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Salt deliquescence along boulder cracks in the Antarctic Dry Valleys: An overlooked source of moisture for rock weathering processes

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ABSTRACT

Cracking is a primary rock-weathering mechanism in arid environments, where dry conditions typically limit the efficacy of water-driven weathering processes. Here, we present results from a field-based experiment in the hyper-arid and frigid Antarctic Dry Valleys (ADV) that documented recurring periods of transient accumulation of liquid water along rock cracks during otherwise dry conditions. This moisture was likely sourced from the deliquescence of hygroscopic salts during sub-saturated humidity conditions. Analysis of meteorological data from 17 stations scattered throughout the ADV revealed that near-surface atmospheric conditions across one of Earth's driest environments can annually support tens of deliquescence-efflorescence cycles of hygroscopic salts, such as, $CaCl_2$, $NaNO_3$, NaCl, and $MgCl_2$. This deliquesced moisture may have an important role in the cracking processes of ADV rocks. In a broader context, the results from the ADV suggest that deliquesced atmospheric humidity may be an overlooked source of moisture available for rock weathering processes in otherwise extremely dry deserts on Earth and possibly Mars.

1. Introduction

1.1. Rock weathering in hyper-arid environments

Rock weathering is broadly regarded as a key and often rate-limiting process in the subsequent evolution of terrestrial landscapes. In hyper-arid environments, physical disintegration, i.e., the breakup of rocks through cracking, is typically a dominant mode of weathering because the dry conditions often limit the efficacy of water-dependent chemical, biological, and frost weathering mechanisms (Cooke and Smalley, 1968; Cooke, 1981). Cracking of rocks or other brittle materials can also occur when subjected to low subcritical stresses (Atkinson, 1984; Anderson, 2005). In arid climates natural stresses are likely dominantly subcritical in magnitude and are commonly attributed to repeated cycles of thermal expansion/contraction in response to diurnal insolation dynamics (e.g., McFadden et al., 2005) or salt weathering (Winkler and Wilhelm, 1970; Sperling and Cooke, 1985; Steiger et al., 2008; Desarnaud et al., 2016). Nonetheless, laboratory as well as field-based studies, have shown that even a slight increase in moisture can significantly increase the rates at which these otherwise 'dry' rock-cracking mechanisms can operate (Waza et al., 1980; Meredith and Atkinson, 1985; Yoshitaka Nara et al., 2010, 2012; Eppes and Keanini, 2017; Eppes et al., 2020). Here, we present field-based evidence from one of Earth's driest and coldest deserts that the deliquescence of atmospheric humidity by hygroscopic salts is an effective pathway for water delivery to rock cracks. This moisture delivery pathway to rock cracks may have an important and previously overlooked pace-setting role in the cracking process of rocks in hyper-arid environments.

