LETTER

Thawing permafrost: an overlooked source of seeds for Arctic cloud formation

Jessie M Creamean1, Thomas C J Hill1, Paul J DeMott1, Jun Uetake1, Sonia Kreidenweis1 and Thomas A Douglas2

1 Department of Atmospheric Science, Colorado State University, Fort Collins, Colorado, United States of America
2 U.S. Army Cold Regions Research and Engineering Laboratory, Fort Wainwright, Alaska, United States of America

E-mail: jessie.creamean@colostate.edu

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Abstract

As the Arctic warms at twice the global rate, radiative feedbacks from clouds will lead to compounding impacts on the surface energy budget that affect both regional and global weather, and climate. In a future warmer world, the Arctic is projected to become cloudier. However, the formation and evolution of Arctic clouds remain highly uncertain in part due to a limited understanding of current and future sources of ice nucleating particles (INPs). In particular, the sources and abundance of biologically-derived INPs are poorly characterized, yet they may be pivotal for cloud ice formation, especially at temperatures in which Arctic mixed-phase clouds (AMPCs) persist (i.e. >−15 °C). Here, we show for the first time that permafrost is a remarkably rich source of biologically-derived INPs, both heat labile (probably proteinaceous) and other organic INPs of biomolecular origin (41%–100% and 99%–100% of the total INPs, respectively). INP concentrations in 1000 to 30,000 year old permafrost were comparable to the most active of other Arctic and midlatitude soil sources (up to 1010 INPs per gram of soil). Thawing of permafrost—which promotes metabolic activity in microbes—and subsequent mobilization of those soils directly into the atmosphere or into lakes, rivers, and the ocean, suggests the intriguing possibility that increasing emissions of INPs from this hitherto overlooked reservoir could be widespread, and, in time, greatly impact Arctic cloud cloud glaciation and radiative properties. This discovery is timely given the rapidly-thawing permafrost in Alaska and across Earth’s high latitudes. Since permafrost covers 15% of Northern Hemisphere land, this novel and prevalent INP source may become central to predictions of aerosol-cloud-precipitation interactions in AMPCs.

1. Introduction

Arctic clouds have profound effects on regional and global energy budgets, and hence climate, with cloud phase (i.e. liquid or ice) being a key modulator of their interactions with radiation (Intrieri et al 2002). Arctic mixed-phase clouds (AMPCs) are prevalent as a key component of the coupled ocean-ice-atmosphere system that modulates the delicate energy balance over frozen surfaces (Shupe 2011a). Such clouds can form within a few hours or less, but then persist for days to weeks (Morrison et al 2012). In addition to atmospheric and ocean dynamical processes, the ability to predict sea ice extent and annual cycle depends on the numerical representation of AMPC microphysical processes, which are inadequately understood (Hunke et al 2010, Morrison et al 2012). Modelling such processes and, hence, the AMPC annual cycle remains a significant challenge due to substantial model biases (Boucher et al 2013, Kay et al 2016, Taylor et al 2019). The ability to accurately represent the sea ice energy balance also has implications for Arctic ecological processes, socioeconomics among communities living in the Arctic and sub-Arctic, industrial dependencies on shipping and resource extraction, and weather teleconnections with midlatitudes (Jeffries et al 2013).

AMPC ice formation, properties, and lifetime are highly sensitive to the quantity and characteristics of aerosols that serve as ice nucleating particles (INPs)
(Morrison et al 2005, Solomon et al 2015, 2018, Kalesse et al 2016, Fridlind and Ackerman 2018). For example, Solomon et al (2018) demonstrated that a doubling of INP concentrations initially specified at 1.3 $1^{-1}$ can increase ice water path by approximately 1.7 times and decrease liquid water path by 1.5 times in AMPCs in Northern Alaska. They concluded that cloud microphysical and radiative properties were more sensitive to perturbations in INP compared to cloud condensation nuclei (CCN) concentrations. The efficacy of a particle to serve as an INP largely depends on its composition, morphology, and size, and thus, its source (Hoose and Möhler 2012).

