Global variability and controls on the accumulation of fallout radionuclides in cryoconite

Caroline C. Clason, Giovanni Baccolo, Edyta Łokas, Philip N. Owens, Przemyslaw Wachniew, Geoff E. Millward, Alex Taylor, Will H. Blake, Dylan B. Beard, Ewa Poniecka, Nick Selmes, Elizabeth A. Bagshaw, Joseph Cook, Ralph Fyfe, Melanie Hay, Deborah Land, Nozomu Takeuchi, Massimiliano Nastasi, Monica Sisti, Francesca Pittino, Andrea Franzetti, Roberto Ambrosini, Biagio Di Mauro

PII: S0048-9697(23)03525-8

DOI: https://doi.org/10.1016/j.scitotenv.2023.164902

Reference: STOTEN 164902

To appear in: *Science of the Total Environment*

Received date: 28 March 2023

Revised date: 12 June 2023

Accepted date: 12 June 2023

Please cite this article as: C.C. Clason, G. Baccolo, E. Łokas, et al., Global variability and controls on the accumulation of fallout radionuclides in cryoconite, *Science of the Total Environment* (2023), https://doi.org/10.1016/j.scitotenv.2023.164902

Global variability and controls on the accumulation of fallout radionuclides in cryoconite

CAROLINE C. CLASON[1*], GIOVANNI BACCOLO[2,3], EDYTA ŁOKAS[4], PHILIP N. OWENS[5], PRZEMYSLAW WACHNIEW[6], GEOFF E. MILLWARD[7], ALEX TAYLOR[7], WILL H. BLAKE[7], DYLAN B. BEARD[7], EWA PONIECKA[8], NICK SELMES[9], ELIZABETH A. BAGSHAW[10], JOSEPH COOK[11], RALPH FYFE[7], MELANIE HAY[12], DEBORAH LAND[13,14], NOZOMU TAKEUCHI[15], MASSIMILIANO NASTASI[16,17], MONICA SISTI[16], FRANCESCA PITTINO[18], ANDREA FRANZETTI[18], ROBERTO AMBROSINI[19] & BIAGIO DI MAURO[20]

[1]*Department of Geography, Durham University, Durham, UK*

[2]*Laboratory of Environmental Chemistry, Paul Scherrer Institut, Villigen, Switzerland*

[3]*Oeschger Centre for Climate Change Research, University of Bern, Bern, Switzerland*

[4]*Institute of Nuclear Physics PAS, Department of Mass Spectrometry, Krakow, Poland*

[5]*Department of Geography, Earth and Environmental Sciences, University of Northern British Columbia, Prince George, Canada*

[6]*Faculty of Physics and Applied Computer Science, AGH University of Science and Technology, Krakow, Poland*

[7]*School of Geography, Earth and Environmental Sciences, University of Plymouth, Plymouth, UK*

[8]*International Institute of Molecular and Cell Biology, Warsaw, Poland*

[9]*Plymouth Marine Laboratory, Plymouth, UK*

[10]*Bristol Glaciology Centre, School of Geographical Sciences, University of Bristol BS8 1SS UK*

[11]*Department of Environmental Science, Aarhus University, Aarhus, Denmark*

[12]*Department of Pathobiology and Population Sciences, Royal Veterinary College, University of London, London, UK*

[13]*Natural England, York, UK*

[14]*British Exploring Society, London, UK*

[15]*Department of Earth Sciences, Chiba University, Chiba, Japan*

[16]*National Institute of Nuclear Physics, Milano-Bicocca section, Milan, Italy*

[17]*Department of Physics, University Milano-Bicocca, Milan, Italy*

[18]*Department of Environmental and Earth Sciences, University Milano-Bicocca, Milano, Italy*

[19]*Department of Environmental Science and Policy, University of Milano, Milan, Italy*

[20]*Institute of Polar Sciences, National Research Council, Milan, Italy*

**Corresponding author email: caroline.clason@durham.ac.uk*

Abstract

The accumulation of fallout radionuclides (FRNs) from nuclear weapons testing and nuclear accidents has been evaluated for over half a century in natural environments; however, until recently their distribution and abundance within glaciers have been poorly understood. Following a series of individual studies of FRNs, specifically ^{137}Cs, ^{241}Am and ^{210}Pb, deposited on the surface of glaciers, we now understand that cryoconite, a material commonly found in the supraglacial environment, is a highly efficient accumulator of FRNs, both artificial and natural. However, the variability of FRN activity concentrations in cryoconite across the global cryosphere has never been assessed. This study thus aims to both synthesize current knowledge on FRNs in cryoconite and assess the controls on variability of activity concentrations. We present a global database of new and previously published data based on gamma spectrometry of cryoconite and proglacial sediments, and assess the extent to which a suite of environmental and physical factors can explain spatial variability in FRN activity concentrations in cryoconite. We show that FRNs are not only found in cryoconite on glaciers within close proximity to specific sources of radioactivity, but across the global cryosphere, and at activity concentrations up to three orders of magnitude higher than those found in soils and sediments in the surrounding environment. We also show that the organic content of cryoconite exerts a strong control on accumulation of FRNs, and that activity concentrations

in cryoconite are some of the highest ever described in environmental matrices outside of nuclear exclusion zones, occasionally in excess of 10,000 Bq kg^{-1}. These findings highlight a need for significant improvements in the understanding of the fate of legacy contaminants within glaciated catchments. Future interdisciplinary research is required on the mechanisms governing their accumulation, storage, and mobility, and their potential to create time-dependent impacts on downstream water quality and ecosystem sustainability.

Keywords: glaciers; contamination; environmental quality; radiocaesium; cryoconite; fallout radionuclides

1. Introduction

The cryosphere is currently undergoing significant change in response to climatic warming, posing a threat to glacier-fed freshwater resources with associated impacts for communities and ecosystems in polar and mountain environments (IPCC, 2019; Clason et al., 2023). Glaciers are important archives of atmospheric constituents, including contaminants, whose deposition history can sometimes be reconstructed through analysis of ice cores (e.g. Barbante et al., 2004). However, glaciers are open systems and materials preserved within their ice and snow will eventually be released into the downstream environment, including legacy contaminants first emitted to the atmosphere decades ago (Bogdal et al., 2009; Ferrario et al., 2017; Beard et al., 2022a, b). As such, glaciers act as secondary sources of contaminants discharged from both snow and ice, and from proglacial areas (Schmid et al., 2011; Pavlova et al., 2016), under present-day accelerated melting.

Cryoconite, a supraglacial material comprising both organic and inorganic components (Cook et al., 2016a), plays an important role as a sink for nutrients and contaminants (Bagshaw et

al., 2013; Baccolo et al., 2017; Li et al., 2017; Huang et al., 2019), and its ability to accumulate fallout radionuclides (FRNs) has received attention in several regions of the cryosphere (e.g. Łokas et al., 2018, 2019, 2022; Huang et al., 2019; Owens et al., 2019, 2023; Baccolo et al., 2020a,b; Clason et al., 2021; Cao et al., 2023). The activities of FRNs found in some cryoconite deposits are among the highest ever observed in environmental matrices outside of nuclear exclusion zones, with activities exceeding 10,000 Bq kg^{-1} for a single radionuclide (Tieber et al., 2009; Baccolo et al., 2017; Owens et al., 2019). FRNs are also present within glacial ice and snow, but at concentrations in the range of mBq kg^{-1} (Olivier et al., 2004; Gäggeler et al., 2020), several orders of magnitude lower than found in cryoconite. Such efficient accumulation of FRNs in cryoconite might be partly explained by its high organic matter content (Cook et al., 2016a), which has geochemical affinity for radionuclides and other particle-bound contaminants (Gadd, 1986). Cryoconite can often be in contact with supraglacial meltwater for prolonged periods, and this interaction with meltwater has also been proposed as a potential mechanism for accumulation of FRNs in cryoconite, via remobilisation, transport, and scavenging of legacy contaminants (Łokas et al., 2014, 2017; Owens et al., 2019; Baccolo et al., 2020a). Analysis of FRNs with different half-lives demonstrates that accumulation of FRNs in cryoconite from meltwater is a continuous process, and that the radioactivity burden of cryoconite increases with time before being released from glaciers to the downstream environment (Baccolo et al., 2020a, b). Once released, cryoconite and sorbed radionuclides are diluted in proglacial waters and sediments, although it has been shown that before being diluted, radionuclides can accumulate in proglacial areas, including lake sediments, posing potential ecological risks (Łokas et al., 2017, 2019; Owens et al., 2019, 2023; Clason et al., 2021).

This work focuses on Caesium-137 (^{137}Cs), Americium-241 (^{241}Am), and Lead-210 (^{210}Pb) as previous studies have demonstrated that they are the most abundant gamma-emitting FRNs present in cryoconite, with activities often exceeding 1000 Bq kg^{-1} for ^{137}Cs and ^{210}Pb (e.g., Baccolo et al., 2020b; Clason et al., 2021; Łokas et al., 2022). ^{137}Cs was introduced into the environment via atmospheric nuclear weapons testing from 1945 onwards, with the majority of test activity occurring in the 1950s and 1960s (UNSCEAR, 2000), prior to the decline of above-ground tests following the introduction of the Nuclear Test Ban Treaty in 1963. Accidental releases of ^{137}Cs from nuclear power and reprocessing plants have also been major contributors to global fallout, including the 1986 Chernobyl reactor fire and the Fukushima accident in 2011 (Bunzl and Kracke, 1990; Chino et al., 2011; Steinhauser et al., 2014), in addition to a range of other local and regional sources (Foucher et al., 2021). ^{137}Cs is a high-yield fission product of ^{235}U with a half-life ($t_{1/2}$) of 30.1 years, and due to this relatively long half-life, and its biogeochemical properties, ^{137}Cs has been able to spread across the globe, becoming one of the most studied radioactive contaminants in the environment (Cambray et al., 1989; Foucher et al., 2021). ^{241}Am ($t_{1/2}$ = 432 years) emits both alpha and gamma radiation. It is present in small quantities in spent nuclear fuel, but its environmental occurrence mostly depends on the decay of its parent radionuclide, ^{241}Pu ($t_{1/2}$ = 14.3 years), an important component of global radioactive fallout from atmospheric nuclear tests (Thakur and Ward, 2018). As a consequence of the decay of ^{241}Pu, the activity of ^{241}Am in the environment is increasing and is expected to peak by the end of the 21st century (Muravitsky et al., 2005). The geographic distribution of ^{137}Cs and ^{241}Am, and indeed all artificial FRNs, is not uniform, and notably the intensity of artificial global fallout in the Southern Hemisphere is lower than in the Northern Hemisphere, partly due to the greater number of above-ground bomb tests and nuclear accidents in the latter (UNSCEAR, 2000). Indeed, ~77% of total worldwide fallout of ^{137}Cs is estimated to have occurred in the Northern Hemisphere

(UNSCEAR, 2000; Evrard et al., 2020). ^{210}Pb ($t_{1/2}$ = 22.3 years) is a natural product of the ^{238}U series, produced from the decay of gaseous ^{222}Rn. In terrestrial soils and sediments, total ^{210}Pb comprises a supported ^{210}Pb component, which is continuously generated in situ from the decay of ^{226}Ra, and an unsupported or excess ^{210}Pb component (^{210}Pb-ex), which is produced by radioactive decay of atmospheric ^{222}Rn. Both wet and dry deposition returns the ^{210}Pb-ex to the Earth's surface. Global distribution of ^{210}Pb is variable and dependent upon air mass sources, while also demonstrating some latitudinal variability (Baskaran, 2011). From a global dataset of atmospheric ^{210}Pb, Zhang et al. (2021) identify the highest concentrations in the mid-latitudes of the Northern Hemisphere, with higher depositional fluxes generally found in the Northern Hemisphere than the Southern Hemisphere due to its larger landmass and associated radon emanation rates (Baskaran, 2011).