1.2. The Antarctic Dry Valleys

The Antarctic Dry Valleys (ADV) (Fig. 1) are amongst the coldest and

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driest 'ice-free' regions on Earth (Doran et al., 2002; Fountain et al., 2010; Obryk et al., 2020). The mean annual air temperatures on the ADV floors range between −15 °C and − 30 °C, depending on the location (Obryk et al., 2020), and precipitation is limited to <50 mm/yr, which occurs primarily as snowfall (Fountain et al., 2010). These hyperarid and frigid conditions have prevailed in the ADV since the Plio-Pleistocene (Fielding et al., 2011; Scopelliti et al., 2013) and resulting in one of the slowest eroding landscapes on Earth with estimated bedrock erosion rates below 1 m/m.y. (Brook et al., 1995; Sugden et al., 1999; Summerfield et al., 1999; Margerison et al., 2005; Staiger et al., 2006; Balco and Shuster, 2009; Putkonen et al., 2014; Marrero et al., 2018). As such, the ADV environment is also regarded as a prime analog site for the present-day hyperarid and cold surface conditions on Mars (Sletten et al., 2003; Tamppari et al., 2012; Head and Marchant, 2014). Rock weathering processes in the ADV have been previously attributed to thermal stress mechanisms (Campbell and Claridge, 1987; Hall, 1999; Lamp et al., 2017) or to hygroscopic salts (Wellman and Wilson, 1965; Selby and Wilson, 1971; Johnston, 1973; Campbell and Claridge, 1987), that accumulate at or near the ADV surface due to the extremely dry conditions (Keys, 1979; Keys and Williams, 1981; Bisson et al., 2015;). The presence of up to ~30 salt phases was previously reported in the ADV soils, including hygroscopic salts, such as NaCl, $MgCl_2$, $NaNO_3$, and $CaCl_2$ (Claridge and Campbell, 1977; Goudie and Cooke, 1984; Keys, 1979; Wilson, 1979; Keys and Williams, 1981; Miotke and von Hodenberg, 1983; Tamppari et al., 2012; Bisson et al., 2015). The origin of salts in the ADV is the subject of numerous studies, with the three primary sources believed to be marine aerosols (Claridge and Campbell, 1968, 1977; Keys and Williams, 1981), chemical weathering (Claridge and Campbell, 1977; Keys and Williams, 1981; Cuozzo et al., 2020) and oxidized nitrogen and sulfur compounds deposited on the East Antarctic Ice Sheet (Bao et al., 2000, 2008; Michalski, 2005). Wind is the most likely transport mechanism of salts in this hyperarid environment of the ADV (Witherow et al., 2006; Bao et al., 2008; Campbell and Sheppard, 2022) and salts are deposited on the surface by dry deposition or precipitation.

1.3. Salt deliquescence in the ADV

Deliquescence occurs when the relative humidity (RH) of the air mass exceeds the deliquescence relative humidity (DRH) of a specific salt or a salt mixture, and atmospheric water is absorbed and forms a brine that can further adsorb water. Efflorescence is the reverse process that occurs when relative humidity is reduced below the efflorescence relative humidity (ERH) and recrystallization occurs. Salt deliquescence/efflorescence dynamics have been previously documented in the ADV soils as the appearance of transient 'wet patches' (Harris and Cartwright, 1981; Head et al., 2007; Gough et al., 2016; Levy, 2021; Toner et al., 2022) or 'wet slope streaks' (Toner et al., 2022) during events of increased atmospheric humidity. The present study tests whether and how such deliquescence/efflorescence dynamics can link to rock-cracking processes in the ADV.

2. Methods

2.1. Field experiment and laboratory analyses

A field-based 10-day experiment was performed using local meteorological measurements (Kestrel 5500), time-lapse photography (Brinno TLC 200), and thermal imaging (FLIR SC430) of a Ferrar Dolerite boulder with incipient cracks near Don Juan pond in Wright Valley (Figs. 1, 2). After the experiment, extraction of the boulders revealed light-toned salts along the rock cracks that were embedded in the soil during the experiment (Fig. 2b, c). Mineralogical and chemical analyses of the salts were performed at the Geological Survey of Israel. Salt taken from a crack in the boulder was dissolved in distilled water, and chemical analysis for major cations was conducted using inductively

