T_a is less than T_s in any mixed-phase cloud, the saturation vapor pressure at the particle surface always exceeds
65 the ambient vapor pressure, driving continuous evaporation of the liquid film even when the surrounding air is at liquid
66 water saturation.

67 The isotope flux F in Eq. 3 is the species-specific form of Eq. 5:

68
$$F = 2\pi \cdot r \cdot D_i \cdot Sh_i \cdot (\rho_s \cdot R_L / \alpha_{lv} - \rho_e \cdot R_v) \quad (6)$$

69 where D_i is the absolute isotopologue-specific diffusivity ($D_i = D_{H_2O} \cdot D_{iso}/D$), and α_{lv} is the liquid-vapor equilibrium
70 fractionation factor at the particle surface temperature (0°C for wet-growth).

71 The isotope ratio of rimed ice from wet-growth at each time step is $\alpha_{ls} \cdot R_L$ and the integration of Eq. 3 proceeds until the
72 film composition reaches an asymptotic value at a prescribed film thickness (Jouzel et al., 1985).

73 Dry-growth occurs when the latent heat of fusion is dissipated without the particle surface warming to 0°C . In that case, a
74 persistent liquid film does not form and the accreted liquid freezes almost instantaneously. The isotope ratio of the rimed ice
75 is then assumed to reflect only the equilibrium liquid-solid fractionation:

76
$$R_{ice} = \alpha_{ls} \cdot R_c \quad (7)$$

77
78 This fractionation results in the d-excess of dry-growth rimed ice being $\sim 2\text{--}3\text{‰}$ lower than the cloud water depending on the
79 ambient temperature.

81 **3.5.2 Sensitivity calculations**

82 Equations 1 through 7 employ multiple parameters to determine the d-excess resulting from vapor deposition or riming
83 processes. Table S3 summarizes the various input parameters and their significance in the process-based isotope

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calculations. To assess the relative impact of these parameters on calculated d-excess, we conducted sensitivity calculations across a range of parameter values. Figure 12 illustrates the calculated sensitivity, focusing solely on one dominant parameter for each process. Detailed results for other parameters are provided in Figs. S9-S11.

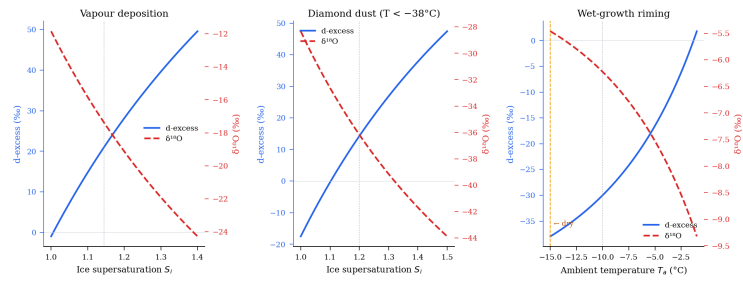


Figure 12. Dominant parameters and their effect on the calculated d-excess of end-members. Input parameters were: for vapor deposition ($\delta^{18}\text{O}_v = -30\text{‰}$, $d_v = 10\text{‰}$, $T_c = -20^\circ\text{C}$, $f=0.997$); for diamond dust ($\delta^{18}\text{O}_v = -50\text{‰}$, $d_v = 15\text{‰}$, $T_c = -40^\circ\text{C}$); for wet-growth riming ($\delta^{18}\text{O}_v = -20\text{‰}$, $d_v = 10\text{‰}$, $\text{LWC} = 0.5 \text{ g m}^{-3}$, $E = 0.8$, $R = 3 \text{ mm}$, $V = 1 \text{ m s}^{-1}$).

For vapor deposition, the sensitivity analysis identifies ice supersaturation as the dominant control on the d-excess of ice at temperatures greater than about -20°C (Fig. 12). The d-excess increases approximately linearly from near 0‰ at $S_i = 1.00$ to $+50\text{‰}$ at $S_i = 1.40$ — driven by the faster diffusion of HDO relative to H_2^{18}O . At lower temperatures, the low $\delta^{18}\text{O}$ of ambient vapor is an equally important secondary control: d-excess of deposited ice increases by about 47‰ as $\delta^{18}\text{O}_{\text{vapor}}$ varies from -60 to -20‰ , because $\alpha_{sv}(\text{H}_2\text{O})$ has a stronger temperature dependence than $\alpha_{sv}(\text{D})$ at $T < -20^\circ\text{C}$, depressing the d-excess of ice and potentially driving it negative. The ambient vapor d-excess has a comparatively minor impact as a given change in vapor d-excess produces an almost equally sized change in ice d-excess, rather than being amplified or damped by the fractionation in the deposition process.

Winter precipitation at Summit forms by vapor deposition throughout a cold column (~ -60 to -30°C) with positive SI (up to $\sim 23\%$), and atmospheric vapor with very low $\delta^{18}\text{O}$ (average winter precipitation $\delta^{18}\text{O}$ of approximately -38‰). The combination of cold temperatures and strongly depleted atmospheric vapor resulted in low d-excess of deposited ice. This was consistent with results from an isotope-enabled GCM by Dütsch et al. (2019), who showed that in iCAM5, the ice supersaturation parameterization strongly controls simulated polar precipitation d-excess. The dimensionless ice supersaturation ratio $S_i (=1 + \text{SI}/100)$ is parameterized as a linear function of temperature ($S_i = a + b \cdot T$) 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

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07 reproduce d-excess values of $\sim +20\text{‰}$ observed in ice cores (Dütsch et al., 2019). However, with $b = -0.006$, the condensate
08 has a d-excess of -10‰ (Fig. 1 of Dütsch et al.) when $\delta^2\text{H}_{\text{vapor}} \approx -460\text{‰}$ and $\text{d-excess}_{\text{vapor}} \approx -5$ (equivalent $\delta^{18}\text{O}_{\text{vapor}} \approx -$
09 57‰). The parameter value of $b = -0.006$ at $T = -50^\circ\text{C}$ yields $S_i = 1.3$ (SI = 30%), and at $T = -30^\circ\text{C}$ yields $S_i = 1.18$ (SI =
10 18‰). For these parameters, our calculations give an average d-excess of -12‰ (compared to -8‰ by Dütsch et al.) and
11 provide an explanation for the low d-excess (and low MDV) of Summit winter precipitation. The variability in winter d-
12 excess at Summit would reflect variations in both vapor $\delta^{18}\text{O}$ and SI rather than a riming influence, consistent with the
13 absence of mixed-phase conditions at the low winter temperatures. Conversely, higher summer SI values at warmer growth
14 temperatures — where ambient vapor would be less depleted — result in higher d-excess through the kinetic fractionation
15 mechanism in vapor deposition (Jouzel and Merlivat, 1984). The variability in summer d-excess at Summit would be
16 modulated by variability in SI and riming under favorable conditions.

17 In the case of riming by wet-growth, the sensitivity analysis identifies ambient temperature as the dominant control on rimed
18 ice d-excess ($\sim 18\text{‰}$ range across -15 to -5°C), followed by liquid water content ($\sim 25\text{‰}$ range across 0.05 to 3.0 g m^{-3}) and
19 particle size ($\sim 16\text{‰}$ range across 0.5 to 5 mm), while cloud water d-excess contributes a moderate effect ($\sim 6\text{‰}$ range). At $T =$
20 -5°C , typical of the riming zone below $\sim 700\text{ m}$ at Ny-Ålesund and of ice-phase growth above the melting layer at tropical
21 and mid-latitude sites, we estimate d-excess values of 0 to $+5\text{‰}$ for cloud water d-excess of $+7$ to $+10\text{‰}$ — substantially
22 lower than the $+15$ to $+20\text{‰}$ produced by vapor deposition under the same source vapor conditions. The boundary for wet or
23 dry-growth riming — defined by whether the supercooled water collection rate exceeds the freeze rate — is temperature and
24 LWC dependent, with wet-growth becoming increasingly restricted to temperatures warmer than -10 to -15°C at low LWC
25 values typical of stratiform clouds. In turbulent cloud conditions, both the droplet collection rate (through enhanced relative
26 fall velocity) and the available LWC (through adiabatic updraft condensation) increase, lowering the minimum LWC
27 threshold for wet-growth by ~ 10 – 15% . This potentially shifts riming from the dry to wet-growth regime even in
28 predominantly stratiform environments, consistent with the findings of Chellini and Kneifel (2024) that riming intensity at
29 Ny-Ålesund correlates with turbulence.