To date, studies on the accumulation of FRNs in cryoconite have focussed on individual glaciers or regions. The objectives of this study are thus: 1) to provide the first global database of FRNs in cryoconite and assess geographical variability of activity concentrations; 2) to evaluate controls on the accumulation of natural and anthropogenic FRNs in cryoconite; and 3) to assess the extent to which FRNs accumulate more efficiently in cryoconite than proglacial sediments. Here we synthesise and evaluate both new and previously published data documenting the presence of FRNs in cryoconite and sediments from proglacial settings, and address the influence of key environmental variables on FRN activity concentrations. We also review current gaps in understanding of the downstream consequences of legacy contaminant release in glaciated catchments, and identify priority areas for future research in this field.

2. Study sites

The data presented here span five continents, including 32 individual sites – 20 of which are previously unpublished – and a total of 520 individual cryoconite samples. We also include data from a further 209 sediment samples from the proglacial areas of 11 glaciers. The spatial coverage of these data provides insight into the variability of fallout radionuclide activity in cryoconite from icefield, ice cap, ice sheet and mountain glacier settings, spanning ~70 to 5500 m a.s.l., and from 79^{o} N to 79^{o} S. These sites represent cool temperate, boreal, and polar temperature domains, and both moist and dry moisture domains (Figure 1; Sayre et al., 2020). Details of individual sites, and associated FRN data, are provided in Tables A1 and A2.

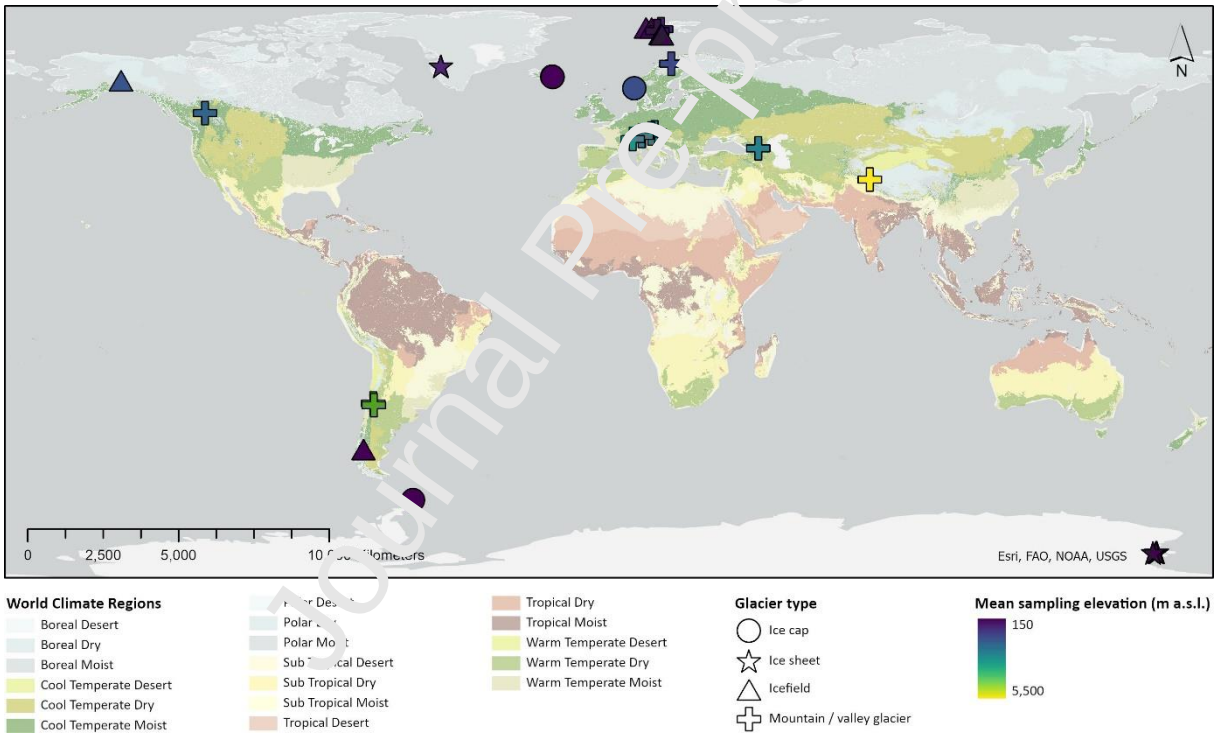


Figure 1. Sampling site locations with glacier type and mean sampling elevation indicated. World climate regions, after Sayre et al. (2020), are also shown.

3. Methods

3.1 Sample collection and preparation

The cryoconite and proglacial sediments described here were sampled in the field between 1992 and 2020. Cryoconite sampling was conducted on the ablation zones of various glaciers, with the samples collected using plastic or stainless-steel tools (disposable pipettes, spatulas, or trowels) and stored in sterile plastic tubes. In some cases, the samples were preserved in 96% ethyl alcohol. In the laboratory, the majority of samples were stored at -20ºC and were subsequently evaporated and dried at 70-105°C. Most of the samples were fine-grained and sufficiently homogeneous that no further treatment (e.g. sieving) was required, and previous work has shown that particle size of cryoconite is less important for FRN accumulation than other physical variables such as organic content (Owens et al., 2019). Samples from Canada were sieved to <63 µm in the laboratory prior to analysis. Proglacial soils and sediments from Svalbard were collected using a stainless steel/PVC cylinder. Proglacial samples from Sweden and Iceland were collected using a plastic trowel, oven/freeze-dried, and sieved to <75 µm prior to analysis. A sediment core from a proglacial lake was also collected in Sweden using a HTH 90 mm diameter gravity corer and was extruded and sliced into 1 cm sections in the field, oven dried at 100°C, and sieved to <75 µm. Further details on specific sampling protocols for previously published data can be found in published sources (see Tables A1 and A2).

All FRN data reported here are based on samples collected by authors of this study, with the exception of cryoconite samples from the Hallstatter/Schladminger and Stubacher Sonnblickkees glaciers which were collected and first reported by Tieber et al. (2009) and Wilflinger et al. (2018) respectively, and proglacial sediments reported by Chmiel et al. (2009) from the Scott Glacier catchment in Svalbard. Most of the data presented here were initially collected for studies of individual glaciers or regions, so it is important to acknowledge that there is variability in both sample collection and preparation methods. From

our collective experience, we recommend the following for future cryoconite sampling and preparation for analysis of FRNs: that at least 6 g of cryoconite (dry weight) is collected per sample; that cryoconite is collected using plastic utensils and stored in sterile, water-tight plastic containers; that cryoconite is freeze-dried where possible; and that cryoconite is analysed in bulk unless containing obviously coarse particles or large granules, in which case it should be sieved to <63 µm.

3.2 Gamma spectrometry

Analysis of FRNs, supported by analytical quality assurance, was carried out by gamma spectrometry in three laboratories in Europe (University Milano-Bicocca, Italy; Institute of Nuclear Physics PAS, Poland; University of Plymouth, UK) and one in Canada (University of Manitoba, Canada). Table A3 provides further information on the analytical capabilities and quality assurance for these facilities. Gamma analysis using a well detector requires the dried cryoconite samples to be packed and sealed in 4 ml plastic/polypropylene vials, and the quantity of material for each of our samples varied between 0.5 g and 22 g. The sealed samples were incubated for a minimum of 24 days to allow establishment of radioactive equilibrium with ^{222}Rn and its progenies, prior to counting. Activity concentrations were determined using different detectors with a counting time of at least 24 hrs (up to 72 hrs for samples with lower weights). The isotopes ^{210}Pb ($t_{½}$ = 22.3 years), ^{241}Am ($t_{½}$ = 432 years), and ^{137}Cs ($t_{½}$ = 30.1 years) were determined by their gamma emissions at 46.52, 59.54, and 661.6 keV, respectively, for the majority of samples, with ^{241}Am in some samples from Svalbard, the Caucasus, and Ecology Glacier, Antarctica analysed by alpha spectrometry (Łokas et al., 2018). The data were verified by inter-laboratory comparisons with soil/sediment and reference materials (IAEA, Vienna, Austria; see Table A4 for exemplar quality assurance data). All values given here are correct to the date of sampling where sampling was conducted

between 2015 and the present day, with other values decay-corrected to 2020 to optimise comparison between sites. We acknowledge that use of different detectors and run times introduces uncertainty when making direct comparisons, however the nature of this study and inclusion of previously published data precluded the use of a single, standardised approach. We also acknowledge that there is uncertainty around the extent to which cryoconite will adsorb/absorb and leach radionuclides, but decay-correcting values from prior to 2015 allows us to represent where radioactivity is likely to have decreased over longer timescales. We report ^{210}Pb-ex unless otherwise stated, which is estimated by subtracting ^{214}Pb from total ^{210}Pb (Zaborska et al., 2007).