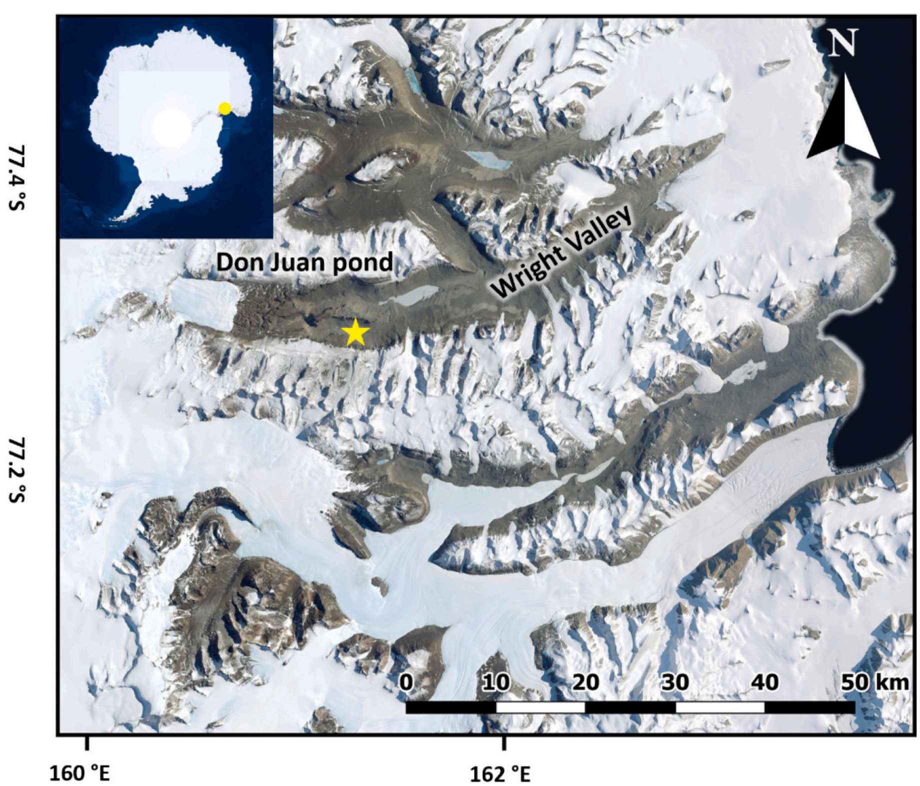

Fig. 1. Annotated satellite image of the Antarctic Dry Valleys. Yellow star marks the field site near Don Juan pond in Wright valley. The satellite image was obtained through the QuickMapServices QGIS plugin, from ESRI server (ArcGIS/World_Imagery). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

