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Radiological and elemental composition of cryoconite and glacier mice from Vatnajökull, Iceland

EMMA SMITH[1], CAROLINE C. CLASON[2*], GEOFFREY MILLWARD[1], ALEX TAYLOR[1] & RALPH FYFE[1]

[1]*School of Geography, Earth and Environmental Science, University of Plymouth, Drake Circus, Plymouth, PL4 8AA, United Kingdom*

[2]*Department of Geography, Durham University, Lower Mountjoy, South Road, Durham, DH1 3LE, United Kingdom*

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

Abstract

Cryoconite has been demonstrated to be an efficient accumulator of some classes of contaminants on glaciers in both mountain and polar environments, however the accumulation of contaminants in cryoconite in Iceland has received very little attention to date. To understand the spatial variability of natural and anthropogenic fallout radionuclides and metals on glaciers in Iceland, we present the first study of this region including both cryoconite from three glaciers: Virkisjökull; Skaftafellsjökull; and Falljökull, together with moss balls ('glacier mice') from Falljökull. The cryoconite samples and glacier mice were analysed using XRF spectrometry to assess their elemental composition and gamma spectrometry to identify, and quantify, fallout radionuclides, primarily ^{7}Be, ^{137}Cs, ^{241}Am, excess ^{210}Pb, and ^{40}K. The results revealed that the cryoconite samples had similar compositions, influenced by local geology and natural sources of volcanic ash and dust. Higher concentrations of radionuclides and heavy metals were found in both cryoconite and glacier mice compared to control samples comprising

nearby proglacial sediments. In comparison to other glaciers in the Northern Hemisphere, however, cryoconite from Icelandic glaciers contains some of the lowest activity concentrations of key radionuclides. Consequently, cryoconite deposits that are released and diluted during the melt and retreat of Icelandic glaciers are unlikely to be of environmental concern following transport to proglacial areas.

Keywords: cryoconite; fallout radionuclides; metals; Iceland; glacier mice

1. Introduction

Cryoconite is a dark, granular solid that consists mainly of fine mineral particles (typically 60-99%), with the remaining composition attributable to organic material and living microorganisms (Cook et al, 2016; Clason et al., 2023a). Cryoconite has now been shown to accumulate significant concentrations of fallout radionuclides (FRNs) and heavy metals across mountain and polar regions globally (Rozwalak et al., 2022; Clason et al., 2023a), even in remote, high latitude locations of the Southern Hemisphere (Buda et al., 2020; Owens et al., 2023). In many sites FRNs have been detected in cryoconite with activity concentrations elevated well above local soil and sediment backgrounds (Łokas *et al*., 2016; 2017; 2018; 2019; 2022; Baccolo *et al*., 2017; 2020a;b; Owens *et al*., 2019; 2023; Clason et al., 2021). Mineral particles found in cryoconite are mostly sourced from local substrates and wind-blown dust, while organic matter is largely derived *in situ* from photosynthetic microbes that inhabit glaciers (Zawierucha *et al*., 2016). Furthermore, cyanobacteria play an important role in the formation of cryoconite granules (Wejnerowski et al, 2023), since they produce extracellular polymeric substances which have properties that contribute to the accumulation of dust and micro-organisms. The chemical reactivity of cryoconite granules also leads to their accumulation of FRNs, trace metals, metalloids, and nutrients such as phosphorus (Beard et

al., 2022; Clason et al., 2023a). Cryoconite is normally found either as a material dispersed on the surface of glaciers or at the base of cryoconite holes in the ablation zone (Owens *et al*, 2019; Baccolo *et al*., 2020a;b). The dark colour of cryoconite also acts to reduce the albedo of glacier surfaces, thereby contributing to enhanced melting of ice beneath the cryoconite layer, promoting the formation of cryoconite holes and contributing to ice surface melting (Takeuchi, 2002).