3.3. Loss on ignition

A subset of 229 cryoconite samples from 11 regions were analysed for loss-on-ignition (LOI) to provide an estimate of the organic content of the samples. Ceramic or aluminium crucibles were preconditioned at 105°C overnight prior to analysis and were allowed to cool in a desiccator before having their weight recorded. All samples were dried at 105°C for a minimum of 2 hours prior to being placed in a furnace. 1 g (± 0.9 g) of each sample of dried cryoconite was placed into a preconditioned crucible and the weight recorded. Each sample was heated at either 600°C or 1050°C for 1 hour before being allowed to cool for 10 minutes and re-weighed. LOI was calculated, as a percentage, as follows:

$$LOI = (W_s - W_A)/(W_s - W_c) \times 100$$

Where W_s is the crucible plus sample dried at 105°C (g), W_A is the crucible plus ignited sample (g), and W_c is the preconditioned crucible (g). The difference in heating temperature

and crucible material used here reflects that samples were analysed for LOI in two different laboratories (University of Plymouth, UK; Institute of Nuclear Physics PAS, Poland).

4. Results and discussion

4.1 Geographical variability in FRN activities in cryoconite

The sites included in our database span a range of glaciological settings and a wide range of climatic regimes, from the polar desert of the Dry Valleys of Antarctica to the moist, coastal region of Vatnajökull in SE Iceland. The highest ^{137}Cs activities for individual samples exceed 100,000 Bq kg^{-1} and are found in cryoconite from glaciers in the Eastern European Alps (Figure 2; Table A1): the Hallstatter / Schladminger glaciers and Stubacher Sonnblickkees. Outside of the Alps, the highest value for individual samples (24,500 Bq kg^{-1}) was found in cryoconite from Norway, with lower but still elevated activities (>4000 Bq kg^{-1}) recorded for other sites in the Alps, Alaska, Sweden, and the Caucasus. The Eastern Alps and the Scandinavian Peninsula were among the regions most impacted by ^{137}Cs deposition following the 1986 Chernobyl event (European Commission, 2009; Meusburger et al., 2020), as reflected in their FRN activities. The lowest ^{137}Cs activities for individual cryoconite samples (<10 Bq kg^{-1}) were found in samples from Iceland, the Chilean Andes, Alaska, and the Antarctic Dry Valleys, with the lowest maximum values also recorded in the Dry Valleys. The highest individual value of ^{210}Pb-ex was found in a sample from the Ladakh Himalaya, at >27,500 Bq kg^{-1}, while elevated values (>10,000 Bq kg^{-1}) were also found in cryoconite from the Alps, Alaska, Sweden, Norway, Svalbard, and the Caucasus. The lowest values of ^{210}Pb-ex in individual samples (<100 Bq kg^{-1}) were found in the Antarctic Dry Valleys, Iceland, the Chilean Andes, Alaska, and the Greenland Ice Sheet.

There is considerable inter- and intra-regional variability in ^{241}Am in cryoconite, which was not detected / reported for sites in the Dry Valleys, some sites in Svalbard, and one site in Greenland. Detector sensitivity and analytical conditions, such as sample size, run times, and backgrounds, preclude the detection of low activities of ^{241}Am in cryoconite above the MDAs (minimum detectable activities) at some sites, while the low activity concentrations reported for other sites are detectable but often return high uncertainties (Table A1). For sites where ^{241}Am is reported, the highest maximum values (>50 Bq kg^{-1}) for individual samples were recorded in cryoconite from Svalbard, the Caucasus, Sweden, Norway, and the European Alps, with the highest value of 120 Bq kg^{-1} recorded for the Morteratsch Glacier in Switzerland.

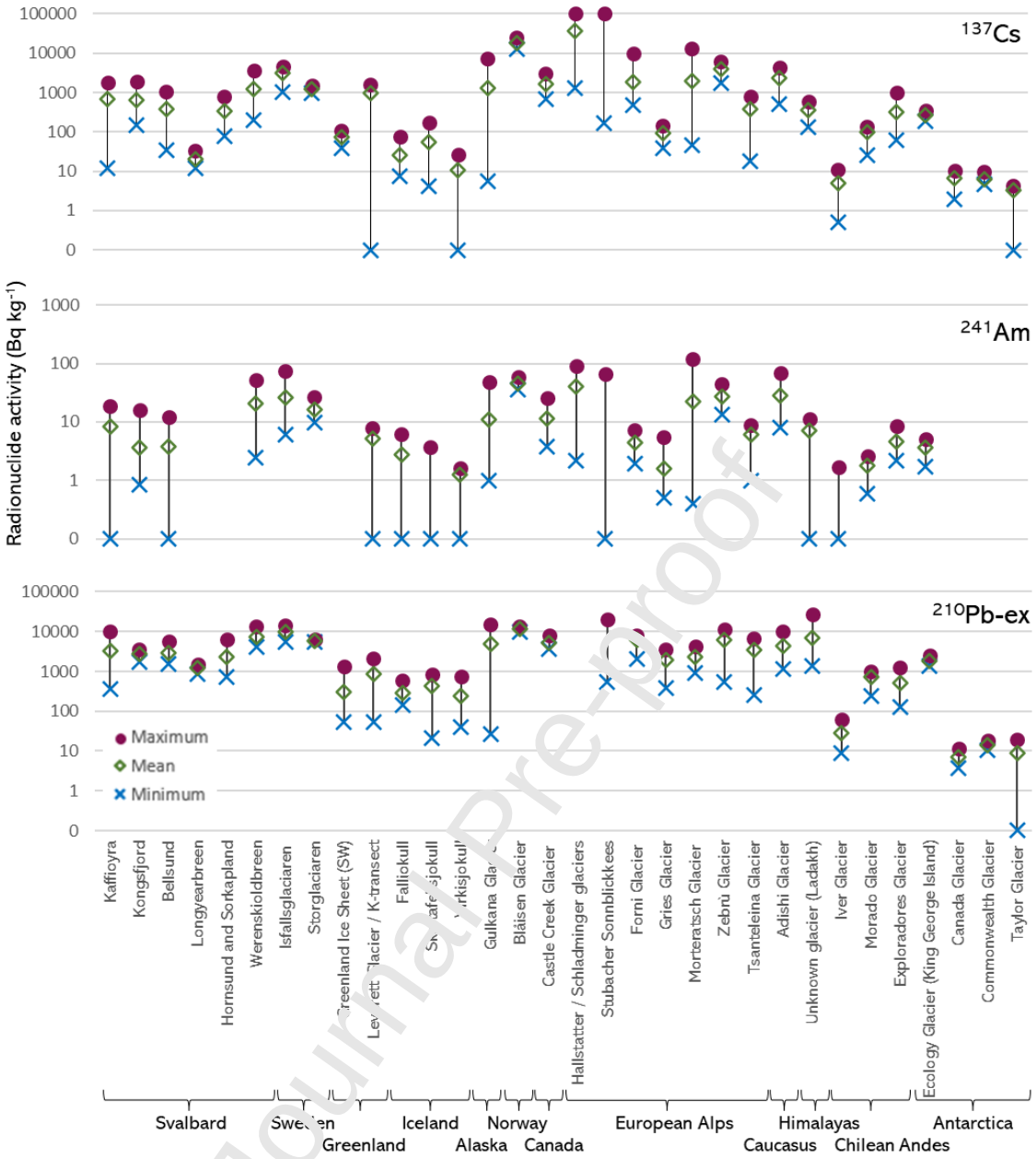


Figure 2. The range and mean values of ^{137}Cs, ^{241}Am, and ^{210}Pb-ex activities recorded in cryoconite samples from across the global cryosphere - see Table A1 for data. Note that logarithmic scales and different scale ranges are used due to the large variability in activity values. Data are presented in latitudinal order from north to south.

The highest mean activities of ^{137}Cs in cryoconite (>2000 Bq kg^{-1}) were found in the European Alps, the Caucasus, Sweden, and Norway, up to a maximum of ~37,400 Bq kg^{-1} on the Hallstatter / Schladminger glaciers. Mean ^{137}Cs values at some sites in Greenland, Iceland,

Antarctica, the Alps, and the Chilean Andes are up to three orders of magnitude lower at <100 Bq kg^{-1} (Figures 2 and 3; Table A1). A similar spatial variability is also evident for ^{210}Pb-ex, with up to four orders of magnitude difference in mean activities between regions, e.g. <10 Bq kg^{-1} in the Antarctic Dry Valleys and ~11,800 Bq kg^{-1} in Norway. The highest mean ^{241}Am values (>40 Bq kg^{-1}) were reported for sites in the Austrian Alps and Norway, and the lowest (<2 Bq kg^{-1}) for sites in Iceland, the Chilean Andes, and the Swiss Alps. Mean activities demonstrate clear regional patterns, however the activity ranges found for most glaciers are high and partly overlap between regions (Figures 2 and 3). While we focus on inter-regional variability here, it has also been observed in the Alps that for a single glacier FRN activities in cryoconite can range by up to three orders of magnitude because of differing degrees of interaction between cryoconite deposits and meltwater transporting legacy nuclides (Baccolo et al., 2020a).

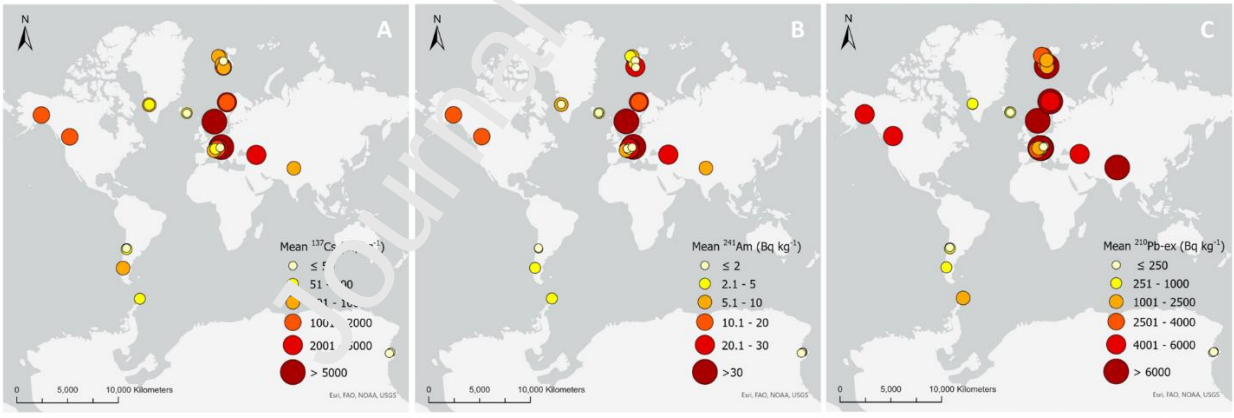


Figure 3. Spatial variability in mean activity concentrations of ^{137}Cs (A), ^{241}Am (B), and ^{210}Pb-ex (**C**) in cryoconite for sites across the global cryosphere.