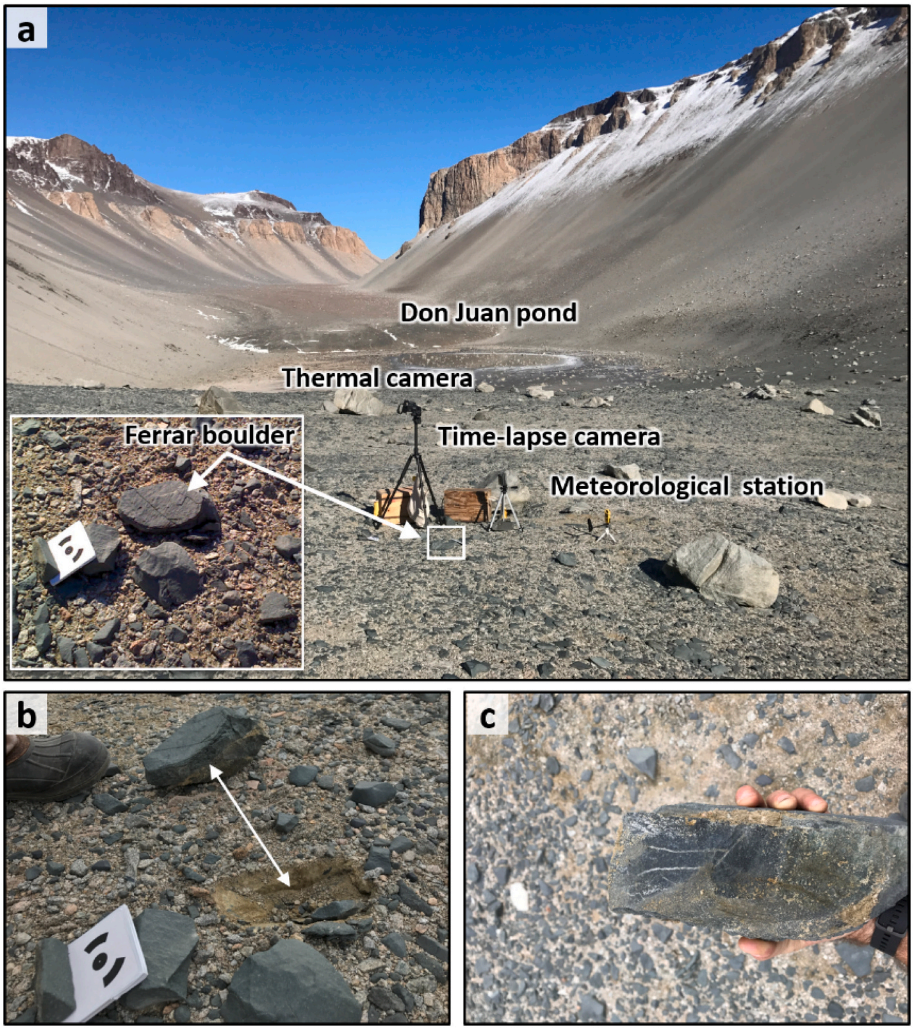

Fig. 2. a) Field experiment setup near Don Juan pond (in the background, west of the experiment site) in Wright Valley. White arrows mark the imaged boulder. The length of the boulder is 25 cm. Monitoring equipment included: a time-lapse optical camera used to detect wetting events, a mobile meteorological station, and a thermal camera. b) Image of the Ferrar dolerite boulder removed from the soil after the experiment (white arrow). c) The underside of the Ferrar dolerite boulder showing accumulation of salts in cracks.

coupled plasma-optical emission spectrometry (Perkin Elmer, Optima 5300) and major anions using ion chromatography (Dionex ICS-2000). Mineralogic analysis was performed using bulk X-ray diffraction. Mineral phase identification was performed using HighScore Plus® software based on the ICSD database.

2.2. Meteorological data

Meteorological data from 17 weather stations scattered throughout the ADV were used to examine the occurrence of deliquescence-efflorescence conditions for $NaNO_3$, $CaCl_2$, NaCl, and $MgCl_2$. The weather stations are part of the McMurdo Long Term Ecological Research Project (MCM LTER) in the ADV. Most stations (11 out of 17) have been operating for over 20 years at 1-h temporal recording resolution. Analysis of these data was conducted to quantify the occurrence of supra-DRH conditions for these salt phases in the ADV environment through time. Deliquescence conditions were defined as the durations in a year when RH values exceed the DRH of the specific salt phase. A Deliquescence/efflorescence cycle was defined as a period between the increase of RH above DRH to when RH decreases below DRH. A minimum duration threshold of 3 h and a minimum of 5 % excess humidity above DRH were used to filter out short events and those with marginal excess RH. The DRH value of $CaCl_2$ used in the analysis refers to the hexahydrate phase - $CaCl_2{\cdot}6H_2O$ ('Antarcticite'), which was first described in the ADV (Torii and Ossaka, 1965).

3. Results

3.1. Salt deliquescence during the field experiment

Time-lapse photography revealed an accumulation of moisture along cracks in the imaged boulder (identified visually by the darkening of the rock, Fig. 3a) during three discrete periods spanning between approximately 6–12 h each (Fig. 3a, supp. Time-lapse video). Since no precipitation was observed during the experiment, we can overrule snow melt as a potential source, although it is a recognized source of water in the ADV during the austral summers (Hagedorn et al., 2010; Liu et al., 2015). The three periods of moisture stability along the boulder cracks during the experiment coincided exclusively with RH values that exceeded 35– 40 % but did not reach the dew point and included sub-zero air temperatures (Fig. 3c, d). During these moisture accumulation events, RH values coincided with supra DRH conditions for chloride salts, such as $CaCl_2$ and $MgCl_2$ (Fig. 3b, d). Chemical analysis of the salt samples taken from a crack in the boulder shows that the major anions were Cl^-, SO_4^{-2}, (55 %, 45 %, respectively) and that the major cations are Ca^{+2}, Na^+, K^+, Mg^{+2}, SiO_2, and Sr^{+2} (71 %, 23 %, 3 %, 2 %,1 %, >1 % respectively). Excess of Cl^- and Ca^+ after accounting for the complete precipitation of halite and gypsum, which were the dominant salt phases found in X-ray diffraction, points to the presence of $CaCl_2$ and possibly other chlorides which were previously found in the ADV (Claridge and Campbell, 1977; Keys, 1979) and specifically in the Wright valley soils (Toner and Sletten, 2013; Toner et al., 2022) and the nearby Don Juan