Mineral dust and biologically-derived aerosols (e.g. intact or cell-free components of specific species of bacteria and fungi, pollen, lichens, algae, diatoms, soil organic matter, and macromolecules) are the most important INPs found in the atmosphere (Conen et al 2011, García et al 2012, Murray et al 2012, Creamean et al 2013, 2019, O’Sullivan et al 2015, Hill et al 2016). Classes of biologically-derived aerosols such as certain bacteria are capable of initiating freezing up to $-1.5$ °C (Vali et al 1976, Despres et al 2012, Murray et al 2012, Frohlich-Nowoisky et al 2016), while pure water will not freeze homogeneously until $-38$ °C.

Previous studies have reported both marine and terrestrial sources impacting Arctic INP populations, some of which indicate the potentially paramount role of biologically-derived INPs. Work by Bigg and colleagues was among the first to elucidate the role of the ocean as a microbial source of Arctic INPs (Bigg and Leck 2001, 2008, Leck and Bigg 2005, Bigg 2011). Specifically, they suggested marine bacteria and fragments of organisms from bubble bursting in open water regions (e.g. leads) were responsible for the sources of INPs they observed in the high Arctic air during their summertime measurements. Recent work reporting open ocean bulk seawater, aerosol, and/or surface microlayer measurements from Creamean et al (2019), Irish et al (2019a, 2019b), Wilson et al (2015), and Zeppenfeld et al (2019) demonstrate how the Arctic Ocean can be a rich source of INPs from marine biological processes, especially from features such as phytoplankton blooms (i.e. Creamean et al 2019, Zeppenfeld et al 2019). Measurements at Arctic coastal locations reported by Creamean et al (2018), Sauti-Temkiv et al (2019), Si et al (2018), and Wex et al (2019) demonstrate that both marine and terrestrial sources impact Arctic INP populations and were thought to be, at least partially, of biological origin. Recently, glacial outwash and ice coring sampling efforts in have elucidated the role of INPs from biological origin and their contribution to airborne Arctic INP populations (Hartmann et al 2019, Tobo et al 2019).

These examples demonstrate the limited body of direct field observations of biologically-derived Arctic INPs, which in general, are rare as compared to the host of work at lower latitudes. Conclusions about the role of biologically-derived particles in ice nucleation are equivocal based on results from climate modelling (Phillips et al 2009, Hooge et al 2010a, Sesartic et al 2012, Burrows et al 2013, Vergara-Temprado et al 2017). Because AMPC temperatures are often $\geq -15$ °C (Shupe et al 2006, Shupe 2011a, Shupe et al 2011b), biologically-derived INPs, which predominate over mineral at warmer than about $-20$ to $-15$ °C (Hartmann et al 2019 and references therein), could play a critical role in cloud formation for much of the Arctic annual cycle. With projected warming, evaluating the role of emissions of INPs from biological processes on cloud formation is essential to understanding the delicate and dynamic Arctic climate.

As Arctic air temperatures rise, permafrost—earth material (vegetation, peat, mineral soil, bedrock) that remains frozen for multiple years—is thawing and degrading (Rowland et al 2010, Slater and Lawrence 2013, Liliedahl et al 2016, Plaza et al 2019). Permafrost extending into the Earth’s surface is capped by a seasonally frozen and thawed zone (the ‘active layer’) that is typically 0.5–1.5 meters thick (Gilichinsky et al 1995). In the High Arctic, permafrost can extend as much as 1000 meters deep into the ground at temperatures of 0 to $-17$ °C (Steven et al 2006). Permafrost landscapes may contain massive ice features like ice wedges and segregated ice (Douglas et al 2011). Ice wedges form from repeated cycles of frost cracking and infiltration of snow, meltwater, soil, and other materials that eventually freezes and, through accumulation, can form features many meters high (Katayama et al 2007, Douglas et al 2011). When the thermal stability of permafrost is altered, rapid (less than a year) degradation, surface subsidence, and formation of pits and valleys (thermokarst) can occur (Jorgenson et al 2001, Jorgenson and Shur 2007). Dominant thaw features include sinkholes (Farquharson et al 2019), wildfire-driven surface thaw (Rowland et al 2010), and large slumps and landslides (Lewkowicz and Way 2019). These permafrost degradation features are associated with release of reservoirs of greenhouse gases (GHGs) (Koven et al 2015, 2017, Schuur et al 2015) and promotion of GHG-generating metabolic activity in microbes that have remained viable in the permafrost for up to 2 million years (Gilichinsky et al 1995). Microbes capable of growth and metabolic activity at low temperatures (Hultman et al 2015) are characteristic of permafrost and include bacteria and archaea that are higher in abundance than in other cryoenvironments (Steven et al 2006, Mondv et al 2014, Kao-Kniffin et al 2015, Mackelprang et al 2017).