30 Dry-growth riming, which operates at temperatures below approximately -15°C and low LWC where collected droplets
31 freeze immediately without forming a persistent film, applies only the equilibrium liquid-ice fractionation without the kinetic
32 film evaporation term. Our calculations show that dry-growth rimed ice has d-excess approximately 2 – 3‰ lower than the
33 collected cloud water at -15°C , declining toward zero difference at -30°C as the equilibrium fractionation factors for
34 deuterium and ^{18}O converge.

35

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37 **3.5.3 Process weighted d-excess**

38 We used a range of vapor and cloud water isotope compositions and riming growth parameters that were reasonable for the
39 study locations to calculate end-member isotopic compositions (Table 4). Process-weighted d-excess of surface precipitation
40 (d_{surface}) was calculated as the mass-weighted sum of the four end-member contributions (Table 5):

41
$$d_{\text{surface}} = f_{\text{dep}} \cdot d_{\text{dep}} + f_{\text{dust}} \cdot d_{\text{dust}} + f_{\text{rim-wet}} \cdot d_{\text{rim-wet}} + f_{\text{rim-dry}} \cdot d_{\text{rim-dry}} \quad (8)$$

42 where f_{dep} , f_{dust} , $f_{\text{rim-wet}}$ and $f_{\text{rim-dry}}$ are the fractional contributions of each growth process summing to unity.

43 For the maritime polar, mid-latitude and tropical sites (Ny-Ålesund, Dumont d'Urville, Cazadero and Rio Claro), rimed ice
44 fractions ranged from 0.2 to 0.6, consistent with estimated RMF (Fig. S8), with the rest being vapor deposition. Diamond-
45 dust was not included for these polar sites as the temperature and SI profiles during the isotope sampling days indicated
46 mixed-phase conditions at all heights (Sect. 3.2). Process-weighted mixtures spanning 40–80% vapor deposition with the
47 remainder as wet and/or dry riming produced d-excess values of −5.8 to +29.3‰, consistent with the observed precipitation
48 d-excess range at these sites. The mixture with 40% vapor deposition and 60% wet-growth riming produces d-excess ≈ 0‰,
49 consistent with heavily rimed events approaching the graupel end-member. Vapor deposition events (100% dep) reach d-
50 excess = +44.2‰ (Table 4), consistent with the highest observed values at Ny-Ålesund.

51 For Summit summer, vapor deposition at $T_c = -25^\circ\text{C}$ and $S_i = 1.20$ (SI = 20%) yields d-excess = +29.4‰ (Table 4).
52 Mixtures of 80% vapor deposition with 20% dry-growth riming or 20% wet-growth riming produce d-excess of +24.4‰ and
53 +17.6‰, respectively, and 30% wet-growth riming with 10% dry-growth results in a d-excess of +9.3‰, consistent with the
54 observed summer d-excess range of approximately +10 to +30‰.

55 For Summit winter, the vapor deposition end-member at $T_c = -35^\circ\text{C}$ and $S_i = 1.20$ produces d-excess = +13.9‰ while the
56 diamond dust end-member at $T_c = -50^\circ\text{C}$ and $S_i = 1.20$ yields d-excess = −1.1‰. Mixtures of vapor deposition and diamond
57 dust spanning 20–80% diamond dust fraction produce d-excess of −7.5 to +8.5‰, consistent with the observed winter d-
58 excess range of approximately 0 to +15‰ at Summit. The strongly negative d-excess of the diamond dust end-member —
59 forming at very cold temperatures from strongly depleted source vapor — drives the mixture d-excess well below the vapor
60 deposition end-member even at modest diamond dust fractions.

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Table 4. Calculated isotope compositions of end-members from different growth processes*

62

End Member	Vapor		T	S _i or	Ice			Additional parameters
	δ ¹⁸ O _v (‰)	d _v (‰)	(°C)	LWC	δ ¹⁸ O (‰)	δ ² H (‰)	d (‰)	
Vapor Deposition								
A	-25	10	-20	1.30	-16.9	-91	44.2	
B	-30	10	-25	1.20	-18.4	-118	29.4	
C	-40	20	-25	1.15	-26.9	-192	23.4	
D	-40	10	-30	1.10	-24.8	-194	4.8	
E	-50	15	-35	1.20	-37.0	-282	13.9	
F	-50	5	-35	1.20	-37.1	-294	2.2	
Diamond dust								
G	-65	10	-50	1.20	-49.6	-410	-1.1	
Wet-growth								
H	-30	10	-10	0.5	-11.4	-120	-31	δ ¹⁸ O _{cw} =-19; d _{cw} =19; R=3; V=1.0; E=0.8
I	-20.0	10	-10	0.5	-6.2	-76	-26.6	-same-
J	-20.0	20	-5	0.5	-7.5	-68	-7.4	-same-
K	-25.0	10	-10	0.5	-4.3	-65	-30.3	δ ¹⁸ O _{cw} =-10; d _{cw} =10; R=3; V=1.0; E=0.8
L	-30.0	20	-10	0.3	-9.5	-105	-1.0	δ ¹⁸ O _{cw} =-17; d _{cw} =21; R=2; V=0.8; E=0.8
Dry-growth								
M	-30	20	-20		-14.4	-111	5.4	δ ¹⁸ O _{cw} =-17; d _{cw} =10

*Input isotope values: the subscripts v and cw refer to ambient vapor and cloud water; T = growth temperature; S_i = ice supersaturation ratio; LWC = liquid water content (g m⁻³); R = particle radius (mm); V = relative fall speed (m s⁻¹); E = collection efficiency.

Table 5. Precipitation isotope compositions based on process-weighted mixtures of end-member components from Table 4.

Components	End-member fractions				δ ¹⁸ O (‰)	δ ² H (‰)	d (‰)
	f _{dep}	f _{dust}	f _{wet}	f _{dry}			
A + K	0.8		0.2		-14.2	-86	29.3
A + K	0.4		0.6		-9.3	-75	-0.5
B + K	0.4		0.6		-13	-110	-5.8
A + J + M	0.4		0.5	0.1	-10.3	-83	3.0
B + J	0.8		0.2		-16.6	-115	17.6
B + J+M	0.6		0.3	0.1	-15.7	-114	11.8
B + M	0.8			0.2	-17.6	-116.0	24.4
F + G	0.8	0.2			-39.5	-308	8.5
F + G	0.2	0.8			-47.1	-384	-7.5

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71 **4 Discussion**

72 The inverse correlation of d-excess with MDV (or rimed mass fraction) and the process-weighted framework developed here
73 have broader implications for the interpretation of d-excess in ice core records and modern precipitation.

74
75 **d-excess–RMF parameterization**

76 The total least squares regression of d-excess and RMF across all sites (except Summit) yields a slope of approximately
77 –124‰ per unit RMF (Fig. S8), providing a simple, observationally constrained parameterization: Each 0.1 unit (10%)
78 increase in RMF corresponds to a d-excess decrease of ~12‰. The convergence of this slope across sites ranging from
79 tropical Rio Claro to polar Ny-Ålesund and Dumont d'Urville, spanning a wide range of cloud temperatures, LWC, and
80 atmospheric conditions, suggests that it reflects a robust microphysical relationship rather than a site-specific artifact. This
81 parameterization offers a practical diagnostic for isotope-enabled climate models such as iCAM5 (Nusbaumer et al., 2017),
82 which currently treat riming as isotopically neutral. Where Doppler radar observations are available alongside modeled
83 precipitation fields, the predicted lowering of d-excess from simulated riming intensity could be compared against observed
84 precipitation d-excess to evaluate whether model microphysics schemes produce physically realistic riming rates.
85 Conversely, where isotope observations exist but radar data do not, the parameterization allows rimed mass fraction to be
86 estimated directly from measured d-excess, providing a microphysical constraint that is otherwise difficult to obtain from
87 surface observations alone.