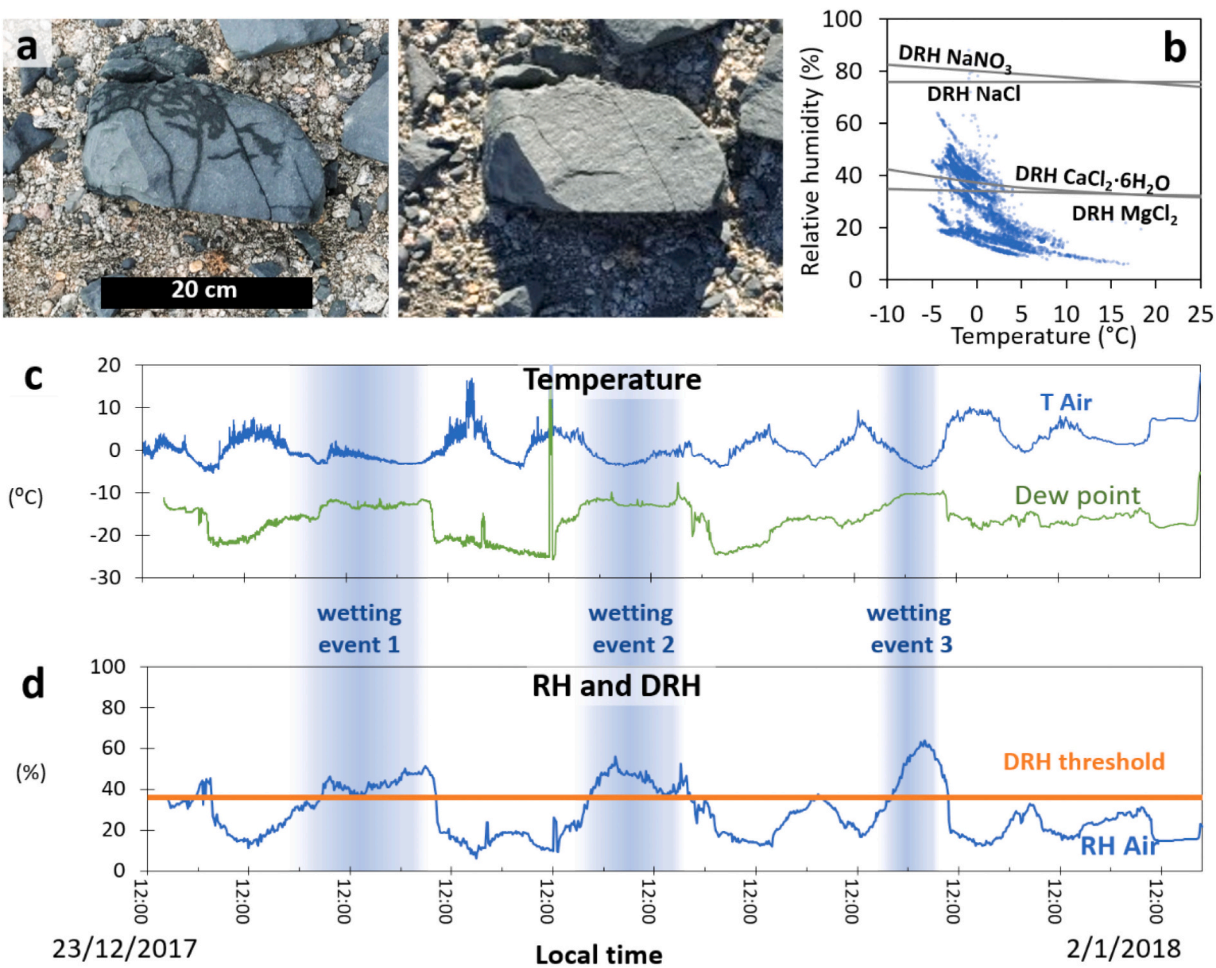

Fig. 3. Moisture delivery to rock cracks via salt deliquescence. a) The Ferrar dolerite boulder during (left) and between (right) wetting events. b) Data with 10-min resolution from field measurements of air temperature vs. relative humidity during the 10 days experiment. Gray lines indicate the DRH of $CaCl_2{\cdot}6H_2O$, $NaNO_3$, NaCl, $MgCl_2$ salts as a function of temperature. c) Ambient air temperature (T) (blue), and dew point (green) through time. Note that air T does not reach the dew point during observed wetting events and that wetting events (blue shading) persist through sub-zero temperatures. d) Relative humidity (blue) and the DRH threshold of 35 % ~ 40 % (orange). All three wetting events were initiated after supra-DRH conditions were achieved and ended when RH declined below the DRH threshold. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

pond – a saline lake rich in $CaCl_2$ (Dickson et al., 2013; Toner et al., 2017). Therefore, the most likely explanation for the observed wetting events in the experiment appears to be the deliquescence of such salts. Altogether moisture sourced from deliquesced atmospheric humidity was found to be stable along the boulder cracks for ~25 % of the otherwise 'dry' 10-day span of the experiment.