Although a handful of studies have assessed INPs from soil or leaf litter at high latitude locations, permafrost has not specifically been evaluated. Conen et al (2016) discovered that decaying leaves were responsible for the abundant source of airborne INPs at a coastal mountain observatory in Norway, while
Schnell and Vali (1973) reported INPs in samples from decaying leaf litter in Siberia. Wilson et al (2006) tested the ice nucleation properties of bacteria isolates from two Canadian locations, but only from the upper 1–2 cm of soil. These studies demonstrate that ice nucleation active bacteria and fungi have been recovered from high latitude soil landscapes, but none tested INPs in the deeper soil (i.e. permafrost). Ponder et al (2005) isolated bacteria from Siberian permafrost and tested the cultured isolates for their ice nucleation activity. However, this study was focused on a small subset of permafrost bacterial strains (nine) and measured little to no ice nucleation activity at −10 °C.

Here, we report, for the first time, that permafrost is a reservoir of INPs on par with other terrestrial topsoils. We first describe the methods for sample collection and the ice nucleation measurements, followed by discussion of the results, and finally close with our main conclusions and broader implications. The goal of this work is to demonstrate that permafrost should be considered in future Arctic aerosol-cloud interaction efforts and to encourage further observational and modelling investigations to better understand the breadth of this potentially critical and widespread INP source. This realization is timely, given the rapidly-thawing permafrost in Alaska, the Arctic, and the sub-Arctic (Slater and Lawrence 2013).

2. Materials and methods

2.1. Permafrost sample collection

Permafrost, ice wedge, and active layer samples were collected from the Cold Regions Research and Engineering Laboratory’s (CRREL) Permafrost Tunnel Research Facility in Fairbanks, Alaska, USA (64.9528 °N, 147.6178 °W; https://www.erdc.usace.army.mil/CRREL/Permafrost-Tunnel-Research-Facility/) (Shur et al 2004, Douglas et al 2011, Burkert et al 2019, Douglas and Mellon 2019). The older permafrost and ice wedge samples were collected in the tunnel in August 2018 while the younger permafrost and active layer samples were collected in August 2019 (see table 1). The ages of the permafrost sample locations have been determined to isolate heat-labile (e.g. proteinaceous) and organic versus inorganic INPs in the youngest and oldest permafrost and ice wedge samples. The stability (or lack thereof) of INPs to these treatments provides an indication of composition. Climate models of all scales require information on INP sources to accurately represent ice nucleation and thus cloud microphysics, especially considering: (1) biologically-derived INPs form ice at cloud temperatures as high as −2 °C while certain mineral dusts, with the exception of some feldspars, glaciate modestly starting at −12 °C and (2) biologically-derived and mineral INP concentrations can vary by several orders of magnitude at any given temperature (Hoossein et al 2010a, 2010b, Burrows et al 2013, Petters and Wright 2017).

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Permafrost and ice wedge core samples in the tunnel were collected using a 10 cm diameter by 10 cm deep hole saw connected to an electric drill. Samples were immediately placed in plastic bags, then transferred and stored frozen at Colorado State University (CSU) at −20 °C for approximately 6 months until analysis. The younger surface permafrost and active layer samples were collected using a gas powered SIPRE augur which provides an 8 cm diameter core. The age of the younger surface permafrost and active layer samples was estimated by assuming about 1 mm year^{-1} of loess deposition in the area (Hamilton et al 1988).

Permafrost samples presented in the current work are compared to other samples, including: (1) high latitude sediment samples collected from 0 to 2 cm deep from a glacial outwash plain region in Svalbard, Norway at the Mt. Zeppelin Observatory during intensive measurement campaigns in July 2016 and March 2017 (details found in Tobo et al 2019) and (2) topsoil samples collected from Wyoming and Colorado in spring 2011, winter and summer 2012, and winter 2013 (details found in Hill et al 2016). The glacial outwash sediment is presented to compare to another high latitude location, while the midlatitude soil sample data are provided in order to compare our results to those from a different geographical region.