88
89 **Paleoclimate implications**

90 The lower d-excess of ice cores from the Last Glacial Maximum (LGM) compared to the Holocene, and the d-excess
91 changes associated with abrupt stadial-interstadial transitions in Greenland (Jouzel et al., 2007; Johnsen et al., 1989), have
92 been interpreted as evidence for major reorganization of moisture source regions. The results of this study suggest an
93 alternative or complementary interpretation that does not require source region changes. The Summit winter case
94 demonstrates that vapor deposition at very low temperatures (–30 to –65°C), from strongly depleted vapor under high ice
95 supersaturation, suppresses precipitation d-excess to low or negative values entirely through in-cloud fractionation physics,
96 without any riming contribution. Glacial conditions at high-latitude ice core sites were characterized by temperatures
97 substantially colder than present, more isotopically depleted atmospheric vapor, and likely higher ice supersaturation during
98 precipitation events (Jouzel and Merlivat, 1984; Dütsch et al., 2019). These are precisely the conditions that the process-
99 weighted framework identifies as producing low d-excess through vapor deposition, independent of moisture source. The
00 glacial-to-Holocene increase in d-excess at Greenland ice core sites may therefore reflect, at least in part, the warming of the
01 precipitating column and the associated increase in vapor $\delta^{18}\text{O}$ and reduction in ice supersaturation — shifting ice growth
02 conditions away from the cold, high-SI regime that characterizes Summit winter today — rather than, or in addition to, a
03 northward shift in moisture source origin. This interpretation is consistent with the back-trajectory analysis of Pettersen et al.

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(2018), which found no systematic difference in moisture source between Summit precipitation types despite large differences in d-excess, and with GCM sensitivity experiments showing that the choice of ice supersaturation parameterization strongly controls simulated polar d-excess (Dütsch et al., 2019). Distinguishing the in-cloud fractionation signal from the source moisture signal in ice core records will require isotope-enabled models that explicitly represent both the temperature and SI dependence of vapor deposition fractionation and the riming contribution documented here.

Spatial variations in d-excess of polar surface snow

In Greenland (Fig. S12a), surface snow d-excess decreases from north to south — from approximately +14‰ in the northwest (near Camp Century; Osterberg et al., 2015), through +9.5‰ at GRIP (Summit), to +8‰ at Dye 3 (Johnsen et al., 1989) — a gradient traditionally attributed to different moisture sources (Johnsen et al., 1989; Masson-Delmotte et al., 2005). However, the same gradient would be consistent with a systematic change in d-excess resulting from vapor deposition at different ice supersaturation and temperature conditions along the north-south transect. Compared to GRIP (Summit), Camp Century has a lower surface elevation and is located closer to the coast (~200 km inland). These differences likely would result in ice growth at Camp Century to occur at lower temperatures and different ice supersaturation conditions than at GRIP. Similarly, lower d-excess from riming contribution would be important at Dye 3 in southcentral Greenland, where proximity to the coast (~120 km inland) and warmer cloud temperatures favor frequent mixed-phase conditions and riming (Borvs et al., 1993). Distinguishing between these controls requires simultaneous Doppler radar observations and isotope sampling of daily precipitation at multiple Greenland sites.

In Antarctica (Fig. S12b), coastal precipitation has consistently lower d-excess than interior plateau precipitation (Wang et al., 2022) — a contrast attributed to the closer proximity of coastal sites to marine moisture sources with higher relative humidity (Masson-Delmotte et al., 2008). However, the coastal Antarctic environment is characterized by frequent frontal precipitation with vigorous dynamics, high LWC in mixed-phase clouds, and extensive riming as documented for Dumont d'Urville (Grazioli et al., 2017). Moderate to heavy riming has been noted at McMurdo (Tridon et al., 2022) and the Mario Zucchelli station (Sarchilli et al., 2020). At both of these locations, snow particle fall velocities are frequently greater than $\sim 1 \text{ m s}^{-1}$ near the surface, consistent with the low d-excess values resulting from more frequent and heavier riming. Warburton (1978) noted that rimed ice crystals were dominant in falling snow near McMurdo station and that their proportion decreased with distance from the coast as growth by vapor deposition became dominant in inland precipitation. Different in-cloud processes may therefore sufficiently explain the spatial d-excess variations across Antarctica rather than, or in addition to, source moisture differences. Distinguishing between these two controls may be possible with more detailed analysis using simultaneous Doppler radar observations and isotope sampling of daily precipitation.

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Deleted: We have used the MDV of snow particles during snow or rain precipitation as a proxy for riming. Its inverse correlation with the d-excess of precipitation collected on the surface in polar to tropical regions shows that isotopic fractionation occurs via riming and results in a lower d-excess of rimed ice compared to that of the liquid. This fractionation is opposite to that during ice growth by the WBF process where the ice phase has a higher d-excess.

The MDV can also be used to characterize the rimed mass fraction (RMF), the ratio of particle mass acquired by riming to the total mass, that is important for understanding the influence of microphysics on precipitation formation and the impact of climate change (Morrison et al., 2020). Improved simulations of future Greenland ice sheet melting and mass budget also require a better characterization of the liquid or ice phase of cloud particles (Morrison et al., 2020). A correlation of d-excess with RMF could be used to determine spatial and temporal patterns of RMF variations in modern precipitation and ice cores. The lower d-excess from riming would also help to explain the spatial variations of surface snow on Greenland and Antarctica that were previously attributed to changes in source moisture origin and reorganization of high-latitude hydrological cycles. The d-excess correlation with RMF and spatial variation of d-excess in the polar regions are discussed below.

4.1 Rimed mass fraction

Methods to calculate RMF from MDV have been proposed by Mosimann (1995) and Kneifel and Moiseev (2020). Mosimann (1995) used a vertically-pointing Doppler radar to measure the MDV of snow in the Swiss Alps. This was correlated with the RMF estimated from a visual analysis of individual snow particles (Mosimann et al., 1994). More recently, Kneifel and Moiseev (2020) used an extensive database of particle shapes, size, and fall velocities measured in precipitation at Hyytiälä, Finland to calculate the RMF. The rimed mass was calculated as the difference between the total mass and unrimed particle mass. The total particle mass was estimated from the fall velocity and mean particle mass for a given size bin (von Lerber et al., 2017). The unrimed mass was determined by assuming that the smallest 5% of the particles in each of the disdrometer's 0.2 mm size bins were unrimed (Moiseev et al., 2017). Using the measured particle size distribution, Kneifel and Moiseev (2020) calculated the expected MDV and a fourth-degree polynomial regression gave the best fit to the calculated RMF and MDV values. The relationship proposed by Kneifel and Moiseev reproduced well the RMF for the original data of Mosimann (1995). We have used the Kneifel and Moiseev (2020) method to calculate the RMF from MDV at all locations in our study.

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69 ***Tropical precipitation and the amount effect***
70 The inverse d-excess–MDV correlation at Rio Claro extends the riming interpretation into the tropics, where stratiform
71 precipitation in mesoscale convective systems is responsible for a large fraction of total rainfall. Low d-excess values in
72 tropical rainfall are commonly attributed to the "amount effect" — the observed depletion of heavy isotopes in high-intensity
73 rainfall — or to sub-cloud evaporation (Dansgaard, 1964; Moerman et al., 2013). However, the results here show that
74 stratiform tropical rainfall with a well-defined melting layer and rimed particles above it carries a low d-excess signal that
75 originates in the ice-phase region, entirely above the sub-cloud layer. In convective mesoscale systems, the seeder-feeder
76 process is well documented during stratiform episodes (Houze, 2014): ice particles grown by vapor deposition in the upper
77 anvil fall into a lower feeder region where supercooled liquid water content is higher and riming occurs. The temporal
78 progression from convective to stratiform precipitation within a single mesoscale event would therefore produce a systematic
79 evolution from very low d-excess — driven by vigorous wet-growth riming in the convective stage — toward higher d-
80 excess as the stratiform stage becomes vapor-deposition dominated. Riming may therefore contribute to the amount effect
81 signal in tropical precipitation, and its role warrants further investigation alongside sub-cloud evaporation and convective
82 intensity.