3.2. Analysis of meteorological data

Results from analysis of 17 meteorological stations in the ADV show that the conditions for deliquescence of the salt phases that were examined, i.e., $CaCl_2$, $NaNO_3$, NaCl, and $MgCl_2$, prevail on average for 69 % (range 43 %–85 %), 16 % (range 4 %–25 %), 32 % (range 10 %–51 %) and 83 % (range 43 %–85 %) of the year, respectively (Fig. 4, Figs. supp. S1–3). In addition, the conditions for discrete deliquescence events happen on average 55 (range 25–83), 30 (range 12–48), 50 (range 20–76), and 39 (range 15–36) times per year, respectively (Fig. 4, Figs. supp. S1–3). For simplicity, deliquescence conditions for single-phase salt-brine systems were assumed. However, laboratory experiments show that the DRH of salt mixtures is expected to be even lower than that of the same single salts (e.g., Yang et al., 2002). Thus, the duration of deliquescence conditions is potentially longer than calculated herein.

4. Discussion

The results of the field-based experiment show evidence that salt deliquescence is an effective mechanism for moisture delivery to the rock surface and cracks in the hyper-arid and frigid conditions of the ADV (Fig. 5). Deliquescence/efflorescence cycles such as those observed during the experiment (Fig. 3) are expected to promote rock cracking by 'salt shattering', which is regarded as an important weathering mechanism in arid soils (Amit et al., 1993; Goudie and Viles, 1997; Rodriguez-Navarro and Doehne, 1999; Goudie, 2013), including the ADV (Johnston, 1973) and Mars as well (Malin, 1974; Jagoutz, 2006). Salt shattering requires that the amount of water be low enough to limit leaching of the salts from the surface/soils, and yet sufficient for salt-water interactions that support cycles of dissolution and crystallization of salts, which can exert local stress when confined within rock pores or fractures (Sperling and Cooke, 1985; Amit et al., 1993; Desarnaud et al., 2016). In addition, recent studies have demonstrated that the rates of mechanical weathering processes, such as those induced by salt hydration or cyclic thermal stress-loading (Richter and Simmons, 1974; McFadden et al., 2005; Viles et al., 2010; Lamp et al., 2017), may accelerate by orders of magnitude in the presence of small amounts of moisture (Eppes and Keanini, 2017; Eppes et al., 2020). This acceleration is associated with the weakening of bonds by water molecules at the tip of cracks that