2.2. Ice nucleation measurements

For processing, a slurry of 4 g of thawed permafrost soil, active layer soil, or ice wedge was mixed into 40 ml of deionized water in a 50 ml polypropylene vial. From each slurry, a 100-fold dilution, followed by serial dilutions at 2000-, 40 000-, 800 000- and 16 000 000-fold were made in DI water. From each sampled position on select cores, 10–20 g of frozen material was scraped off and used to obtain dry weights (105 °C for 24 h). Dry matter contents were ~95% for the old permafrost (30 000 years old) sample and ~50% for the surface and active layer. For the ice wedge, INP concentrations are expressed per g of ice.

INP concentrations in the samples were measured using CSU’s Ice Spectrometer (IS), which is an immersion freezing measurement device with well-established, documented experimental protocols (Hiranuma et al 2015, Mccluskey et al 2018, Suski et al 2018). Frozen aliquots of 50 µl were counted at 0.5 °C intervals as temperature was lowered at ~0.33 °C min^{-1} to approximately −30 °C. The IS is semi-automated (LabVIEW; NI, Inc.) and detects aliquot freezing with a CCD camera system.

Thermal and chemical treatments were conducted to isolate heat-labile (e.g. proteinaceous) and organic versus inorganic INPs in the youngest and oldest permafrost and ice wedge samples. The stability (or lack thereof) of INPs to these treatments provides an indication of composition. Climate models of all scales require information on INP sources to accurately represent ice nucleation and thus cloud microphysics, especially considering: (1) biologically-derived INPs form ice at cloud temperatures as high as −2 °C while certain mineral dusts, with the exception of some feldspars, glaciate modestly starting at −12 °C and (2) biologically-derived and mineral INP concentrations can vary by several orders of magnitude at any given temperature (Hoose et al 2010a, 2010b, Burrows et al 2013, Petters and Wright 2017).
2015, Demott et al 2016, Kanji et al 2017, Vergara-Temprado et al 2017, Mccluskey et al 2019). However, due to limited observations and a lack of knowledge about the roles of different potential INP classes and their sources, models currently struggle to disentangle the effects of biological versus mineral INPs, leading to significant biases and uncertainties in cloud formation and the ability to project future changes (Mccluskey et al 2019). We need more comprehensive characterizations of INPs, as opposed to solely total INP concentrations. To assess the contribution of heat-labile entities such as proteins, a 1.5 ml aliquot of suspension was re-tested after heating to 95 °C for 20 min. To remove all organic INPs of biomolecular origin, 0.75 ml of 30% H2O2 was added to a 1.5 ml aliquot of suspension and the mixture heated to 95 °C for 20 min while illuminated with UVB fluorescent bulbs to generate hydroxyl radicals (residual H2O2 is removed using catalase (Mccluskey et al 2018)), and the sample retested in the IS. Remaining INPs were likely to be mineral (Conen et al 2011, Hill et al 2016, Mccluskey et al 2018).

From the fraction of drops frozen and the volume and dilution of suspension used, estimated INP concentrations were calculated using the equation in Vali (1971):

\[ K(\theta) \langle L^{-1} \rangle = \frac{\ln(1-f)}{V_{\text{drop}}} \times \frac{V_{\text{suspension}}}{m_{\text{material}}}, \]

where \( f \) is the proportion of droplets frozen, \( V_{\text{drop}} \) is the volume of each aliquot, \( V_{\text{suspension}} \) is the volume of the suspension, and \( m_{\text{material}} \) is the mass of soil in the suspension. Procedural controls were used to correct for background INPs. Confidence intervals (95%) were calculated based on the methodology of Agresti and Coull (1998).

### 3. Results and discussion

Cumulative INP spectra from the permafrost and ice wedge serial dilution samples are shown in figure 1. All the samples exhibited high concentrations of INPs, particularly at warmer temperatures (≥−15 °C) as indicated by the steep gradient within this range. There was no direct correlation between permafrost age and INP concentrations. However, we note that the oldest sample (30 000 years old) had the highest INP concentrations. There was also no difference between the ice wedge and permafrost soil samples. Above −8 °C, permafrost and ice wedge INP concentrations were up to 1 order of magnitude less than glacial dust and midlatitude soil samples reported previously (Hill et al 2016, Tobo et al 2019). However, below this threshold, permafrost and ice wedge INP concentrations were comparable or up to 1 order of magnitude higher than the previously-reported glacial and midlatitude topsoil concentrations. Previous work by Schnell and Vali (1976) reported that INPs are more enriched in soils of colder climates, aligning with the current work.