4.2 Controls on the variability of FRN activity

Inter-regional patterns in FRN activity are controlled by a combination of climatological, environmental, and biogeochemical factors. Comparison of ^{137}Cs and ^{210}Pb-ex activities in

cryoconite with well-documented latitudinal global fallout proportions (UNSCEAR, 2000; Evrard et al., 2020) demonstrate a general accordance for sites in the Southern Hemisphere and the mid-latitudes (Figure A1). However, a number of sites do not follow this trend, with disproportionately high values for some sites above 60°N, most notably certain sites in Norway, Sweden and Svalbard. Reported FRN activities in cryoconite are lower in the Southern Hemisphere, increasing from very low values in the arid Antarctic Dry Valleys and Andean sites to higher values in more humid areas of the Andes and King George Island. The disproportionately elevated values at high northerly latitudes may be explained in-part by the influence of long-range atmospheric transport during positive phases of the North Atlantic Oscillation, which transports contaminants to the Arctic from distal sources (Stohl, 2006). However, the large variation in activities for Northern Hemisphere sites illustrates that latitude alone cannot account for FRN variability. Similarly, we find that although proximity to major FRN sources such as Chernobyl may explain some of the most extreme values when comparing artificial radionuclides in cryoconite, we find no significant correlation between ^{137}Cs and distance from Chernobyl based on the Pearson's Correlation Coefficient (Figure A2), and a moderate negative correlation for ^{241}Am and which deteriorates to no significant correlation when only Northern Hemisphere sites are considered. In comparison with activities from meta-analyses of radionuclides in soils and sediments globally (e.g. Evrard et al., 2020; Foucher et al., 2021; Figure A1), we find that ^{137}Cs activities in cryoconite are up to three orders of magnitude higher, demonstrating the unique affinity between FRNs and cryoconite.

We explored the correlation between FRN values and a range of other environmental variables (Figure A2) to investigate the potential influence of factors such as precipitation, continentality, and elevation. We use distance from the sea as a proxy for continentality, and

find no correlation with artificial radionuclides, but a moderate positive correlation with ^{210}Pb-ex. We also find a moderate positive correlation between ^{210}Pb-ex and mean elevation of sampling sites when Northern Hemisphere sites only are considered. No correlation was found between mean annual precipitation and ^{210}Pb-ex activity concentrations, however there is a moderate positive correlation between precipitation and both ^{137}Cs and ^{241}Am (Figure A2), highlighting the importance of atmospheric contaminant scavenging by precipitation (Franz and Eisenreich, 1998; Herbert et al., 2006). Artificial FRN activities increase with annual mean precipitation from the lowest values in Antarctica (e.g. ^{137}Cs = 3 Bq kg^{-1}; ^{214}Am = 0 Bq kg^{-1} for Taylor Glacier), to the high values reported for Scandinavian and Alpine glaciers, such as Blåisen Glacier (^{137}Cs = 18000 Bq kg^{-1}; ^{214}Am = 46 Bq kg^{-1}) and the Hallstatter/Schladminger glaciers (^{137}Cs = 37438 Bq kg^{-1}; ^{214}Am = 40 Bq kg^{-1}), with an exception of Iceland where the activities are amongst the lowest despite moderate precipitation (e.g. ^{137}Cs = 11 Bq kg^{-1}; ^{214}Am = 1 Bq kg^{-1} at Virkisjökull). There is high variability of ^{210}Pb-ex in terms of relationship to precipitation, and the strong degree of continentality at Ladakh might explain the high ^{210}Pb-ex concentration in cryoconite at this site which is more than 1400 km from the sea. Unsupported ^{210}Pb is often more abundant in the atmosphere at continental sites (Rangarajan et al., 1986; Baskaran, 2011), although enrichment of air masses with ^{210}Pb-ex is also dependent on patterns of air mass circulation (Zhang et al., 2021). Principal component analysis (PCA) of standardized geographical and environmental variables was conducted to further explore variance within the data, revealing clustering of regions (Figure A3; Table A5). There are large positive loadings from mean ^{137}Cs, ^{241}Am and ^{210}Pb-ex for principal component 1, explaining 43% of the data variance, with latitude, mean annual precipitation, and mean elevation exerting the strongest influence on principal component 2, explaining a further 20% of the variance. The PCA further supports that no single environmental factor is the primary control on variability of FRN activities.

While not assessed here, variations in FRN activities between glaciers will likely reflect the different inventories of FRNs within snow and ice layers, in addition to the residence time of water within glaciers and their accumulation area ratios, further controlled by glacier hypsometry and mass balance. Intra-glacier variations in cryoconite activities also likely reflect their different histories of exposure to FRNs, with cryoconite in cryoconite holes capturing radionuclides from both ice and meltwater interactions (Baccolo et al., 2020a). Heterogeneity in melt rates both between glacier sites and within the same site could thus have ramifications for quantification of supraglacial FRN inventories, with samples retrieved from high melt areas near the margin versus samples from lower melt areas further up-glacier yielding different FRN accumulation rates. Supraglacial meltwater pathways will also likely act as a control on intra-glacier FRN variability by determining the extent to which meltwater interacts with cryoconite deposits. Supraglacial pathways can be relatively stable at seasonal or even multi-year timescales, but are likely to be dynamic at decadal scales, influencing patterns of FRN mobilisation, scavenging, and accumulation. Furthermore, near-surface hydrology and the weathering crust can play an important role in regulating supraglacial discharge (Stevens et al., 2018) and in areas like the Antarctic Dry Valleys water in cryoconite holes can be relatively isolated yet contribute a significant portion of total runoff (Fountain et al., 2004). While no study has yet attempted to predict the fluxes of FRNs to proglacial areas from the supraglacial environment and through mobilisation of cryoconite, future research might incorporate modelling of melt rates and hydrological pathways to improve understanding of cryoconite-meltwater interactions (Cook et al., 2016b).

A crucial factor controlling FRN accumulation in cryoconite is its ability to capture and retain radionuclides. The capacity of cryoconite to bind radionuclides partly depends on the

extracellular polymeric substances (EPS) produced mainly by cyanobacteria (Pereira et al., 2011; d'Abzac et al., 2013), which have been demonstrated to be important for biogeochemical cycling (Nagar et al., 2021), and the very high FRN activities found in some samples indicate that this capacity is not easily saturated. Individual cryoconite granules are composed of recently accumulated biomass, although they may contain fragments of older granules that disintegrate after reaching a threshold size (Takeuchi et al., 2010). The granules sampled from a glacier thus represent a spectrum of life histories spanning different times of exposure to FRNs, reflecting highly variable radioactivity levels even on individual glaciers. Analysis of organic content via loss-on-ignition (LOI) for a subset of individual cryoconite samples in this study (Figure 4) supports that both ^{137}Cs and ^{210}Pb-ex activities are positively correlated with LOI and significant above the 95% confidence level. This relationship is strongly correlated when considering all sites, or only those in the Northern Hemisphere, and demonstrates a moderate, but still significant correlation when samples from Norway - which have particularly high organic content and ^{137}Cs concentrations - are excluded. The relative depletion of ^{137}Cs and ^{210}Pb in cryoconite for some sites might thus reflect a lack of older cryoconite material in high-ablation regions or highly dynamic supra-glacial environments, preventing prolonged periods of cryoconite-meltwater interaction and limiting accumulation of FRNs. When excluding Norwegian samples, the degree of correlation between FRN content and organic matter is higher for ^{210}Pb-ex than for ^{137}Cs, with a coefficient of determination exceeding 0.6 for ^{210}Pb-ex compared with 0.37 for ^{137}Cs. This might be explained by differences in the deposition mechanisms of the two radionuclides: continuous deposition for ^{210}Pb-ex, but event-related for ^{137}Cs. ^{137}Cs accumulation in cryoconite is also more influenced by local factors than ^{210}Pb-ex, whose deposition is more uniform both temporally and spatially (Persson and Holm, 2011). Moreover, it has been shown that the

degree of affinity for organic matter is higher for ^{210}Pb-ex than for ^{137}Cs (Teramage et al., 2013), which is compatible with our data.

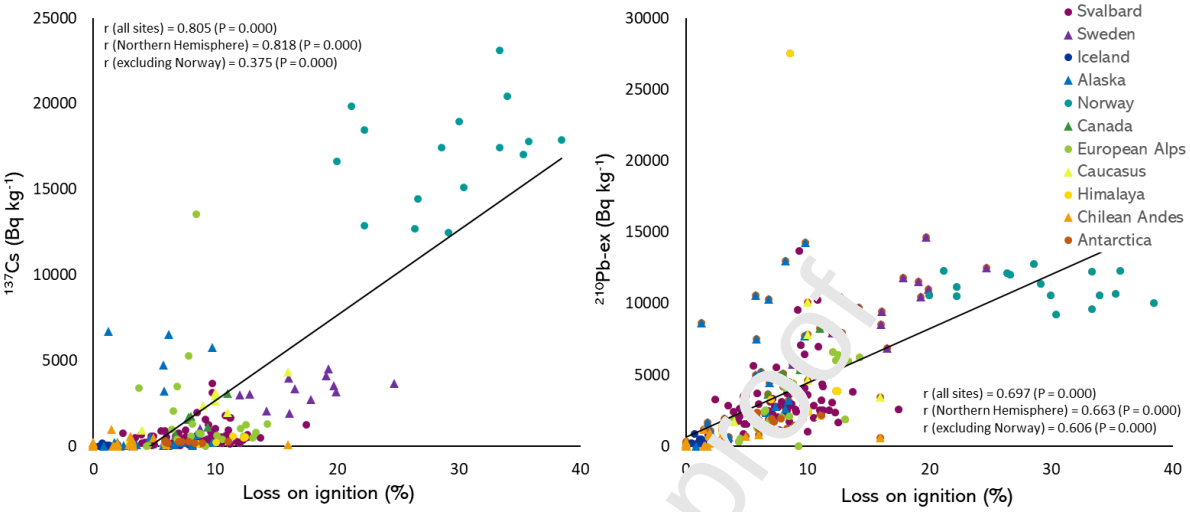


Figure 4. Correlation of Loss-on-Ignition (LOI; representing organic matter content) with ^{137}Cs and ^{210}Pb-ex for 229 individual cryoconite samples from regions in this study. The Pearson's Correlation Coefficient is given for samples from all sites, samples from the Northern Hemisphere only, and excluding samples from Norway.