83
84 ***Mid-latitude frontal precipitation***
85 The systematic temporal evolution of d-excess during mid-latitude frontal precipitation events may also be better understood
86 through the process-based interpretation developed here. At Cazadero, the lowest d-excess values (−22.9 to −13.9‰)
87 occurred in the early hours of the 1 March 2009 event (Fig. 6 and Sect. 3.3) followed by progressively higher d-excess and
88 lower δ¹⁸O (Fig. S13). Low d-excess values in rainfall have commonly been attributed to sub-cloud evaporation, which we
89 have shown may have had only a minor influence on the Cazadero d-excess values (Sect. 3.3). The presence of a melting
90 layer throughout the event (Fig. S6) confirms that precipitation was stratiform during both the early low d-excess and later
91 higher d-excess periods. The temporal evolution of d-excess is physically consistent with the known evolution of riming
92 intensity within frontal precipitation systems: in the early stage, embedded convective cells and turbulent motions within the
93 stratiform region generate higher LWC and more vigorous riming — conditions identified in the sensitivity analysis as
94 producing the lowest d-excess — while the mature stratiform stage has weaker vertical motions, lower LWC, and vapor-
95 deposition-dominated growth producing higher d-excess (Matejka et al., 1980; Houze, 2014). The temporal MDV–d-excess
96 correlation within the Cazadero event is therefore consistent with the expected evolution of riming intensity within a single
97 frontal precipitation event, and mirrors the tropical case in reflecting the convective-to-stratiform transition.

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26 **5 Conclusions**

27 We investigated the effect of riming on the d-excess of precipitation at six locations spanning the tropics to the polar regions:
28 Summit, Greenland; Ny-Ålesund and Andenes, Norway; Dumont d'Urville, Antarctica; Cazadero, California; and Rio Claro,
29 Brazil. The mean Doppler velocity (MDV) of precipitating hydrometeors was used as an independent, physically based
30 indicator of riming intensity. Riming increases both particle density — replacing low-density dendritic ice with dense
31 accreted ice — and effective diameter, both of which raise terminal fall speed substantially. MDV is therefore a direct proxy
32 for the degree of riming that is entirely independent of the isotopic measurements.

33
34 Daily or sub-daily precipitation d-excess was correlated with concurrent MDV measured near the surface (150–300 m AGL)
35 during snowfall and above the melting layer (~2–4.5 km AGL) during rainfall. An inverse correlation between d-excess and
36 MDV was found at all locations except Summit winter, spanning a wide range of temperatures, moisture sources, and
37 atmospheric conditions. Because the MDV values used for rainfall were extracted from the ice-phase region above the
38 melting layer, they are unaffected by sub-cloud evaporation. Sub-cloud evaporation may slightly lower the d-excess of
39 individual rain samples at the surface, but it cannot produce the systematic, multi-site inverse correlation of d-excess and
40 MDV shown here.

41
42 Vertical profiles of temperature, relative humidity, and ice supersaturation index (SI) derived from radiosonde soundings at
43 Summit, Ny-Ålesund, and Dumont d'Urville, provide independent atmospheric context that corroborates the microphysical
44 interpretation. At Summit, these profiles show that the low winter temperatures (<−38°C) in the entire precipitating column
45 preclude mixed-phase conditions and riming while summer profiles reveal the occurrence of near-liquid-saturation
46 conditions at temperatures between −4 and −15°C, consistent with wet-growth riming. The low d-excess of winter Summit
47 precipitation was shown to result from ice growth by vapor deposition at very low temperatures (−30 to −65°C) and ice
48 supersaturated conditions from vapor depleted in $\delta^{18}\text{O}$, consistent with published isotope-enabled GCM simulations (Dütsch
49 et al., 2019). At Ny-Ålesund and Dumont d'Urville, the T and SI profiles reveal a two-layer growth structure in which vapor
50 deposition dominates in the upper, ice-supersaturated column while riming occurs in the lower, near-liquid-saturated layer,
51 consistent with the seeder-feeder cloud structure documented at those sites.

52
53 The physical mechanism responsible for the lower d-excess of rimed ice operates through evaporation of the accreted liquid
54 film during wet-growth riming. When the rate of supercooled droplet collection exceeds the rate at which latent heat of
55 fusion can be conducted away, the particle surface temperature rises to 0°C while ambient air remains at temperatures below
56 freezing. This temperature differential drives evaporation of water from the liquid film into the subsaturated ambient air.
57 Because HDO diffuses more slowly than H_2^{18}O in air, evaporation preferentially removes ^{18}O relative to D from the accreted
58 liquid, lowering the d-excess of the liquid that ultimately freezes. Sensitivity calculations show that this mechanism produces

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In this study, we investigated the effect of the riming process of snow formation, where supercooled liquid droplets collide with falling ice particles in mixed-phase clouds and freeze directly on the particle surface, on the d-excess of ice. We used the terminal fall velocity of snow particles, measured with vertically-pointing Doppler radars, as an independent indicator of riming at several locations in the Arctic (Summit, Greenland, Ny-Ålesund, Svalbard and Andenes, Norway), Antarctic (Dumont d'Urville), mid-latitudes (Cazadero, California) and tropics (Rio Claro, Brazil). Concurrent measurements of d-excess and terminal fall velocity in daily or sub-daily precipitation are shown to be inversely correlated, except for the winter precipitation at Summit, Greenland. The d-excess in the winter precipitation at Summit is lower compared to the summer, while the terminal fall velocities are similar. This may be an artefact of smaller, less dense particles in the winter resulting in lower terminal fall velocity or a much lower d-excess in riming growth at colder temperatures. Excluding the data from Summit, a strong correlation ($R^2 = 0.59$) of d-excess and terminal fall velocity in precipitation from the rest of the polar and non-polar locations indicates robust dependence of d-excess on riming. This is the result of isotopic fractionation during riming by evaporation, and possibly partial loss of the accreted liquid by splashing and shedding, before freezing is complete. Isotopic fractionation during riming is contrary to the existing view, which holds that riming occurs without fractionation, but is consistent with previously published field, experimental, and modelling studies.

Consideration of lower d-excess resulting from riming helps to reconcile the spatial d-excess variations in surface snow observed across Greenland and Antarctica. In Greenland, increased riming to the south likely results in lower d-excess values. Similarly, lower d-excess in the coastal areas and higher values inland on Antarctica, which have been thus far difficult to explain based on source moisture origins, are consistent with increased riming in the coastal areas.

Although our study focused on stratiform precipitation in order to allow the use of terminal fall velocity as an indicator of riming, the results are applicable for interpreting the lower d-excess of all precipitation. In particular, the lower d-excess of precipitation that is conventionally attributed to sub-cloud evaporation in mid-latitude and tropical precipitation likely results instead from riming with only a minor contribution from sub-cloud evaporation.

The fraction of snow particle mass attributed to riming (RMF) was calculated by using the terminal fall velocity and a strong inverse correlation is also shown between d-excess and RMF. Therefore, d-excess variations in precipitation and ice cores can be used to characterize RMF and its spatial or temporal variability.

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88 rimed ice with substantially lower d-excess than the liquid (by as much as 20–30‰) while dry-growth riming at temperatures
89 below approximately −15°C produces only a modest d-excess reduction of 2–3‰ through equilibrium liquid-ice
90 fractionation.

91

92 A process-weighted framework was developed to quantify the d-excess of precipitation as a mass-weighted mixture of four
93 end-member growth modes: vapor deposition, diamond dust, wet-growth riming, and dry-growth riming. Process mixtures
94 spanning 20–80% vapor deposition (or 100% for Summit winter) with the remainder as riming reproduce the observed d-
95 excess range across all study sites. These calculations demonstrate that in-cloud microphysical processes alone can account
96 for the observed spatial and temporal d-excess variability from tropical to polar precipitation regimes.

97

98 Our findings suggest that low d-excess in precipitation and ice cores, traditionally attributed to changes in source moisture
99 origin or sub-cloud evaporation, warrants reexamination. This does not imply that vapor composition is irrelevant: the $\delta^{18}\text{O}$
00 and d-excess of ambient vapor are explicit inputs to the fractionation calculations, and their seasonal and spatial variability
01 contributes meaningfully to d-excess variability. Rather, we argue that the in-cloud fractionation component — controlled by
02 growth temperature, ice supersaturation, and riming intensity — is of comparable or greater magnitude than the vapor signal
03 across many precipitation regimes. Separating these two contributions requires independent constraints on in-cloud
04 conditions, such as those provided by the radar-based riming proxy introduced here. The conventional attribution of d-excess
05 variability solely to oceanic evaporation conditions (sea surface temperature and relative humidity) requires additional
06 caution: the source evaporation signal is substantially modified by Rayleigh distillation and vapor mixing during
07 atmospheric transport and during ice growth by vapor deposition and riming.
08

09 **Data availability**

10 All data used in this study were obtained from published sources or public databases, as cited in the text.

11 **Supplement link**

12 Supplementary material is provided in a separate file.

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13 **Author contributions**

14 PKA conceived the project and designed the methodology, compiled isotope and radar data, and conducted the analyses; CS,
15 FJL and MDS contributed to refining the methodology; AF assisted in processing radar data; PKA wrote the original draft;
16 CS, FJL and MDS critically reviewed and edited the original draft; PKA prepared the final manuscript.