Anthropogenic FRNs such as ^{137}Cs have been released into the global environment, mainly in the Northern Hemisphere, since the second half of the 20th century (Łokas *et al*, 2019) as a result of atmospheric nuclear weapons testing, nuclear accidents, and the fragmentation of satellites (Steinhauser et al., 2014). However, the presence of ^{241}Am in the environment is a result of ^{241}Pu decay leading to increased ^{241}Am activity which is predicted to peak around 2100 (Baccolo *et al.*, 2020a). The presence of heavy metals in cryoconite results from their increased deposition, by approximately tenfold, since the start of the industrial era (c.1850) as a result of mining, industry, fuel burning, agriculture, transportation and waste disposal (Łokas *et al*., 2019). The atmosphere, oceanic currents, and rivers are regarded as significant transport pathways for artificial radionuclides and heavy metals (Łokas *et al*., 2019). Airborne FRNs, possibly originating from the stratosphere (Corcho Alverado *et* al., 2014), attach onto aerosols or fine dusts (Wilflinger *et al*., 2018), which are ultimately deposited on glacier surfaces. Particulate fallout is aided by snowfall which effectively scavenges trace constituents from the atmosphere (Singh et al., 2013). During the melt season water flows across the surface of glaciers, particularly in the ablation zone where cryoconite is commonly found (Owens *et al*., 2019). Hence, meltwater acts to mobilise and transport contaminants previously stored in the snow and ice, which are accumulated in cryoconite deposits due to its chemically reactive properties (Baccolo *et al*., 2017; 2020a;b; Owens *et al*., 2019).

Glaciers and ice sheets cover about 10% of Earth's surface, and act as an important source of freshwater for downstream ecosystems and populations (Clason et al., 2023b). Understanding the spatial variability of natural and anthropogenic contaminants in glaciated catchments is important with respect to both the long-term health of the polar environment and for assessing pressures on water quality. Cryoconite is one of the most radioactive of natural materials, excluding those found in nuclear exclusion zones (Baccolo *et al*., 2017). When glaciers melt and retreat, cryoconite is released into the proglacial environment. Consequently, the raised activity concentrations of FRNs found in cryoconite may act as a secondary source of radioactive contaminants downstream (Owens *et al*., 2019), with the potential for pollution of local vegetation, aquatic habitats, and drinking water. Furthermore, the release of particulate radionuclides from glaciers into downstream ecosystems could be taken up by biota, allowing transfer within trophic chains causing contamination in higher species such as reindeer (Łokas *et al*., 2016; 2019). FRNs may also be taken up into other components of the supraglacial ecosystem, such as 'glacier mice', little-studied ovate moss balls that are rich in microbial life, contain a range of moss species, and have been observed to move across glacier surfaces in a 'herd-like' fashion (Coulson & Midgely, 2012; Hotaling et al., 2020). It is thus important to investigate the properties of materials in the transient supraglacial ecosystem to understand whether the concentrations of the natural and anthropogenic contaminants it contains could impact environments downstream. Although there are now numerous reports of the concentrations of natural and anthropogenic contaminants in cryoconite from glaciers across the globe (Rozwalak et al., 2022), Icelandic glaciers are a notable exception.

In this contribution we report the elemental compositions and activity concentrations of radionuclides in cryoconite samples collected from the ice surfaces of Virkisjökull,

Skaftafellsjökull and Falljökull in Iceland. We also contribute new knowledge of the elemental composition and activity concentrations of radionuclides in samples of glacier mice from Falljökull, and explore the variability of natural and anthropogenic contaminants together with the factors giving rise to their distribution.