Contemporary volcanic activity in Iceland and Chile could also affect the geochemical composition of cryoconite in these regions, in turn affecting its ability to accumulate FRNs, however further research is required to assess spatial variability in cryoconite geochemistry and the role of mineralogy, which has been shown to play a role in ^{137}Cs sorption in other environments (e.g. Nakao et al., 2015). The high clay content of cryoconite may also play a role in accumulation of ^{137}Cs, as demonstrated for soils in Japan following the Fukushima accident (Fujii et al., 2014; Baccolo et al., 2017), although some studies have shown that the particle size composition of cryoconite is less important than organic matter content (Owens et al., 2019). The controls on FRN accumulation at individual glacier sites most likely vary considerably between regions, reflecting a combination of intrinsic and extrinsic factors, such

that while proximity to FRN sources may override other factors at some sites, for other sites precipitation amount, glacial melt rate, or cryoconite organic matter composition and geochemistry may play a much more central role.

4.3 Transport and accumulation of FRNs in glaciated catchments

In addition to considering the role of cryoconite in scavenging FRNs on the surface of glaciers, it is important to consider the transport and release of FRNs in cryoconite, and implications for downstream environments. Hydro-sedimentological processes are key for mobilising cryoconite and associated FRNs into the proglacial environment in response to both longer-term patterns of glacier melt and event-scale increases in melt and precipitation, in addition to erosion and re-mobilisation of sediments within proglacial areas (e.g. Leggat et al., 2015). Numerous studies have reported that glaciers are retreating rapidly, with a total mass loss of 267 ± 16 Gt per year between 2000 and 2019 (Hugonnet et al., 2021), exposing new land to subaerial processes. Few studies have determined FRN activities in the soils and sediments of proglacial areas, which can also be helpful tracers of glacier retreat (Navas et al., 2017; 2020; Lizaga et al., 2019), and in most cases concentrations are considerably lower than those recorded in cryoconite (e.g. Hasholt et al., 2000; Table A2). Comparison of cryoconite with proglacial material for selected sites (Figure 5) demonstrates the ability of cryoconite to efficiently accumulate FRNs in comparison to proglacial sediments, with up to three orders of magnitude between activities in cryoconite and proglacial sediments for some sites. A study in British Columbia, Canada (Owens et al., 2019) documented declining FRN concentrations in fluvial sediments with increasing distance from the glacier terminus, while studies in Arctic Sweden (Clason et al., 2021) and Svalbard (Łokas et al., 2017) found that some radionuclides were higher in sediments from southern proglacial outlets than those from northern outlets, highlighting the importance of local variability in glacier topography and hydrology.

The lower activities found in proglacial sediments may be due to the dilution of cryoconite released from the glacier with sediment from other sources, such as eroding channel banks, hillslopes, and subglacial substrates, which typically contain much lower concentrations of FRNs (e.g. Evrard et al., 2020; Foucher et al., 2021). However, the activity of FRNs detected in soils and sediments in depositional environments can be relatively high, suggesting that downstream accumulation of FRNs, or localised off-ice 'hotspots' is possible for some sites (Łokas et al., 2017; Clason et al., 2021). Much of the total inventory of FRNs stored in and on glaciers is likely to be legacy in nature, having been deposited in the years following nuclear events. Given that it can take decades or longer for material to be transported from the accumulation zone to the termini of glaciers, we can expect the release of historical contaminants to increase under current and future warming. Inventories of FRNs in glacier ice remain very poorly constrained, however, so better understanding of contaminant load should be a focus for future studies to both contribute to environmental risk assessment and improve knowledge of FRN inventories in polar and mountain regions.

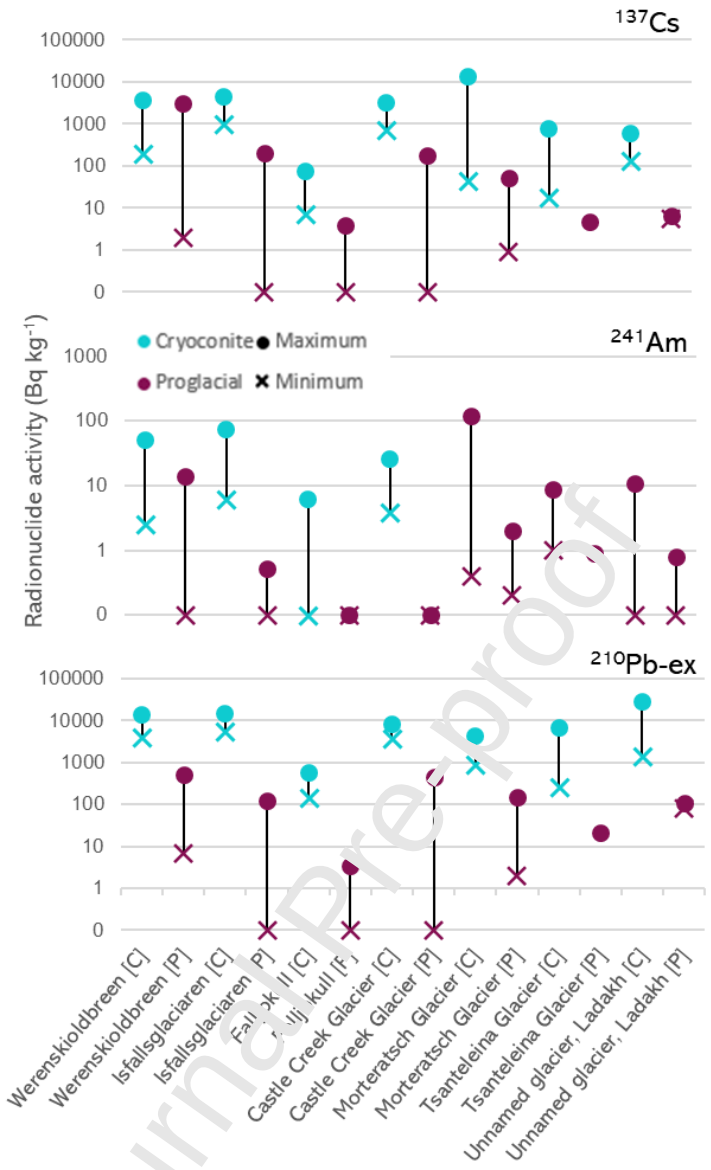


Figure 5. The range (minimum and maximum) of ^{137}Cs, ^{241}Am, and ^{210}Pb-ex activities recorded in selected cryoconite [C], and proglacial soil / sediment [P] samples. Note that logarithmic scales and different scale ranges are used due to large variability in activity values.

4.4 Downstream implications of legacy contaminant release

Here we have demonstrated the prevalence of high activities of FRNs in cryoconite throughout the global cryosphere. This highlights the importance of cryoconite within glacial systems for the accumulation of atmospherically deposited materials, including contaminants, and the role played by these unique biomes in biogeochemical transfers and cycling. What

remains poorly understood are the consequences of legacy contaminant release downstream, enhanced by continued melting and down-wasting of glaciers, for both ecosystem health and socio-economic impacts (Figure 6), with very few studies exploring both supraglacial and proglacial FRN activities to date (e.g. Owens et al., 2019; 2023; Clason et al., 2021). The behaviour and retention of FRNs in the environment, particularly in soils, depends on local geochemistry and organic content, including the inhabiting microbial community. Factors such as organic matter content, pH, and permeability of soils, in addition to reworking of proglacial deposits (Łokas et al., 2017), will affect the migration of FRNs once they reach the proglacial environment (Walther and Gupta, 2015). Proglacial soils are usually low in organic carbon (Bradley et al., 2014) with a low sorption capacity, which may result in relatively high mobility of FRNs in some regions. However, this mobility is limited in regions such as the High Arctic and Antarctica due to low bioturbation and long frost periods (Łokas et al., 2014). Further dispersal depends on the soil's microorganisms, which play a major role in controlling the cycling and toxicity of FRNs by direct reduction, uptake, and accumulation, as well as turnover of organic matter (Simonoff et al., 2007). The microbial community structure in glacier forefields differs significantly from the communities of cryoconite holes (Edwards et al., 2013) which could have a direct impact on the fate of FRNs released from glaciers. The freshly exposed soils in front of receding glaciers also undergo microbial community succession, gradually increasing in evenness, phylogenetic diversity, and number of phylotypes (Franzetti et al., 2020). FRN contamination has been shown to impact the microbial community structure and functionality of soil ecosystems in non-glaciated environments (Rogiers et al., 2021) which could negatively impact the establishment of soil communities in rapidly exposed soils. Data on the impact of FRN activity on proglacial soils and adjacent communities remain scarce, and more research is necessary to predict the downstream fate and toxicity of FRNs in these environments.

Figure 6. Chain of radionuclide transport and accumulation mechanisms within terrestrial glaciated environments.

In addition to affecting soil microbial communities, the introduction of FRNs within downstream environments has the potential to influence the health and fertility of flora and fauna. Vegetation in glaciated areas tends to have less developed root systems as an adaption to frozen ground. It therefore relies on absorbing nutrients atmospherically and aquatically (Meyers, 2012), and is particularly susceptible to the accumulation of FRNs. Furthermore, biomagnification through higher trophic levels of grazing fauna can lead to even higher concentrations of FRNs (Hong et al., 2011; Stocki et al., 2016), with potential socio-economic and health implications for communities that rely on livestock for food and livelihoods. These

populations are often indigenous communities who may have less access to standardised environmental quality testing (Stensrud, 2016). The impact of consuming local fauna, and by proxy the contaminated fauna and flora they have digested, as well as uptake from contaminated and eco-toxic water sources, could pose issues for both animal and human health (Donaldson et al., 2016). However, the extent to which the release of FRNs from glaciers poses a risk, if at all, is currently unknown, and there are notable research gaps around the processes involved in the accumulation and secondary deposition of FRNs downstream of glaciers, and associated effects on flora, fauna, human health and socio-cultural wellbeing. In addition to improving understanding of contaminant accumulation in cryoconite in under-represented regions of the cryosphere, these gaps should be addressed to assess and mitigate potential future risk to communities living within and downstream of glaciated environments.