17 **Competing interests**

18 There are no competing interests

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24 [kindly facilitated access to raw MRR data for 2014–2017 from Ny-Ålesund that were collected as part of the Indian Arctic](#)
25 [Programme and accessed through the NCPOR data repository \(<https://data.ncpor.res.in/newhtml/3>\)](#). MDS was supported by
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29 **References**

30 Atlas, D., Srivastava, R. C. and Sekhon, R. S.: Doppler radar characteristics of precipitation at vertical incidence. Rev.
31 Geophys., 11, doi:10.1029/rg011i001p00001, 1973.

32

33 Atmospheric Radiation Measurement (ARM) user facility: Millimeter Wavelength Cloud Radar (MMCRMOM). 2010-06-01
34 to 2017-09-01, Summit Station, Greenland (SMT) External Data (satellites and others) (X1). Compiled by N. Bharadwaj.

35 ARM Data Center. Data set accessed 2023-03-27 at <http://dx.doi.org/10.5439/1025228>, 2010 [dataset].

36

37 Atmospheric Radiation Measurement (ARM) user facility: Active Remote Sensing of CLOUDS (ARSCL) product using Ka-
38 band ARM Zenith Radars (ARSCLAZRIKOLLIAS), 2020-02-22 to 2020-03-24, ARM Mobile Facility (ANX) Andenes,

Deleted: <http://dx.doi.org/10.5439/1025228>, 2010.

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40 Norway; AMF1 (main site for COMBLE) (M1). Compiled by K. Johnson, M. Jensen and S. Giangrande. ARM Data Center.
41 Data set accessed 2026-01-17 at <http://dx.doi.org/10.5439/1228768>, 2019 [dataset].

Deleted: <http://dx.doi.org/10.5439/1228768>, 2019.

42 [v](#)
43 Bailey, A., Noone, D., Dee, S.G., Nusbaumer, J., Conroy, J.L., Stevenson, S. and Atwood, A.: Toward a process-oriented
44 understanding of water in the climate system: recent insights from stable isotopes, *Env. Res.: Climate* 4 012002,
45 doi:10.1088/2752-5295/ada17b, 2025.

Deleted: ¶

... [14]

46
47 Bailey, H.L., Klein, S.L. and Welker, J.M.: Synoptic and mesoscale mechanisms drive winter precipitation $\delta^{18}\text{O}/\delta^2\text{H}$ in
48 south-central Alaska, *J. Geophys. Res., Atmospheres* 124, no. 7 (2019): 4252-4266.

49
50 Bailey, I. H., Hulston, J. R., Macklin W. C. and Stewart, J. R.: On the isotopic composition of hailstones. *Jour. Atm. Sci.* 26,
51 689-694, 1969.

52
53 [Borys, R.D., Del Vecchio, D., Jaffrezo, J.-L., Davidson, C.I., Mitchell, D.L.: Assessment of Ice Particle Growth Processes at](#)
54 [Dye-3, Greenland, *Atm. Env.*, 27A, 2815-2822, 1993.](#)

Moved (insertion) [23]

55
56 Casado, M., Cauquoin, A., Landais, A., Israel, D., Orsi, A., Pangui, E., Landsberg, J., Kerstel, E., Prie, F. and Doussin, J.-F.:
57 Experimental determination and theoretical framework of kinetic fractionation at the water ~~vapor~~-ice interface at low
58 temperature, *Geochim. Cosmochim. Acta*, 174, 54-69, 2016.

Deleted: vapour

59
60 Castellani, B. B., Shupe, M. D., Hudak, D. R. and Sheppard, B. E.: The annual cycle of snowfall at Summit, Greenland. *J.*
61 *Geophys. Res.: Atm.*, 120, 6654-6668, 2015.

62
63 Chellini, G. and Kneifel, S.: Turbulence as a key driver of ice aggregation and riming in Arctic low-level mixed-phase
64 clouds, revealed by long-term cloud radar observations, *Geophys. Res. Lett.* 51, e2023GL106599, 2024.

65
66 Ciais, P. and Jouzel J.: Deuterium and oxygen 18 in precipitation: Isotopic model, including mixed cloud processes. *J.*
67 *Geophys. Res.: Atm.* 99, 16793-16803, 1994.

68
69 Coplen, T. B., Neiman, P. J., White, A. B. and Ralph, F. M.: Categorization of northern California rainfall for periods with
70 and without a radar bright band using stable isotopes and a novel automated precipitation collector, *Tellus B: Chem. Phys.*
71 *Met.* 67, 28552-28574, 2015, [\[dataset\]](#).

Deleted: .

72
73 Dansgaard, W.: Stable isotopes in precipitation, *Tellus* 16, 436-468, 1964.

Deleted: 46¶

| [46](#)

83
84 Dansgaard, W., Johnsen, S. J., Møller, J. and Langway, C. C. Jr.: One thousand centuries of climatic record from Camp
85 Century on the Greenland ice sheet, *Science* 166, 377-381, 1969.
86
87 Demoz, B. B., Warburton, J. A. and R. H. Stone: The influence of riming on the oxygen isotopic composition of ice-phase
88 precipitation, *Atmos. Res.* 26, 463-488, 1991.
89
90 Durán-Alarcón, C., Boudevillain, B., Genthon, C., Grazioli, J., Souverijns, N., van Lipzig, P.M., Gorodetskaya, I. V. and
91 Berne, A.: The vertical structure of precipitation at two stations in East Antarctica derived from micro rain radars, *The*
92 *Cryosph.* 13, 247-264, 2019.
93
94 Dütsch, M., Blossey, P. N., Steig, E. J. and Nusbaumer, J. M.: Nonequilibrium fractionation during ice cloud formation in
95 iCAM5: Evaluating the common parameterization of supersaturation as a linear function of temperature, *Jour. Adv.*
96 *Modeling Earth Sys.* 11, 3777-3793, 2019.
97
98 Ebell, K., Walbröl, A. and Krobot, K.: Measurements of micro rain radar at AWIPEV, Ny-Ålesund (2017-2021), [Data set].
99 <https://doi.org/10.1594/PANGAEA.958967>, PANGAEA, 2023 [\[dataset\]](#).
00
01 Ebell, K., Buhren, C., Gierens, R., Chellini, G., Lauer, M., Walbröl, A., Dahlke, S., Krobot, P. and Mech, M.: Multi-year
02 precipitation characteristics based on in-situ and remote sensing observations at Ny-Ålesund, Svalbard. *EGUsphere* 1-34,
03 2024.
04
05 Federer, B., Brichet, N. and Jouzel, J.: Stable isotopes in hailstones. Part I: The isotopic cloud model, *J. Atm. Sci.*, 39, 1323-
06 1335, 1982.
07
08 Fitch, K. E. and Garrett, T. J.: Graupel precipitating from thin Arctic clouds with liquid water paths less than 50 g m⁻²,
09 *Geophys. Res. Lett.* 49, e2021GL094075, 2022.
10
11 Garrett, T. J. and Yuter, S. E.: Observed influence of riming, temperature, and turbulence on the fallspeed of solid
12 precipitation, *Geophys. Res. Lett.*, 41, 6515-6522, 2014.
13
14 Graf, P., Wernli, H., Stephan, P. and Sodemann, H.: A new interpretative framework for below-cloud effects on stable water
15 isotopes in [vapor](#) and rain, *Atm. Chem. Phys.*, 19, 747-765, 2019.
16
47

Deleted: Davidson, C. I., Harrington, J. R., Stephenson, M. J., Small, M. J., Boscoe, F. P. and Gandle, R. E.: Seasonal variations in sulfate, nitrate and chloride in the Greenland ice sheet: Relation to atmospheric concentrations, *Atm. Env.*, 23, 2483-2493, 1989. [¶](#)

Moved up [23]: L.,

Moved down [24]: *Atm.*

Deleted: Di Natale, G., Turner, D. D., Bianchini, G., Del Guasta, M., Palchetti, L., Bracci, A., Baldini,

Deleted: Maestri, T., Cossich, W., Martinazzo, M. and Facheris, L.: Consistency test of precipitating ice cloud retrieval properties obtained from the observations of different instruments operating at Dome C (Antarctica),