2. Study area

Virkisjökull, Skaftafellsjökull, and Falljökull are situated in south-east Iceland (Figure 1), and are all outlet glaciers that descend from the Vatnajökull ice cap (Everest *et al.*, 2017). Virkisjökull and Falljökull flow southwest from their source at over 1500 m above sea level (asl) down to ~150 m asl. They are considered twin valley glaciers which separate around the Rauðikambur nunatak at ~600 m asl (Phillips *et al.*, 2014; Everest *et al*., 2017) before reconnecting at ~210 m asl and terminating at ~130 m asl at a proglacial lake. Skaftafellsjökull descends south from the Vatnajökull ice cap (Vilmundardóttir *et al*., 2014) and terminates at ~120 m asl. All three are temperate glaciers, and in recent decades have undergone a period of rapid retreat in response to warmer summers and milder winters (Phillips et al., 2014; Evans *et al*., 2017). Approximately 60% of Iceland's glaciers overlay active volcanoes, for example there are seven volcanoes located under the Vatnajökull ice cap (Benediktsson et al., 2024), with Grímsvötn, Katla, and Öræfajökull notably having deposited tephra over Skaftafellsjökull and nearby areas (Vilmundardóttir *et al*., 2014). The proglacial regions of Iceland's glaciers are typified by significant *sandur* such as Skeiðarársandur to the south of Vatnajökull (Arnalds *et al*., 2016), and Iceland experiences up to 135 days of dust storms annually. The area north of Vatnajökull experiences desert-like conditions, while the ice cap can have up to 4000 mm of precipitation annually (Einarsson, 1984). The predominant wind direction is from the east, and the geology consists mainly of igneous basaltic rocks (Vilmundardóttir *et al*., 2014).

The sea surrounding Iceland is one of the least polluted ocean areas of the world (Ministry of Environment, Iceland, 2006). The low contamination status is a result of its remote geographical location, its small population, and a relatively low number of industries releasing insignificant quantities of contaminants into the aquatic environment. The main industrial sources of pollutants are the shipyards, a tannery, a zinc-chrome coating plant, and aluminium and ferrosilicon plants (Ministry of Environment, Iceland, 2006). Contamination from heavy metals, radionuclides and other pollutants is thus largely transported from sources beyond Iceland.