INP concentrations for the ice wedge, oldest permafrost, and youngest permafrost samples dropped significantly when treated with heat and with peroxide (figure 2, table 2), indicating a substantial quantity of proteinaceous INPs (or other heat-labile ice nucleating molecules) and the predominance of organic INPs in all samples. Interestingly, the largest quantity of what were likely proteinaceous INPs, as indicated by their sensitivity to 95 °C, was found in the ice wedge sample (figure 2(c))—INPs were mostly ≥2 orders of magnitude less at all temperatures after heating. Ice wedges contain abundant bacteria and archaea as compared to other ice habitats (Katayama et al 2007, 2009, 2010), but their microbial communities can reflect surrounding soil communities (active layer and permafrost) (Wilhelm et al 2012). Additionally, Wilhelm et al (2012) found that the most abundant taxa in ice wedges were Pseudomonas spp., belonging to the P. fluorescens group (Anzá et al 2000), which are known to contain efficient ice nucleating proteins (i.e. can nucleate at −3.8 °C) (Hazra et al 2004). Pseudomonas spp. have also been found to be abundant in permafrost soils (Warren et al 1986, Singh et al 2017). Of course, ice nucleating proteinaceous (or other molecular classes of) material may be produced by other microbes.

For the peroxide-treated samples, concentrations were 1–5 orders of magnitude less than in untreated samples. Specifically, the youngest permafrost sample contained roughly 10 times more organic INPs than the oldest sample at −15 °C and below, consistent with where most soil organic carbon is found.
Figure 1. Cumulative INP spectra from serial dilutions of permafrost and ice wedge samples collected at the CRREL Permafrost Tunnel in Fairbanks, AK. The ages (in years old or ‘yo’) are provided. For context, glacial outwash plain dust (Tobo et al 2019) and midlatitude soil (Hill et al 2016) data are shown. The grey markers represent four blanks run during permafrost/ice wedge sample processing. Asterisks indicate samples that were dried prior to testing. Error bars indicate 95% confidence intervals.

(i.e. in the top 1 meter of permafrost (Hugelius et al 2014)). The organic nature of the ice nucleating ability of the permafrost samples is consistent with glacial outwash plain data reported by Tobo et al (2019). Previous studies have indicated that interactions between glacial ice and permafrost sediment can occur in regions where glaciers reside on or near permafrost (Waller et al 2012), even in Svalbard valley glaciers (Etzelmüller et al 1996, Morrison et al 2005). Samples from Tobo et al (2019) were collected from the Brøggerbreen glacier near Ny-Ålesund in Svalbard where nearby permafrost exists (Humlum et al 2003). Given the similarity in results for our permafrost samples and the glacial outwash plain sediment from Tobo et al (2019), it is possible the outwash soil from Tobo et al (2019) was thawed permafrost sediment exposed once the glacier had retreated, mixed with other glacial minerals, producing unique outwash microbial community.

4. Summary and broader implications

We report the first observations of INP measurements from permafrost and ice wedge samples spanning from <500 to 30,000 years old. INP concentrations from these samples rivalled and, at certain temperature ranges, surpassed those of previously reported glacial outwash plain dust and midlatitude soil.