5. Conclusions

Our global synthesis of FRN activities in 520 cryoconite samples from 32 different glacier environments shows, for the first time, that the accumulation of both artificial and natural FRNs in cryoconite is a commonplace and universal process, with FRN accumulation found in both polar and mountain glacier settings. Activities of ^{137}Cs, ^{241}Am, and ^{210}Pb in cryoconite are highly variable, both within individual glaciers and inter-regionally, but the activities we report are consistently, and significantly, above values typically reported in other environmental matrices such as soils and sediments. Indeed, observed activity concentrations in cryoconite can exceed common environmental values by up to 3 or even 4 orders of magnitude, with some activities for single FRNs exceeding 10,000 Bq kg^{-1}. Intra-glacier variability of FRN activity in cryoconite likely reflects differences in interaction between cryoconite and supraglacial meltwater, through which legacy contaminants stored in snow and

ice are released and mobilized in the glacio-hydrological environment. Between regions, the considerable variability in FRN activities is controlled by a combination of climatological, environmental, and biogeochemical factors, with a notable strong correlation between cryoconite organic content and radioactivity. The extent to which the release of FRNs from glaciers poses a downstream ecological threat is uncertain, but improved understanding of the behaviour of legacy contaminants within glacier catchments should be a future research priority to contribute to future environmental protection in ecologically vulnerable polar and mountain regions.

Our compilation and synthesis of a unique database of radionuclides in cryoconite samples was obtained using samples from a wide range of international sites analysed by non-systematic sample manipulation and instrumental approaches. Future progress in the understanding, and environmental significance, of FRN accumulation in cryoconite is currently hindered by a lack of analytical unity with respect to the coherence of data arising from a range of standard operating procedures. For example, various run times are used and, in some cases, but not all, radiological data may be supported by suitable Quality Control (QC) procedures. Therefore, in order to enhance the international comparability of future cryoconite datasets we propose the adoption of a standard operating procedure for FRN measurements in cryoconite, including sample preparation, run times, and QC, to ensure the continuity and quality of this database.

Acknowledgements

Sample collection in Sweden and Iceland was funded by INTERACT Transnational Access and QRA grants awarded to CCC respectively. EP thanks the “Black and Bloom” team who

were funded by NERC, and EB thanks the U.S. National Science Foundation McMurdo Dry Valleys Long-Term Ecological Research Program at Taylor Valley, Antarctica for field assistance. We thank the British Exploring Society for collecting samples in Ladakh on our behalf, Alexandre Anesio for providing samples from Sweden to EP, and Krzysztof Zawierucha for providing data from Norway and Svalbard. PNO acknowledges financial support from the Natural Sciences and Engineering Research Council of Canada and Forest Renewal British Columbia. Samples from Andean glaciers were collected under permission N. 019/2018 issued by CONAF (Chile), and analyses for Alaska, Norway, and the Chilean Andes were supported by the National Science Centre grant no. 2018/31/B/ST10/03057.

Author contributions

CCC devised and led this collaborative study. RA, EB, GB, CCC, JC, AF, RF, DL, EL, PNO, EP, FP NS, NT and PW conducted field sampling, GB, DB, CCC, EL, GEM, MN, PNO, MS and AT conducted sample analysis, and GB, CCC, EL, PNO and PW led interpretation of the collective data. All authors contributed to preparation of the manuscript.

Data availability

Data presented in this manuscript are accessible at http://dx.doi.org/10.17632/6v7pghpw34.2 .

References

Baccolo, G., Di Mauro, B., Massabò, D., Clemenza, M., Nastasi, M., Delmonte, B., Prata, M., Prati, P., Previtali, E., Maggi, V., 2017. Cryoconite as a temporary sink for anthropogenic species stored in glaciers. Sci. Rep. 7, 9623. doi:10.1038/s41598-017-10220-5

Baccolo, G., Nastasi, M., Massabò, D., Clason, C., Di Mauro, B., Di Stefano, E., Łokas, E., Prati, P., Previtali, E., Takeuchi, N., Delmonte, B., Maggi, V., 2020a. Artificial and natural radionuclides in cryoconite as tracers of supraglacial dynamics: Insights from the Morteratsch glacier (Swiss Alps). Catena 191**,** 104577. doi:10.1016/j.catena.2020.104577

Baccolo, G., Łokas, E., Gaca, P., Massabò, D., Ambrosini, R., Azzoni, R. S., Clason, C., Di Mauro, B., Franzetti, A., Nastasi, M., Prata, M., Prati, P., Previtali, E., Delmonte, B. & Maggi, V., 2020b. Cryoconite: an efficient accumulator of radioactive fallout in glacial environments. Cryosphere 14, 657-672, doi:10.5194/tc-14-657-2020

Bagshaw, E.A., Tranter, M., Fountain, A.G., Welch, K., Basagic, H.J., Lyons, W.B., 2013. Do cryoconite holes have the potential to be significant sources of C, N, and P to downstream Depauperate Ecosystems of Taylor Valley, Antarctica? Arctic Antarctic Alpine Res. 45, 440-454. doi:10.1657/1938-4246-45.4.440

Barbante, C., Schwikowski, M., Döring, T., Gäggeler, H. W., Schotterer, U., Tobler, L., Van De Velde, K., Ferrari, C., Cozzi, G., Turetta, A., Rosman, K., Bolshov, M., Capodaglio, G., Cescon, P., Boutron, C., 2004. Historical record of European emissions of heavy metals to the atmosphere since the 1650s from alpine snow/ice cores drilled near Monte Rosa. Environ. Sci. Technol 38, 4085-4090 doi:10.1021/es049759r

Baskaran, M., 2011. Po-210 and Pb-210 as atmospheric tracers and global atmospheric Pb-210 fallout: a Review. J. Environ. Radioact. 102(5), 500-513. doi: 10.1016/j.jenvrad.2010.10.007

Beard, D.B., Clason, C.C., Rangecroft, S., Poniecka, E., Ward, K.J., Blake, W.H., 2022a. Anthropogenic contaminants in glacial environments I: Inputs and accumulation. Prog. Phys. Geog. 46(4), 630-648. doi: 10.1177/03091333221107376

Beard, D.B., Clason, C.C., Rangecroft, S., Poniecka, E., Ward, K.J., Blake, W.H..

, 2022b. Anthropogenic contaminants in glacial environments II: Release and downstream consequences. Prog. Phys. Geog. 46(5), 790-808. doi:10.1177/03091333221127342

Bogdal, C., Schmid, P., Zennegg, M., Anselmetti, F. S., Scheringer, M., Hungerbühler, K., 2009. Blast from the past: Melting glaciers as a relevant source for persistent organic pollutants. Environ. Sci. Technol. 43**,** 8173-8177. doi:10.1021/es901628x

Bradley, J.A., Singarayer, J.S., Anesio, A.M., 2014. Microbial community dynamics in the forefield of glaciers. Proc. R. Soc. B. 281 20140882. doi:10.1098/rspb.2014.0882

Bunzl, K., Kracke, W., 1990. Simultaneous determination of ^{238}Pu, $^{239+240}$Pu, ^{241}Pu, ^{241}Am, ^{241}Cm, ^{244}Cm, ^{89}Sr, and ^{90}Sr in vegetation samples, and application to Chernobyl-fallout contaminated grass. J. Radioanal. Nuclear Chem. 138, 83–91. doi:10.1007/BF02049350

Cambray, R.S., Playford, K., Lewis, G.N.J., Carpenter, R.C., 1990. *Radioactive fallout in air and rain: results to the end of 1988*. AERE R13575, HMSO, United Kingdom

Cao, L., Zhou, Z., Zhang, K., Wang, N., Liu, Z., 2023. Determination of plutonium in cryoconite on glacier surfaces in the northeast Tibetan Plateau: Implications for source identification and accumulation. Sci. Total Environ. 887, 164140. doi:10.1016/j.scitotenv.2023.164140

Chino, M., Nakayama, H., Nagai, H., Terada, H., Katata, G., Yamazawa, H., 2011. Preliminary estimation of release amounts of 131I and 137Cs accidentally discharged from the Fukushima Daiichi Nuclear Power Plant into the atmosphere. J. Nuclear Sci. Tech. 48**,** 1129-1134. doi:10.1080/18811248.2011.9711799

Chmiel, S., Reszka, M., Rysiak, A., 2009. Heavy metals and radioactivity in environmental samples of the Scott glacier region on Spitsbergen in summer 2005. Quaestiones Geographicae 28A/1, 23-29

Clason, C.C., Blake, W.H., Selmes, N., Taylor, A., Boeckx, P., Kitch, J., Mills, S.C., Baccolo, G., Millward, G.E., 2021. Accumulation of legacy fallout radionuclides in cryoconite on Isfallsglaciären (Arctic Sweden) and their downstream spatial distribution. Cryosphere 15, 5151-5168. doi:10.5194/tc-15-5151-2021

Clason, C. Rangecroft, S., Owens, P.N., Łokas, E., Baccolo, G., Selmes, N., Beard, D., Kitch, J., Dextre, R.M., Morera, S., Blake, W., 2023. Contribution of glaciers to water, energy, and food security in mountain regions: current perspectives and future priorities, Annals Glaciol. doi:10.1017/aog.2023.14

Cook, J., Edwards, A., Takeuchi, N., Irvine-Fynn, T., 2016a. Cryoconite: The dark biological secret of the cryosphere. Prog. Phys. Geog. 40, 66-111. doi:10.1177/0309133315616574

Cook, J.M., Hodson, A.J., Irvine-Fynn, T.D., 2016b. Supraglacial weathering crust dynamics inferred from cryoconite hole hydrology. Hydrol. Process. 30(3), 433-446. doi:10.1002/hyp.10602

Donaldson, S., Adlard, B., Odland, J.Ø., 2016. Overview of human health in the Arctic: conclusions and recommendations. Int. J. Circumpolar Health 75, 33807-2. doi: 10.3402/ijch.v75.33807

D'abzac, P., Bordas, F., Joussein, E., Van Hullebusch, E.D., Lens, P.N.L., Guibaud, G., 2013. Metal binding properties of extracellular polymeric substances extracted from anaerobic granular sludges. Environ. Sci. Pollut. Res. Int. 20, 4509-4519. doi:10.1007/s11356-012-1401-3

Edwards, A., Rassner, S.M.E., Anesio, A.M., Worgan, H.J., Irvine-Fynn, T.D.L., Wyn Williams, H., Sattler, B., Wyn Griffith, G., 2013. Contrasts between the cryoconite and ice-marginal bacterial communities of Svalbard glaciers. Polar Res. 32, 19468-9. doi:10.3402/polar.v32i0.19468

European Commission, Directorate-General for Research and Innovation, De Cort, M., Dubois, G., Fridman, S., et al., 2009. *Atlas of caesium deposition on Europe after the Chernobyl accident*, Publications Office.