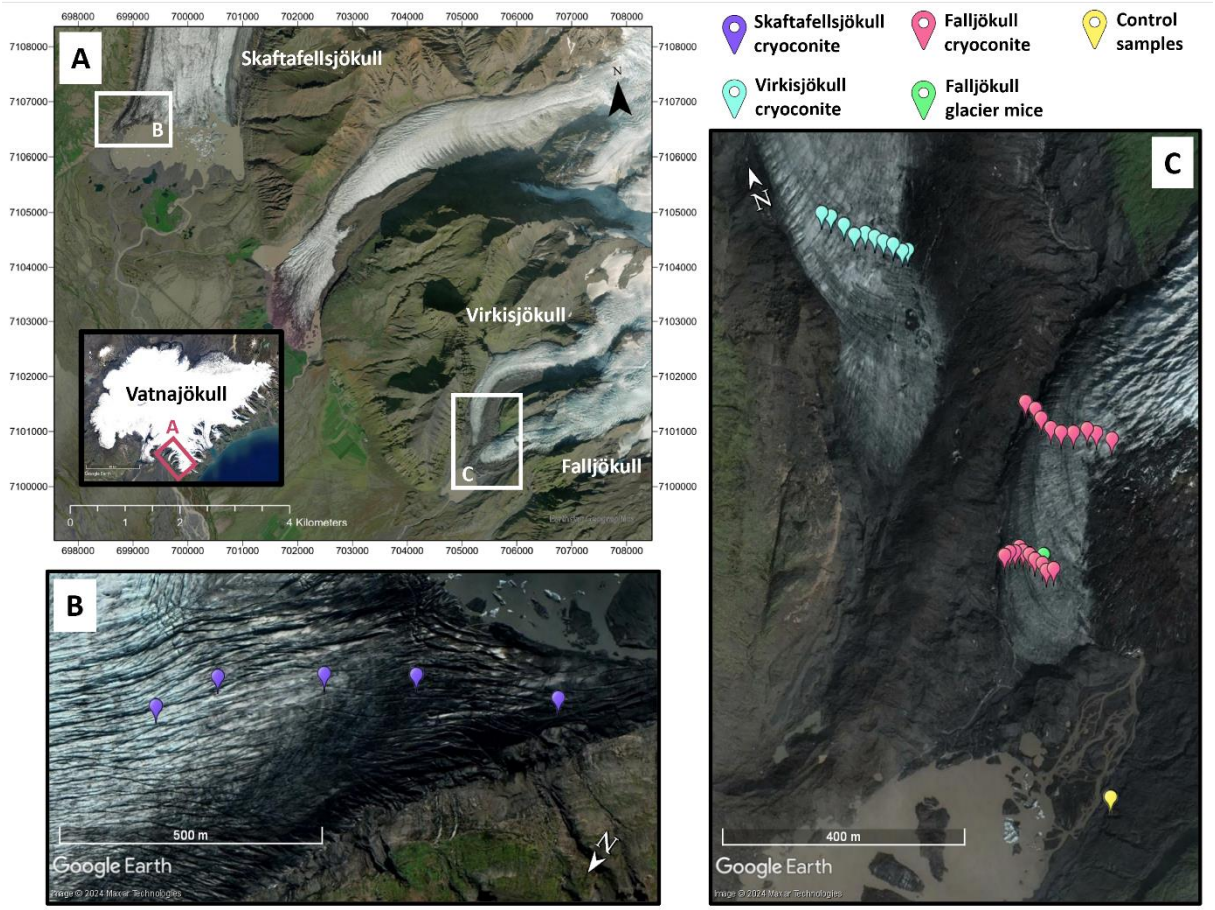

Figure 1. (a) Study site including the position of outlet glaciers on the Vatnajökull ice cap; (b) cryoconite sampling locations on Skaftafellsjökull (purple); (c) cryoconite sampling locations on Virkisjökull (blue), cryoconite (pink) and glacier mice (green) sampling locations on Falljökull, and locations of the control samples (yellow).

3. Methods

3.1 Sample collection and preparation

A total of 10 cryoconite samples were retrieved from the surface of Virkisjökull, 5 from Skaftafellsjökull, and 19 from Falljökull (Fig, 1) in July 2018, to characterise intra- and inter-glacier variability in elemental composition and FRN content. Three control samples (proglacial sediment) were also collected from an off-ice site in the Virkisjökull-Falljökull forefield. Control samples were selected to provide a comparison of typical geochemistry within sediments disconnected from glacial meltwater drainage pathways. Cryoconite samples were collected from the ice surface or from within water-filled cryoconite holes using a plastic pipette or a small plastic spatula, and control samples collected using a plastic trowel. Samples were stored in water-tight containers for transportation to the Consolidated Radioisotope Facility (CORIF) at the University of Plymouth, UK where they were freeze dried prior to geochemical and radiological analysis. Moss balls (glacier mice) were also present in abundance on the surface of Falljökull, and three were collected and preserved in bulk. On arrival at CORIF the cryoconite samples were prepared immediately for radiological analysis in order to capture the short-lived isotope ^{7}Be ($t_{1/2}$ = 54 days). The cryoconite samples were sieved, using a plastic mesh designed to isolate the <63µm fraction. The cryoconite solids and moss ball samples were freeze-dried for 48 h. Dried cryoconite samples were powdered by hand using a mortar and pestle. Loose particulate matter in the strands of the moss balls was shaken out and disaggregated using a plastic knife for subsequent counting. The prepared samples were stored in acid-cleaned plastic bags in a refrigerator held at -30ºC.

3.2 X-Ray Fluorescence (XRF) spectrometry

The geochemical compositions of the cryoconite and control samples were analysed by XRF as loose powders which were packed into 40 mm diameter cups fitted with a 6 µm polypropylene spectromembrane (Chemplex, USA). All samples were packed to the same volume and left to settle for 24 hours prior to analysis. Analyses were undertaken by wavelength-dispersive X-Ray fluorescence (WD XRF) spectrometry (Axios Max, PANalytical, Netherlands). The instrument was operated at 4 kW using a Rh target X-ray tube. During sequential analysis of elements tube settings ranged from 25 kV, 160 mA for low atomic weight elements up to 60 kV, 66 mA for higher atomic weight elements. All analyses were undertaken using the Omnian analysis application (PANalytical, Netherlands) under He gas. Repeatability of the approach was assessed by repacking and analysing cryoconite samples in triplicate with relative standard deviation found to be <10% across triplicates. This analytical procedure has been validated by Clason et al. (2021) whereby an inter-comparison of results obtained from a calibrated ICP-OES procedure which showed that XRF-Derived concentrations were in close agreement, within 15% relative to ICP-OES for the elements of interest.