Deleted: *Meas. Tech.* 15, 7235-7258, 2022. [¶](#)

Deleted: Field, P. R. and Heymsfield, A. J.: Importance of snow to global precipitation. *Geophys. Res. Lett.* 42, 9512-9520, 2015. [¶](#)

Deleted: Fujita, K. and Abe, O.: Stable isotopes in daily precipitation at Dome Fuji, East Antarctica, *Geophys. Res. Lett.*, 33, L18503, 2006. [¶](#)

Deleted: Gettelman, A., Morrison, H., Santos, S., Bogenschutz, P. and Caldwell, P. M.: Advanced two-moment bulk microphysics for global models. Part II: Global model solutions and aerosol–cloud interactions, *J. Clim.* 28, 1288-1307, 2015. [¶](#)

Deleted: vapour

Deleted: 47 [¶](#)

45 Grazioli, J., Genthon, C., Boudevillain, B., Duran-Alarcon, C., Del Guasta, M., Madeleine, J.-B. and Berne, A.:
46 Measurements of precipitation in Dumont d'Urville, Adélie Land, East Antarctica, The Cryos., 11, 1797-1811, 2017.
47
48 Harimaya, T. and Sato, M.: Measurement of the riming amount on snowflakes, J. Fac. Sci. Hokkaido University:
49 Geophysics, 8, 355-366, 1989.
50
51 Heymsfield, A. J., Schmitt, C. and Bansemer, A.: Ice cloud particle size distributions and pressure-dependent terminal
52 velocities from in situ observations at temperatures from 0 to− 86 C, J. Atm. Sci., 70, 4123-4154, 2013.
53
54 Heymsfield, A. J. and Kajikawa, M.: An improved approach to calculating terminal velocities of plate-like crystals and
55 graupel, J. Atm. Sci., 44, 1088-1099, 1987.
56
57 Houze Jr, R. A., Cloud dynamics. Vol. 104. Academic press, 2014.
58
59 Johnsen, S. J., Dansgaard, W. and White, J. W. C.: The origin of Arctic precipitation under present and glacial conditions,
60 Tellus B: Chem. Phys. Meteo., 41, 452-468, 1989.
61
62 Jouzel, J., Delaygue, G., Landais, A., Masson-Delmotte, V., Risi, C., Vimeux, F.: Water isotopes as tools to document
63 oceanic sources of precipitation, Water Resources Res. 49, 7469-7486, 2013.
64
65 Jouzel, J. and Merlivat, L.: Deuterium and oxygen 18 in precipitation: Modeling of the isotopic effects during snow
66 formation, J. Geophys. Res.: Atmospheres, 89-D7, 11749-11757, 1984.
67
68 Jouzel, J., Merlivat, L. and Federer, B.: Isotopic study of hail: The δD-δ18O relationship and the growth history of large
69 hailstones, Quart. J. Royal Met. Soc., 111, 495-516, 1985.
70
71 Jouzel, J., Stievenard, M., Johnsen, S. J., Landais, A., Masson-Delmotte, V., Sveinbjornsdottir, A., Vimeux, F., Von
72 Grafenstein, U. and White J. W. C.: The GRIP deuterium-excess record, Quat. Sci. Rev. 26, 1-17, 2007.
73
74 Karrer, M., Dias Neto, J. von Terzi, L. and Kneifel S.: Melting behavior of rimed and unrimed snowflakes investigated with
75 statistics of triple-frequency Doppler radar observations, J. Geophys. Res.: Atmospheres, 127, e2021JD035907, 2022.
76
77 Kneifel, S. and Moisseev., D.: Long-term statistics of riming in nonconvective clouds derived from ground-based Doppler
78 cloud radar observations, J. Atm. Sci., 77, 3495-3508, 2020.

Deleted: d'Urville

Deleted: Holme, C., Gkinis, V., Lanzky M., Morris, V., Olesen, M., Thayer, A., Vaughn, B. H. and Vinther, B. M.: Varying regional δ 18 O–temperature relationship in high-resolution stable water isotopes from east Greenland, Clim. Past, 15, 893-912, 2019.

Moved down [25]: J.

Deleted: Hogan, A. W., Barnard, S. C., Kebschull, K., Townsend, R., Samson, J. A.: Aerosol variation in the western hemisphere Arctic,

Deleted: Aerosol. Sci., 15, 13-33, 1984.

Deleted: 48

91
92
93
94
95
96
97
98
99
00
01
02
03
04
05
06
07
08
09
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24

Kopeck, B. G., Feng, X., Posmentier, E. S. and Sonder, L. J.: Seasonal deuterium excess variations of precipitation at Summit, Greenland, and their climatological significance, J. Geophys. Res.: Atmospheres 124, 72-91, 2019, [\[dataset\]](#).

[Korolev, A. V., Mazin, I. P.: Supersaturation of water vapor in clouds." J. *Atm. Sci.* 60, 2957-2974, 2003.](#)

Korolev, A., McFarquhar, G., Field, P. R., Franklin, C., Lawson, P., Wang, Z., E. Williams et al.: Mixed-phase clouds: Progress and challenges, *Meteo. Mon.* 58, 5-1, 2017.

Kumjian, M. R. and Ryzhkov, A. V.: The impact of evaporation on polarimetric characteristics of rain: Theoretical model and practical implications, J. App. *Meteo. Clim.*, 49, 1247-1267, 2010.

[Lauer, M., Rinke, A., Gorodetskaya, I., Sprenger, M., Mech, M. and Crewell, S.: Influence of atmospheric rivers and associated weather systems on precipitation in the Arctic, *Atm. Chem. Phys.*, 23, 15, 8705-8726, 2023.](#)

Leroy-Dos Santos, C.: From atmospheric water isotopes measurement to firn core interpretation in Adelie Land: A case study for isotope-enabled atmospheric models in Antarctica: [Zenodo. *https://doi.org/10.5281/zenodo.7708489*, 2023.](#) [\[dataset\]](#).

Leroy-Dos Santos, C.: A 4.5 year-long record of Svalbard water vapor isotopic composition documents winter air mass origin V1, [Zenodo. *https://doi.org/10.5281/zenodo.3689566*, 2020](#) [\[dataset\]](#).

Lin, Y. and Colle, B. A.: A new bulk microphysical scheme that includes riming intensity and temperature-dependent ice characteristics, *Monthly Weather Rev.*, 139, 1013-1035, 2011.

[Locatelli, J. D. and Hobbs, P. V.: Fall speeds and masses of solid precipitation particles, J. Geophys. Res. 79, 2185-2197, 1974.](#)

Lowenthal, D. H., Borys, R. D., Cotton, W., Saleeby, S., Cohn, S. A. and Brown, W. O. J.: The altitude of snow growth by riming and vapor deposition in mixed-phase orographic clouds, *Atm. Env.*, 45, 519-522, 2011.

Lowenthal, D., Gannet Hallar, A., McCubbin, I., David, R., Borys, R., Blossey, P., Muhlbauer, A., Kuang, Z. and Moore, M.: Isotopic fractionation in wintertime orographic clouds, *J. Atm. Ocean. Tech.*, 33, 2663-2678, 2016.

Deleted: .

Moved (insertion) [24]

Moved down [26]: *Atm.*

Moved down [27]: *Chem.*

Deleted: Lachlan-Cope, T., Listowski, C. and O'Shea, S.: The microphysics of clouds over the Antarctic Peninsula-Part 1: Observations,

Deleted: *Phys.16*, 15605-15617, 2016.

Landais, A., Steen-Larsen, H. C., Guillevic, M., Masson-Delmotte, V., Vinther, B. and Winkler, R.: Triple isotopic composition of oxygen in surface snow and water vapor at NEEM (Greenland), *Geochim. Cosmochim. Acta*, 77, 304-316, 2012.

Deleted: [Data set].

Deleted: .

Deleted: [Data set]. *Zenodo. *https://doi.org/10.5281/zenodo.3689566*, 2020.*

Moved down [28]: J.,

Deleted: Listowski, C., Delanoë,

Deleted: Kirchgaessner, A., Lachlan-Cope, T. and King, J.: Antarctic clouds, supercooled liquid water and mixed phase, investigated with DARDAR: geographical and seasonal variations, *Atm. Chem. Phys.*, 19, 6771-6808, 2019.