Evrard, O., Chaboche, P.-A., Ramon, R., Foucher, A., Laceby, J.P., 2020. A global review of sediment source fingerprinting research incorporating fallout radiocesium (137Cs), Geomorphol 362, 107103. doi: 10.1016/j.geomorph.2020.107103

Ferrario, C., Finizio, A., Villa, S., 2017. Legacy and emerging contaminants in meltwater of three Alpine glaciers. Sci. Total Environ. 574, 350-357. doi:10.1016/j.scitotenv.2016.09.067

Foucher, A., Chaboche, P.-A., Sabatier, P., Evrard, O., 2021. A worldwide meta-analysis (1977–2020) of sediment core dating using fallout radionuclides including ^{137}Cs and $^{210}Pb_{xs}$. Earth System Sci. Data. 13(10), 4951-4966. doi: 10.5194/essd-13-4951-2021

Fountain, A.G., Tranter, M., Nylen, T.L., Lewis, K.J., Mueller, D.R., 2004. Evolution of cryoconite holes and their contribution to meltwater runoff from glaciers in the McMurdo Dry Valleys, Antarctica. J. Glaciol. 50(168), 35-45. doi:10.3189/172756504781830312

Franz, T.P., Eisenreich, S.J., 1998. Snow scavenging of polychlorinated biphenyls and polycyclic aromatic hydrocarbons in Minnesota. Environ. Sci. Technol. 32, 1771-1778. doi:10.1021/es970601z

Franzetti, A., Pittino, F., Gandolfi, I., Azzoni, R. S., Diolaiuti, G., Smiraglia, C., Pelfini, M., Compostella, C., Turchetti, B., Buzzini, P., Ambrosini, R., 2020. Early ecological succession patterns of bacterial, fungal and plant communities along a chronosequence in a recently deglaciated area of the Italian Alps. FEMS Microbiol. Ecol. 96. doi:10.1093/femsec/fiaa165

Fujii, K., Ikeda, S., Akama, A., Komatsu, M., Takahashi, M., Kaneko, S., 2014. Vertical migration of radiocesium and clay mineral composition in five forest soils contaminated by

the Fukushima nuclear accident. Soil Sci. Plant Nutrition 60, 751-764. doi:10.1080/00380768.2014.926781

Gadd, G.M., 1996. Influence of microorganisms on the environmental fate of radionuclides. Endeavour 20, 150-156. doi:10.1016/s0160-9327(96)10021-1

Gäggeler, H.W., Tobler, L., Schwikowski, M., Jenk, T.M., 2020. Application of the radionuclide 210Pb in glaciology – an overview. J. Glaciol. 66, 447-456. doi:10.1017/jog.2020.19

Hasholt, B., Walling, D.E., Owens, P.N., 2000. Sedimentation in arctic proglacial lakes: Mittivakkat Glacier, south-east Greenland. Hydrol. Process. 14, 679-699. doi:10.1002/(SICI)1099-1085(200003)14:4%3C679::AID-HYP966%3E3.0.CO;2-E

Herbert, B.M.J., Villa, S., Halsall, C.J. 2006. Chemical interactions with snow: Understanding the behavior and fate of semi-volatile organic compounds in snow. Ecotoxicol. Environ. Saf. 63.3-16, doi:10.1016/j.ecoenv.2005.05.012

Hong, G.H., Baskaran, M., Molaroni, S.M., Lee, H.-M., Burger, J., 2011. Anthropogenic and natural radionuclides in caribou and muskoxen in the Western Alaskan Arctic and marine fish in the Aleutian Islands in the first half of 2000s. Sci. Total Environ. 409, 3638-3648.oi:10.1016/j.scitotenv.2011.06.044

Huang, J., Kang, S., Ma, M., Guo, J., Cong, Z., Dong, Z., Yin, R., Xu, J., Tripathee, L., Ram, K., Wang, F., 2019. Accumulation of atmospheric mercury in glacier cryoconite over western China. Environ. Sci. Technol. 53, 6632-6639. doi:10.1021/acs.est.8b06575

Hugonnet, R., Mcnabb, R., Berthier, E., Menounos, B., Nuth, C., Girod, L., Farinotti, D., Huss, M., Dussaillant, I., Brun, F., Kääb, A., 2021. Accelerated global glacier mass loss in the early twenty-first century. Nature 592, 726-731. doi:10.1038/s41586-021-03436-z

IPCC. 2019. *IPCC Special Report on the Ocean and Cryosphere in a Changing Climate*. *In:* H.-O. Pörtner, D. C. R., V. Masson-Delmotte, P. Zhai, M. Tignor, E. Poloczanska, K. Mintenbeck, A. Alegría, M. Nicolai, A. Okem, J. Petzold, B. Rama, N.M. Weyer (eds.). Cambridge University Press, Cambridge, UK and New York, NY, USA, 755 pp. doi:10.1017/9781009157964

Leggat, M.S., Owens, P.N., Stott, T.A., Forrester, B.J., Dery, S.J., Menounos, B., 2015. Hydro-meteorological drivers and sources of suspended sediment flux in the pro-glacial zone of the retreating Castle Creek Glacier, Cariboo Mountains, British Columbia, Canada. Earth Surface Process. Landf. 40(11), 1542-1559. doi:10.1002/esp.3755

Li, Q., Kang, S., Wang, N., Li, Y., Li, X., Dong, Z. & Chen, P. 2017. Composition and sources of polycyclic aromatic hydrocarbons in cryoconites of the Tibetan Plateau glaciers. *Sci Total Environ,* 574**,** 991-999, doi:10.1016/j.scitotenv.2016.09.159

Lizaga, I., Gaspar, L., Quijano, L., Le'con, G., Navas, A., 2019. NDVI, ^{137}Cs and nutrients for tracking soil and vegetation development on glacial landforms in the Lake Parón Catchment (Cordillera Blanca, Perú). Sci. Total Environ. 651(1), 250-260. doi:10.1016/j.scitotenv.2018.09.075

Łokas, E., Bartmiński, P., Wachniew, P., Mietelski, J. W., Kawiak, T., Środoń, J., 2014. Sources and pathways of artificial radionuclides to soils at a High Arctic site. Environ. Sci. Pollut. Res. Int. 21, 12479-12493. doi:10.1007/s11356-014-3163-6

Łokas, E., Zaborska, A., Kolicka, M., Różycki, M., Zawierucha, K., 2016. Accumulation of atmospheric radionuclides and heavy metals in cryoconite holes on an Arctic glacier. Chemosphere. 160, 162-172. doi:10.1016/j.chemosphere.2016.06.051

Łokas, E., Wachniew, P., Jodłowski, P., Gąsiorek, M., 2017. Airborne radionuclides in the proglacial environment as indicators of sources and transfers of soil material. J. Environ. Radioact. 178-179. 193-202, doi:10.1016/j.jenvrad.2017.08.018

Łokas, E., Zawierucha, K., Cwanek, A., Szufa, K., Gaca, P., Mietelski, J.W., Tomankiewicz, E., 2018. The sources of high airborne radioactivity in cryoconite holes from the Caucasus (Georgia). Sci. Rep. 8, 10802-10. doi:10.1038/s41598-018-29076-4

Łokas, E., Zaborska, A., Sobota, I., Gaca, P., Milton, J.A., Kocurek, P., Cwanek, A., 2019. Airborne radionuclides and heavy metals in high Arctic terrestrial environment as the indicators of sources and transfers of contamination. Cryosphere 13, 2075-2086. doi:10.5194/tc-13-2075-2019

Łokas, E., Wachniew, P., Baccolo, G., Gaca, P., Janko, K., Milton, A., Buda, J., Komedera, K., Zawierucha, K., 2022. Unveiling the extreme environmental radioactivity of cryoconite from a Norwegian glacier. Sci. Total Environ. 814, 152656. doi:10.1016/j.scitotenv.2021.152656

Meyers, R.A. (ed.). 2012. *Encyclopedia of Sustainability Science and Technology*. Springer New York, doi:10.1007/978-1-4419-0851-32021.

Meusburger, K., Evrard, O., Alewell, C., Borrelli, P., Cinelli, G., Ketterer, M., Mabit, L., Panagos, P., van Oost, K., Ballabio, C. 2020. Plutonium aided reconstruction of caesium atmospheric fallout in European topsoils. Sci. Rep. 10, 11858. doi:10.1038/s41598-020-68736-2

Muravitsky, A.V., Razbudey, V.F., Tokarevsky, V.V., Vorona, P.N., 2005. Time-dependent 241Am activity in the environment from decay of 241Pu released in the Chernobyl accident. Appl. Radiat. Isot. 63, 487-492. doi:10.1016/j.apradiso.2005.03.018

Nagar, S., Antony, R., Thamban, M., 2021. Extracellular polymeric substances in Antarctic environments: A review of their ecological roles and impact on glacier biogeochemical cycles. Polar Sci. 30, 100686. doi:10.1016/j.polar.2021.100686

Nakao, A., Nakao, A., Tanaka, R., Ogasawara, S., Yanai, J., 2015. Aeolian-dust-derived micaceous minerals control radiocesium retention in andosols in Japan. Soil Sci. Soc. Amer. J. 79(6), 1590-1600. doi:10.2136/sssaj2015.05.0173

Navas, A., Oliva, M., Ruiz-Fernández, J., Gaspar, L., Quintano, L., Lizaga, I., 2017. Radionuclides and soil properties as indicators of glacier retreat in a recently deglaciated permafrost environment of the Maritime Antarctica. Sci. Total Environ. 609, 192-204. doi:10.1016/j.scitotenv.2017.07.115