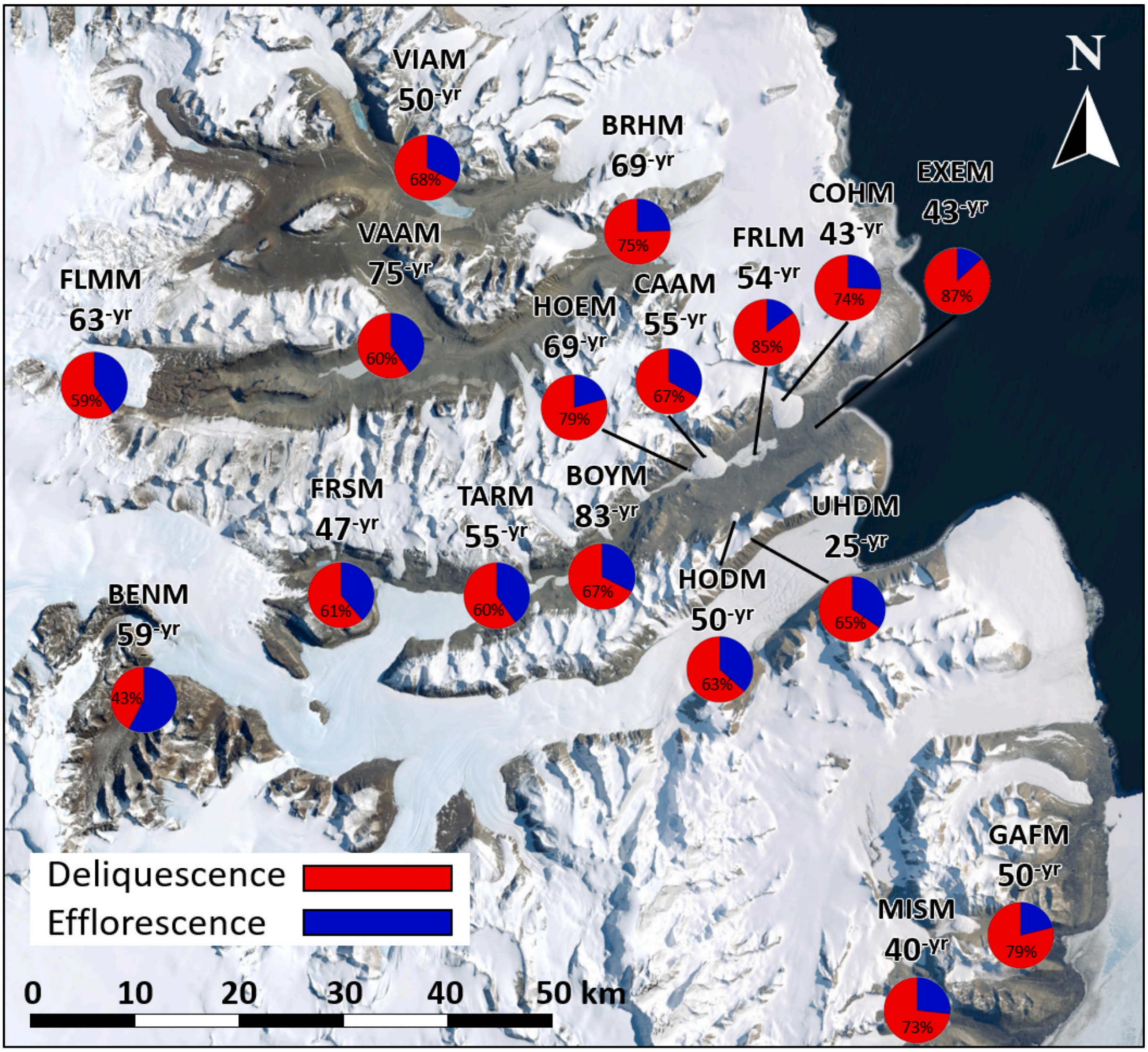

Fig. 4. Deliquescence conditions of $CaCl_2$ in 17 meteorological stations in the ADV. The pie plots show the time fraction of deliquescence conditions (red). The number of estimated deliquescence-efflorescence cycles per year is marked below the station name. A full description of the stations can be found at https://mcm.lternet.edu/meteorology-data-sets#met-15. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

propagate slowly in response to subcritical stresses (Atkinson, 1984; Meredith and Atkinson, 1985; Nara and Kaneko, 2006; Voigtländer et al., 2018; Eppes et al., 2020). The delivery of moisture to the tips of cracks, via salt deliquescence, can thus also accelerate crack propagation under external subcritical stress.

The analysis of data obtained from the permanent ADV meteorological stations (Fig. 4) shows that the atmospheric conditions that enabled the deliquescence events near Don Juan pond are prevalent throughout the ADV and therefore suggest that deliquesced moisture during otherwise 'dry' conditions may be an important moisture delivery pathway for rock weathering throughout the ADV. The immediate area of Don Juan pond features anomalously high concentrations of deliquescent salts (Torii and Ossaka, 1965; Dickson et al., 2013; Toner and Sletten, 2013; Gough et al., 2016; Toner et al., 2017) and therefore excludes straightforward extrapolation of the local experimental results (Fig. 3) to the entire ADV landscape. However, observations of soluble salts accumulation in soils and on rock surfaces throughout the ADV (though most likely in lower concentrations), suggests that deliquescence can have an important role in delivering moisture for rock-weathering processes throughout the ADV landscape. We thus suggest that salt deliquescence may play a dual role in the acceleration of rock cracking in arid regions. The first as a source of stress loading, where cycles of salt dissolution and crystallization can exert local stress when confined within rock pores or fractures (Sperling and Cooke, 1985; Amit et al., 1993; Desarnaud et al., 2016). The second is the acceleration of subcritical rock cracking in the presence of even small amounts of water at the tip of cracks (Waza et al., 1980; Meredith and Atkinson, 1985; Yoshitaka Nara et al., 2010, 2012; Eppes and Keanini, 2017; Eppes et al., 2020).