Deleted: 49

49 Macklin, W. C. and Payne, G. S.: A theoretical study of the ice accretion process, Quart. J. Royal Meteo. Soc., 93, 195-213,
50 1967.

51
52 Mahernndl, N., Moser, M., Lucke, J., Mech, M., Risse, N., Schirmacher, I. and Maahn, M.: Quantifying riming from airborne
53 data during the HALO-(AC)3 campaign, Atmos. Meas. Tech., 17, 1475-1495, 2024.

54
55 [Majoube, M. \(1971\). Fractionnement en oxygène-18 et en deutérium entre l'eau et sa vapeur. J. Chim. Phys., 68, 1423–1436.](#)

Moved (insertion) [25]

56
57 Masson-Delmotte, V., Landais, A., Stievenard, M., Cattani, O., Falourd, S., Jouzel, J., Johnsen S. J., White, J.W.C., Werner,
58 M., Sveinbjörnsdottir, A. and Fuhrer, K.: Holocene climatic changes in Greenland: Different deuterium excess signals at
59 Greenland Ice Core Project (GRIP) and NorthGRIP, J. Geophys. Res.: Atmospheres, 110-D14, 2005.

60
61 Masson-Delmotte, V., S. Hou, A. Ekaykin, J. Jouzel, A. Aristarain, R. T. Bernardo, D. Bromwich et al.: A review of
62 Antarctic surface snow isotopic composition: Observations, atmospheric circulation, and isotopic modeling, J. Clim., 21,
63 3359-3387, 2008.

64
65 [Matejka, T. J., Houze Jr, R. A., Hobbs, P. V.: Microphysics and dynamics of clouds associated with mesoscale rainbands in](#)
66 [extratropical cyclones. Quarterly J. Royal Met. Soc., 106, 29-56, 1980.](#)

Moved (insertion) [28]

67
68 Matrosov, S. Y.: Frozen hydrometeor terminal fall velocity dependence on particle habit and riming as observed by
69 vertically pointing radars, J. App. Meteo. Clim., 62, 1023-1038, 2023.

70
71 [Maturilli, M., M. Kayser \(2016\): Homogenized radiosonde record at station Ny-Ålesund, Spitsbergen in 2014 \[dataset\].](#)
72 [PANGAEA, https://doi.org/10.1594/PANGAEA.845338, In supplement to: Maturilli, M; Kayser, M \(2016\): Arctic](#)
73 [warming, moisture increase and circulation changes observed in the Ny-Ålesund homogenized radiosonde record.](#)
74 [Theoretical and Applied Climatology, 17 pp, https://doi.org/10.1007/s00704-016-1864-0](#)

Deleted: McIlhattan, E. A., Pettersen, C., Wood, N. B. and L'Ecuyer, T. S.: Satellite observations of snowfall regimes over the Greenland Ice Sheet, The Cryosph., 14, 4379-4404, 2020.

75
76 [Maturilli, M.; M. Kayser \(2017\): Homogenized radiosonde record at station Ny-Ålesund, Spitsbergen, 2015-2016 \[dataset](#)
77 [publication series\]. PANGAEA, https://doi.org/10.1594/PANGAEA.875196](#)

78
79 [Maturilli, M; E. Dünschede \(2023\): Homogenized radiosonde record at station Ny-Ålesund, Spitsbergen, 2017-2022 \[dataset](#)
80 [publication series\]. PANGAEA, https://doi.org/10.1594/PANGAEA.961203](#)

81

Deleted: 50

85 Mellat, M., Bailey, H., Mustonen, K.-R., Marttila, H., Klein, E. S., Gribanov, K., Bret-Harte M. S., et al.: Hydroclimatic
86 controls on the isotopic ($\delta^{18}\text{O}$, $\delta^2\text{H}$, d-excess) traits of pan-Arctic summer rainfall events, *Front. Earth Sci.*, 9, 651731, 2021,
87 [\[dataset\]](#).
88
89 [Merlivat, L.: Molecular diffusivities of \$\text{H}_2^{16}\text{O}\$, \$\text{HD}^{16}\text{O}\$ and \$\text{H}_2^{18}\text{O}\$ in gases, *J. Chem. Phys.*, 69, 2864–2871, 1978.](#)
90
91 Merlivat, L. and Jouzel, J.: Global climatic interpretation of the deuterium-oxygen 18 relationship for precipitation, *J.*
92 *Geophys. Res.: Oceans*, 84-C8, 5029-5033, 1979.
93
94 [Merlivat, L., and Nief, G.: Fractionnement isotopique lors des changements d'états solide–vapeur et liquide–vapeur de l'eau.](#)
95 [Tellus](#), 19, 122–127, 1967.
96
97 Miller, N. B., Shupe, M. D., Cox, C. J., Walden, V. P., Turner, D. D. and Steffen, K.: Cloud radiative forcing at Summit,
98 Greenland, *J. Clim.*, 28, 6267-6280, 2015.
99
00 Mitchell, D. L., Zhang, R. and Pitter, R. L.: Mass-dimensional relationships for ice particles and the influence of riming on
01 snowfall rates, *J. App. Meteo. Clim.*, 29, 153-163, 1990.
02
03 Moerman, J. W., Cobb, K. M., Adkins, J. F., Sodemann, H., Clark, B. and Tuen, A. A.: Diurnal to interannual rainfall $\delta^{18}\text{O}$
04 variations in northern Borneo driven by regional hydrology, *Earth Planet. Sci. Lett.*, 369, 108-119, 2013.
05
06 Moisseev, D., von Lerber, A. and Tiira, J.: Quantifying the effect of riming on snowfall using ground-based observations, *J.*
07 *Geophys. Res.: Atmospheres*, 122, 4019-4037, 2017.
08
09 [Mosimann, L.: An improved method for determining the degree of snow crystal riming by vertical Doppler radar, *Atm. Res.*,](#)
10 [37, 305-323, 1995.](#)
11
12 [Mülmenstädt, J., Sourdeval, O., Delanoë, J. and Quaas, J.: Frequency of occurrence of rain from liquid-, mixed-, and ice-](#)
13 [phase clouds derived from A-Train satellite retrievals, *Geophys. Res. Lett.*, 42, 6502-6509, 2015.](#)
14
15 [NCPOR – National Center for Polar and Ocean Research, India: MRR data from Himadri Station, last accessed 8 May 2026](#)
16 [\(<https://npdc.ncpor.res.in/>\) \[dataset\]](#).
17

Deleted: .

Moved (insertion) [27]

Moved down [29]: J.

Deleted: Morrison, H., van Lier-Walqui, M., Fridlind, A.M., Grabowski, W.W., Harrington, J.Y., Hoose, C., et al.: Confronting the challenge of modeling cloud and precipitation microphysics,

Deleted: Adv. Modeling Earth Sys., 12, e2019MS001689, 2020.

Moved down [30]: J.

Deleted: Mosimann, L., Weingartner, E. and Waldvogel, A.: An analysis of accreted drop sizes and mass on rimed snow crystals,

Deleted: Atm. Sci., 51, 1548-1558, 1994.

Deleted: 51

30 Nusbaumer, J., Wong, T. E., Bardeen, C. and Noone, D.: Evaluating hydrological processes in the Community Atmosphere
31 Model Version 5 (CAM5) using stable isotope ratios of water, *J. Adv. Modeling Earth Sys.*, 9, 949-977, 2017.

32

33 Ohtake, T.: Observations of size distributions of hydrometeors through the melting layer, *J. Atm. Sci.*, 26, 545-557, 1969.

34

35 Ohtake, T.: Atmospheric ice crystals at the South Pole in summer, *Antarct. J.*, 13, 174-175, 1978.

36

37 Orr, B. W. and Kropfli, R. A.: A method for estimating particle fall velocities from vertically pointing Doppler radar, *J. Atm.*
38 *Oceanic Tech.*, 16, 29-37, 1999.

39

40 Osterberg, E. C., Hawley, R. L., Wong, G., Kopec, B., Ferris, D. and Howley, J.: Coastal ice-core record of recent northwest
41 Greenland temperature and sea-ice concentration, *J. Glaciology*, 61, 1137-1146, 2015.

42

43 Pettersen, C., Bennartz, R., Merrelli, A. J., Shupe, M. D., Turner, D. D. and Walden, von P.: Precipitation regimes over
44 central Greenland inferred from 5 years of ICECAPS observations, *Atm. Chem. Phys.*, 18, 4715-4735, 2018.