Navas, A., Lizaga, I., Gaspar, L., Latorre, B., Dercon, G., 2020. Unveiling the provenance of sediments in the moraine complex of Aldegonda Glacier (Svalbard) after glacial retreat using radionuclides and elemental fingerprints. Geomorphol. 367, 107304. doi:10.1016/j.geomorph.2020.107304

Olivier, S., Bajo, S., Fifield, L.K., Gäggeler, H.W., Papina, T., Santschi, P.H., Schotterer, U., Schwikowski, M., Wacker, L., 2004. Plutonium from global fallout recorded in an ice core from the Belukha Glacier, Siberian Altai. Environ. Sci. Technol. 38, 6507-6512. doi:10.1021/es0492900

Owens, P.,N., Blake, W.H., Millward, G.E., 2019. Extreme levels of fallout radionuclides and other contaminants in glacial sediment (cryoconite) and implications for downstream aquatic ecosystems. Sci. Rep. 9, 12531. doi:10.1038/s41598-019-48873-z

Owens, P.N., Stott, T.A., Blake, W.H., Millward, G.E., 2023. Legacy radionuclides in cryoconite and proglacial sediment on Orwell Glacier, Signy Island, Antarctica. J.Environ. Radioact. 264, 107206. doi:10.1016/j.jenvrad.2023.107206

Pavlova, P.A., Zennegg, M., Anselmetti, F.S., Schmid, P., Bogdal, C., Steinlin, C., Jäggi, M., Schwikowski, M., 2016. Release of PCBs from Silvretta glacier (Switzerland) investigated in lake sediments and meltwater. Environ. Sci. Pollut. Res. Int. 23, 10308-10316. doi:10.1007/s11356-015-5854-z

Pereira, S., Micheletti, E., Zille, A., Santos, A., Moradas-Ferreira, P., Tamagnini, P., De Philippis, R., 2011. Using extracellular polymeric substances (EPS)-producing cyanobacteria for the bioremediation of heavy metals: do cations compete for the EPS functional groups and also accumulate inside the cell? Microbiology 157, 451-458. doi:10.1099/mic.0.041038-0

Persson, B.R.R., Holm, E., 2011. Polonium-210 and lead-210 in the terrestrial environment: a historical review. J. Environ. Radioact. 102(5), 420-429. doi:10.1016/j.jenvrad.2011.01.005

Rangarajan, C., Madhavan, R., Gopalakrishnan, S.S., 1986. Spatial and temporal distribution of lead-210 in the surface layers of the atmosphere. J. Environ. Radioact. 3, 23-33. doi:10.1016/0265-931X(86)90047-0

Rogiers, T., Claesen, J. R., Van Gompel, A., Vanhoudt, N., Mysara, M., Williamson, A., Leys, N., Van Houdt, R., Boon, N., Mijnendonckx, K., 2021. Soil microbial community structure and functionality changes in response to long-term metal and radionuclide pollution. Environ. Microbiol. 23, 1670-1683. doi:10.1111/1462-2920.15394

Sayre, R., Karagulle, D., Frye, C., Boucher, T., Wolff, N.H., Breyer, S., Wright, D., Martin, M., Butler, K., Van Graafeiland, K., Touval, J., Sotomayor, L., McGowan, J., Game, E.T., Possingham, H., 2020. An assessment of the representation of ecosystems in global protected areas using new maps of World Climate Regions and World Ecosystems. Global Ecol. Conserv. 21, e00860. doi:10.1016/j.gecco.2019.e00860

Schmid, P., Bogdal, C., Blüthgen, N., Anselmetti, F. S., Zwyssig, A., Hungerbühler, K., 2011. The missing piece: sediment records in remote mountain lakes confirm glaciers being

secondary sources of persistent organic pollutants. Environ. Sci. Technol. 45.03-208, doi:10.1021/es1028052

Simonoff, M., Sergeant, C., Poulain, S., Pravikoff, M.S., 2007. Microorganisms and migration of radionuclides in environment. Comptes rendus. Chimie 10, 1092-1107. doi:10.1016/j.crci.2007.02.010

Steinhauser, G., Brandl, A., Johnson, T.E., 2014. Comparison of the Chernobyl and Fukushima nuclear accidents: A review of the environmental impacts. Sci. Total Environ. 470-471, 800-817., doi:10.1016/j.scitotenv.2013.10.029

Stensrud, A.B., 2016. Climate change, water practices and relational worlds in the Andes. Ethnos 81, 75-98. doi:10.1080/00141844.2014.929597

Stevens, I.T., Irvine-Fynn, T.D.L., Porter, P.R., Cook, J.M, Edwards, A., Smart, M., Moorman, B.J., Hodson, A.J., Mitchell, A.C., 2018. Near-surface hydraulic conductivity of northern hemisphere glaciers. Hydrol. Process. 32(7), 850-865. doi:/10.1002/hyp.11439

Stocki, T.J., Gamberg, M., Loseto, L., Pellerin, E., Bergman, L., Mercier, J.F., Genovesi, L., Cooke, M., Todd, B., Sandles, D., Whyte, J., Wang, X., 2016. Measurements of cesium in Arctic beluga and caribou before and after the Fukushima accident of 2011. J. Environ. Radioact. 162-163, 379-387. doi:10.1016/j.jenvrad.2016.05.023

Stohl, A., 2006. Characteristics of atmospheric transport into the Arctic troposphere. J. Geophys. Res. 111.11306-n/a, doi:10.1029/2005JD006888

Takeuchi, N., Nishiyama, H., Li, Z., 2010. Structure and formation process of cryoconite granules on Ürümqi glacier No. 1, Tien Shan, China. Ann. Glaciol. 51, 9-14. doi:10.3189/172756411795932010

Teramage, M.T., Onda, Y., Kato, H., Wakiyama, Y., Mizugaki, S., Hiramatsu, S., 2013. The relationship of soil organic carbon to ^{210}Pbex and ^{137}Cs during surface soil erosion in a hillslope forested environment. Geoderma 192, 59-67. doi:10.1016/j.geoderma.2012.08.030

Thakur, P., Ward, A.L., 2018. ^{241}Pu in the environment: insight into the understudied isotope of plutonium. J. Radioanal. Nuclear Chem. 317, 757-778. doi:10.1007/s10967-018-5946-6

Tieber, A., Lettner, H., Bossew, P., Hubmer, A., Sattler, B., Hofmann, W., 2009. Accumulation of anthropogenic radionuclides in cryoconites of Alpine glaciers. J. Environ. Radioact. 100.90-598, doi:10.1016/j.jenvrad.2009.04.008

UNSCEAR. 2000. *Sources And Effects Of Ionizing Radiation*. United Nations Publications, New York.

Walther, C., Gupta, D.K. (eds.). 2015. *Radionuclides in the Environment*, Springer International. Springer International Publishing, Switzerland, doi:10.1007/978-3-319-22171-7_5

Wilflinger, T., Lettner, H., Hubmer, A., Bossew, P., Sattler, B., Slupetzky, H., 2018. Cryoconites from Alpine glaciers: Radionuclide accumulation and age estimation with Pu and Cs isotopes and ^{210}Pb. J. Environ. Radioact. 186, 90-100. doi:10.1016/j.jenvrad.2017.06.020

Zaborska, A., Carroll, J., Papucci, C., Pempkowiak, J., 2007. Intercomparison of alpha and gamma spectrometry techniques used in ^{210}Pb geochronology. J. Environ. Radioact 93(1), 38-50. doi:10.1016/j.jenvrad.2006.11.007

Zhang, F., Wang, J., Baskaran, M., Zhong, Q., Wang, Y., Paatero, J., Du, J., 2021. A global dataset of atmospheric ^{7}Be and ^{210}Pb measurements: annual air concentration and depositional flux. Earth System Sci. Data 13(6), 2963-2994. doi:10.5194/essd-13-2963-2021

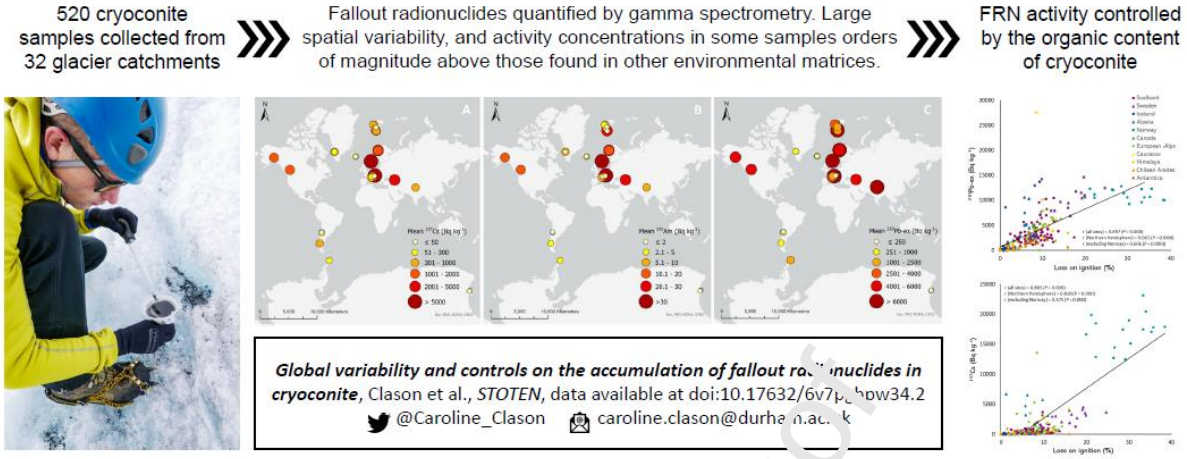
Declaration of interests

☐ 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.

☒ The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Caroline Clason reports financial support was provided by INTERACT. Caroline Clason reports financial support was provided by Quaternary Research Association. Ewa Poniecka reports financial support was provided by Natural Environment Research Council. Ewa Poniecka reports travel was provided by National Science Foundation. Philip Owens reports financial support was provided by Natural Sciences and Engineering Research Council of Canada. Philip Owens reports financial support was provided by Forest Renewal British Columbia. Edyta Lokas reports financial support was provided by National Science Centre Poland.

Graphical abstract



Highlights:

- Accumulation of fallout radionuclides is commonplace in cryoconite on glaciers around the world.
- There is large inter-regional variability in fallout radionuclide activity concentrations in cryoconite.
- Fallout radionuclides concentrations can be orders of magnitude higher than found in other environmental matrices.
- The organic content of cryoconite plays a central role in its ability to accumulate fallout radionuclides.