As permafrost thaws, microbes, organic matter, and their INPs could be aerosolized directly into atmosphere from wind erosion or exchange with thermokarst lakes, rivers, and coastal waters to be emitted at the water-air interface (Benner et al 2005, Schuur et al 2015, Park et al 2019, Wild et al 2019). The presence of the active layer could prevent deeper permafrost soils from direct interaction with the atmosphere; however, there are other possible mechanisms for exchange. For example, microbial emissions can transfer to the atmosphere via exchange from the thawed permafrost (Katayama et al 2007) to thermokarst lakes (Matheus Carnevali et al 2015), especially from retrogressive slumping (Ward Jones et al 2019), and ultimately to the air from bubbling up through such lakes. Organic matter-rich thermokarst lake sediments may be eroded through freeze-thaw cycles and mobilized through soil-water interactions (Carson and Hussey 1962, Hinkel and Nelson 2003, Schuur et al 2015, Vonk et al 2015). Such lakes can also drain into tundra river systems and, through riverine transport, can expand the spatial reach of thermokarst materials including microbes and the nutrients they feed on (Rowland et al 2010, Reyes and Lougheed 2015). Additionally, thermokarst erosion, landslides, and hillslope thaw slumping along coastlines, river banks, and lake shores have been increasing due to warmer Arctic summers and can introduce large quantities of thawed soil into lakes, streams, rivers, and oceans (Burn and Lewkowicz 1990, Lantuit and Pollard 2008, Rowland et al 2010, Wobus et al 2011, Malone et al 2013, Vonk et al 2015, Lewkowicz and Way 2019). Once in water, particulate material, including INPs, can be ejected into the atmosphere as lake, river, or sea spray aerosol (Wilson et al 2015, Demott et al 2016, Pietsch et al 2017, Knackstedt et al 2018, Moffett et al 2018)—the same mechanism could be applicable to permafrost INPs found in water catchments.
These studies indicate permafrost is rapidly thawing and degrading, which could expose large areas of thawed permafrost—and thus, its INPs—directly to the atmosphere or indirectly through soil-water-air exchanges. We suggest that INPs liberated from thawing permafrost—a hitherto overlooked source of Arctic INPs—have the potential to significantly influence INP abundance in the atmosphere and cloud radiative properties in the Arctic. Understanding permafrost as a substantial source of INPs and their possible impacts on clouds is urgent given the widespread thawing of permafrost in the Arctic.
Table 2. Percentages of total INPs that were proteinaceous (from heat treatment; first row for each permafrost soil type) and organic (from peroxide treatment; second row for each permafrost soil type and first and only row for glacial dust samples) for the youngest (surface) permafrost, oldest permafrost, ice wedge, and glacial dust samples (from Tobo et al 2019).

| Soil type    | –10 °C  | –15 °C  | –20 °C |
|--------------|---------|---------|--------|
| 1000 yo permafrost | 85%   | 24%    | 49%    |
|              | 100%   | 100%   | 100%   |
| 30 000 yo permafrost | 97%   | 84%    | 85%    |
|              | 100%   | 100%   | 100%   |
| 24 000 yo ice wedge | 100%  | 100%   | 100%   |
|              | 100%   | 100%   | 100%   |
| glacial dust 1  | 100%   | 100%   | 100%   |
|              | 100%   | 100%   | 99%    |
| glacial dust 2  | 100%   | 100%   | 99%    |

(Farquharson et al 2019), especially considering permafrost covers 15% of land in the Northern Hemisphere (Obu et al 2019). Recent studies have reported that the response of permafrost to rising global temperatures and diminishing sea ice is unclear, yet is abrupt in approximately 20% of the permafrost zone (Turetsky et al 2020). Thus, permafrost thaw is accelerating faster than can be predicted (Vaks et al 2020), having implications for collapsing ground, rapid erosion, and landslides. However, the resulting effects on clouds have not been considered when assessing the risks of thawing permafrost (Schuur and Abbott 2011). Atmospheric INP concentrations in the Arctic are rather low (i.e. typically range from 10−5 to 1 l−1; Creamean et al 2018 and references therein), but possible future increases in INPs as permafrost thaws rapidly, assuming they become airborne and reach cloud level, could have significant influences on AMPCs given their sensitivity to INPs (Solomon et al 2018).

Future research should focus on climate impacts of thawing permafrost through its potential to affect clouds, in addition to its contributions of GHGs. Due to a projected cloudier (a 1% decrease in sea ice leads to a 0.5% increase in cloud cover, suggesting that a further decline in sea ice cover will result in an even cloudier Arctic; Liu et al 2012) and rainier (60% of future Arctic precipitation is predicted to be rain compared to 35% in the present day; Bintanja and Selten 2014, Bintanja and Andry 2017) ‘New Arctic’, there is a critical need to understand the role of regionally-sourced aerosols in cloud and precipitation formation (Jeffries et al 2013, Overland et al 2018).

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Data availability

Any data that support the findings of this study are included within the article supplementary data files stack.iop.org/ERL/15/084022/mmedia.

ORCID iDs

Jessie M Creamean @ https://orcid.org/0000-0003-3819-5600
Paul J DeMott @ https://orcid.org/0000-0002-3719-1889

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