45

46 Pfahl, S. and Sodemann, H.: What controls deuterium excess in global precipitation? *Climate of the Past* 10, 771-781, 2014.

47

48 Praz, C., Roulet Y.-A. and Berne, A.: Solid hydrometeor classification and riming degree estimation from pictures collected
49 with a Multi-Angle Snowflake Camera, *Atm. Meas. Tech.*, 10, 1335-1357, 2017.

50

51 Protat, A. and Williams, C. R.: The accuracy of radar estimates of ice terminal fall speed from vertically pointing Doppler
52 radar measurements, *J. App. Meteo. Clim.*, 50, 2120-2138, 2011.

53

54 Pruppacher, H. R. and Klett, J. D.: Microphysics of clouds and precipitation, Springer New York, DOI 10.1007/978-0-306-
55 48100-0, 2010.

56

57 Santos, V. dos, Durán-Quesada, A. M. Sánchez-Murillo, R. and Gastmans, D.: High-frequency isotope compositions reveal
58 different cloud-top and vertical stratiform rainfall structures in the inland tropics of Brazil, *Geophys. Res. Lett.*, 51,
59 e2024GL109886, 2024, [dataset].

60

61 Sarchilli, C., Ciardini, V., Grigioni, P., Iaccarino, A., De Silvestri, L., Proposito, M., Dolci, S., et al.: Characterization of
62 snowfall estimated by in situ and ground-based remote-sensing observations at Terra Nova Bay, Victoria Land, Antarctica, *J.*
63 *Glaciology*, 66, 1006-1023, 2020.

Deleted: Rasmussen, S. O., Dahl-Jensen, D., Fischer, F., Fuhrer, K., Hansen, S. B., Hansson, M., Hvidberg, C. S. et al.: Ice-core data used for the construction of the Greenland Ice-Core Chronology 2005 and 2021 (GICC05 and GICC21), *Earth Sys. Sci. Data Discuss.*, 15, 3351–33641, 2023.

Ricaud, P., Durand, P., Grigioni, P., Del Guasta, M., Camporeale, G., Roy, A., Attié, J.-L. and Bognar, J.: In situ observations of supercooled liquid water clouds over Dome C, Antarctica, by balloon-borne sondes, *Atm. Meas. Tech.*, 17, 5071-5089, 2024.

Deleted: .,

Deleted: .

Deleted: 52

77
78
79
80
81
82
83
84
85
86
87
88
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90
91
92
93
94
95
96
97
98
99
00
01
02
03
04
05
06
07
08
09

Schumacher, C., R. A. Houze Jr.: The TRMM precipitation radar's view of shallow, isolated rain *J. Applied Meteo.* 42, 1519-1524, 2003.

Seidl, A. W., Johannessen, A., Dekhtyareva, A., Huss, J. M., Jonassen, M. O., Schulz, A., Hermansen, O., Thomas, C. K. and Sodemann, H.: The ISLAS2020 field campaign: Studying the near-surface exchange process of stable water isotopes during the arctic wintertime, *Earth Sys. Sci. Data: Discussions*, 2024, [dataset].

Shupe, M. D., Turner, D. D., Walden, von P., Bennartz, R., Cadeddu, M. P., Castellani, B. B., Cox, C. J., et al.: High and dry: New observations of tropospheric and cloud properties above the Greenland Ice Sheet, *Bull. Amer. Meteo. Soc.*, 94, 169-186, 2013.

Shupe, M.D., Kollias, P., Poellot, M. and Eloranta, E.: On Deriving Vertical Air Motions from Cloud Radar Doppler Spectra. *J. Atm. Ocean Tech.*, 25, 547-557, DOI: <https://doi.org/10.1175/2007JTECHA1007.1>, 2008.

Stewart, M. K.: Stable isotope fractionation due to evaporation and isotopic exchange of falling waterdrops: Applications to atmospheric processes and evaporation of lakes *J. Geophys. Res.*, 80, 1133-1146, 1975.

Takahashi, T. and Fukuta, N.: Supercooled cloud tunnel studies on the growth of snow crystals between -4 and -20°C, *J. Met. Soc. Japan*, 66, 841-855, 1988.

Tridon, F., Silber, I., Battaglia, A., Kneifel, S., Fridlind, A., Kalogeras, P. and Dhillon, R.: Highly supercooled riming and unusual triple-frequency radar signatures over McMurdo Station, Antarctica, *Atm. Chem. Phys.*, 22, 12467-12491, 2022.

Uemura, R., Matsui, Y., Yoshida, N., Abe, O. and Mochizuki, S.: Isotopic fractionation of water during snow formation: Experimental evidence of kinetic effect, *Polar Meteo. Glaciology*, 19, 1-14, 2005.

Wang, J., Pang, H., Wu, S., Schoenemann, S. W., Uemura, R., Ekaykin, A., Werner, M., et al.: The Ant-Iso dataset: a compilation of Antarctic surface snow isotopic observations, *Earth Sys. Sci. Data: Discussions*, 2022.

Warburton, J. A.: Atmospheric processes and the chemistry of snow on the Ross Ice Shelf, Antarctica, *J. Glaciology*, 20, 149-162, 1978.

Moved (insertion) [29]

Deleted: Schlosser, E., Dittmann, A., Stenni, B., Powers, J. G., Manning, K. W., Masson-Delmotte, V. and Cagnati, V. M.: An., Grigioni, P., Scarchilli, C., The influence of the synoptic regime on stable water isotopes in precipitation at Dome C, East Antarctica, *The Cryosphere*, 11, 2345-2361, 2017.

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Deleted: <https://doi.org/10.1175/2007JTECHA1007.1>

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Deleted: Tian, C., Wang, L., Kaseke, K. F. and Bird, B. W.: Stable isotope compositions (δ^2H , $\delta^{18}O$ and $\delta^{17}O$) of rainfall and snowfall in the central United States. *Sci. Rep.* 8, 6712, 2018.

Deleted: von Lerber, A., Moiseev, D., Bliven, L. F., Petersen, W., Harri, A.-M. and Chandrasekar, V.: Microphysical properties of snow and their link to Z-e-S relations during BAECC 2014, *J. App. Meteo. Clim.*, 56, 1561-1582, 2017.

Deleted: 53

29 [Warburton, J. A., deFelice, T. P.: Oxygen isotopic composition of central Sierra Nevada precipitation, I. Identification of ice-](#)
30 [phase water capture regions in winter storms, *Atm. Res.*, 20, 11-22, 1986.](#)

31 Weiss, R. R., Locatelli, J. D. and Hobbs, P. V.: Deduction of ice particle types in the vicinity of the melting layer from
32 Doppler radar measurements, *J. App. Meteo. Clim.*, 16, 314-316, 1977.

33

34 Weiss, R. R. and Hobbs, P. V.: The use of a vertically pointing pulsed Doppler radar in cloud physics and weather
35 modification studies, *J. App. Meteo. Clim.*, 14, 222-231, 1975.

36

37 White, A. B., Gottas, D. J., Strem, E. T., Ralph, F. M. and Neiman, P. J.: An automated bright band height detection
38 algorithm for use with Doppler radar spectral moments, *J. Atm. Oceanic Tech.*, 19, 687-697, 2002.

39

40 White, A. B., Neiman, P. J., Ralph, F. M., Kingsmill, D. E. and Persson, P. O. G.: Coastal orographic rainfall processes
41 observed by radar during the California Land-Falling Jets Experiment, *J. Hydrometeo.*, 4, 264-282, 2003.

42

43 Wiener, V., Roussel, M.-L., Genthon, C., Vignon, É., Grazioli, J. and Berne A.: A 7-year record of vertical profiles of radar
44 measurements and precipitation estimates at Dumont d'Urville, Adélie Land, East Antarctica, *Earth Sys. Sci. Data*, 16 821-
45 834, 2024, [\[dataset\]](#).

46

47 Xie, X., Evaristo, R., Troemel, S., Saavedra, P., Simmer, C. and Ryzhkov A.: Radar observation of evaporation and
48 implications for quantitative precipitation and cooling rate estimation, *J. Atm. Oceanic Tech.*, 33, 1779-1792, 2016.

49

50 Zawadzki, I., Szyrmer, W., Bell, C. and Fabry, F.: Modeling of the melting layer. Part III: The density effect, *J. Atm. Sci.*,
51 62, 3705-3723, 2005.

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