Laboratory experiments show that ERH can be lower than DRH for a given salt and temperature due to a kinetic barrier for the nucleation of a crystalline phase (Martin, 2000; Gough et al., 2016). In our experiment, there is no evidence for the reported hysteresis between DRH and ERH. This could be because of the heterogeneity and impurity of natural brines that can readily facilitate the nucleation of salt crystals. Furthermore, the onset and termination of the deliquescence events that were documented during the field experiment were not driven by diurnal oscillations in air temperature and resulting changes in RH and instead appeared to be more closely associated with pulses of increased vapor pressure (Fig. Supp. S4).

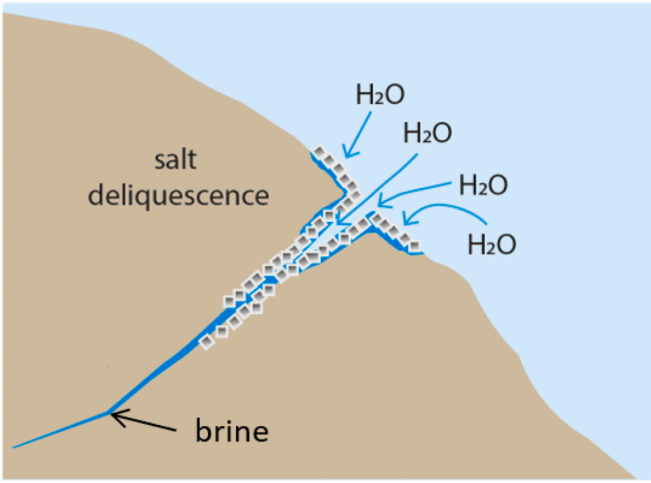

Fig. 5. Illustration of moisture delivery into rock cracks by salt deliquescence. Deliquesced moisture (brine) within rock cracks can accelerate the propagation of cracks by weakening rock chemical bonds by water molecules at the tip of cracks that propagate slowly in response to subcritical stresses. Such stresses can originate from cycles of salt crystallization (deliquescence/efflorescence cycles) and/or thermal expansion/contraction in response to diurnal insolation dynamics.

We propose that deliquescence/efflorescence cycles may be an important driver of rock weathering in the ADV and potentially other hyper-arid regions where an accumulation of hygroscopic salts is observed, such as the Atacama (Davila et al., 2010), Negev (Amit et al., 1993) and Namib (Goudie et al., 1997; Goudie, 1998) deserts (Fig. 5). This may also apply to the surface of Mars, where the presence of salts and deliquescence/efflorescence conditions was previously suggested (Davila et al., 2010; Fischer et al., 2014; Toner et al., 2015; Gough et al., 2016, 2019).

5. Conclusion

Based on our results from a field experiment that show discrete wetting events of a rock surface in sub-saturated air and sub-freezing conditions, following an increase in air RH, we conclude that salt deliquescence may be an effective and overlooked mechanism of water delivery into rock cracks in hyper-arid and cold conditions on Earth and possibly on Mars. Analysis of data from 17 permanent meteorological stations shows that conditions for the deliquescence of several salt phases found in the ADV prevail throughout the region and suggests that such moisture delivery may be widespread in the ADV. Deliquescence/efflorescence cycles can potentially contribute to rock cracking both by facilitating additional stress loading related to salt expansion during crystallization or hydration and also by providing water for accelerating subcritical crack propagation driven by any source of stress.

Supplementary data to this article can be found online at https://doi.org/10.1016/j.geomorph.2023.108800.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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