3.3 Gamma spectrometry

The freeze-dried cryoconite samples were packed and sealed in gas tight plastic vials of 4 ml volume containing up to 7 g of particulate sample. The prepared moss ball samples were packed into aluminium boxes each holding approximately 25 g of solid phase. All samples were incubated for 21 days to allow establishment of radioactive equilibrium between constituents of the ^{238}U decay scheme. Subsequently, activity concentrations of the target radionuclides were determined using a well High Purity Germanium (HPGe) gamma spectrometer (EG&G Ortec GWL-170-15-S) built with an ultra-low background specification, particularly for ^{210}Pb detection. The gamma spectrometer was calibrated using a low-background soil spiked with a

certified mixed radioactive standard containing 12 radionuclides covering the gamma energy range 46 to 1850 keV (#80717-669 supplied by Eckert & Ziegler Analytics, Georgia, USA). All calibration relationships were derived using EG&G GammaVision software. All samples were counted for a minimum of 24h. Total ^{210}Pb was measured by its gamma emissions at 46.5 keV and its unsupported component calculated by the subtraction of the ^{226}Ra activity, which in turn was measured by the gamma emissions of ^{214}Pb at 295 and 352 keV, ^{137}Cs was determined by its gamma emissions at 662 keV and ^{241}Am by the line at 59.4 keV. The stability of the instrument was verified by undertaking regular quality control analyses of reference materials, namely moss soil IAEA-2009-03, soil IAEA-TEL-2012-03, and spruce needles IAEA-TEL-2016-03 (IAEA, Vienna, Austria) (Table A1). All activity concentrations were decay corrected to the date of sampling and uncertainties were derived from counting statistics and reported at ±*2 sigma*.

Loss on ignition (LOI) for each sample was conducted by estimating the loss in weight following the heating of accurately weighed samples contained in porcelain crucibles to 600^{o}C for 6 hours, following the procedure adopted by Łokas et al. (2022).

3.4 Calculation of Enrichment Factors

To emphasise the differences in the elemental concentrations from the various sample sites, concentrations for both cryoconite and the control samples were normalised in terms of the Enrichment Factor (EF), defined as:

$$EF = \frac{\frac{M_m}{Al_m}}{\frac{M_s}{Al_s}} \qquad (1)$$

where M_m and Al_m are the measured metal and Al concentrations in cryoconite samples, respectively, and M_s and Al_s are the metal and Al concentrations applicable to the composition of the upper continental crust (Rudnick & Gao, 2014). $1 < EF < 3$ specifies minor enrichment, $3 < EF < 5$ specifies moderate enrichment, $5 < EF < 10$ specifies moderate to severe enrichment, and $EF > 10$ is classed as severe enrichment.

4. Results

4.1 Inorganic composition of Icelandic cryoconite

The analytical data is summarised as the mean percentages of the chemical composition of cryoconite and the control samples (Figure 2). The sum of the concentrations from the XRF spectrometric analyses adds up to ~93% as a result of other minor constituents in the sample that were not determined. The mean concentrations of major constituents SiO_2, Al_2O_3 and CaO were higher in the control samples compared to the cryoconite samples (510,300; 215,300; 112,100 mg kg^{-1} and 428,900; 142,400; 82,800 mg kg^{-1} respectively), while cryoconite had higher mean concentrations of Fe_2O_3 and P_2O_5 compared to the controls (156,900; 3995 mg kg^{-1} and 116,300; 2826 mg kg^{-1} respectively). For the minor constituents (Figure 3), cryoconite from Virkisjökull had the highest concentrations of Ti (22,160 mg kg^{-1}), and Zr (340 mg kg^{-1}), cryoconite from Skaftafellsjökull had the greatest concentrations of Ni, Zn and Cu (89; 149 and 129 mg kg^{-1}), and Falljökull cryoconite contained the highest concentrations of Nb (38 mg kg^{-1}). Cryoconite from Virkisjökull and Falljökull contained similar concentrations of Cu and Ni (92; 59 and 82; 55 mg kg^{-1} respectively), which were both below the means for the control samples (91 and 63 mg kg^{-1}). The mean concentration of Co in the control samples (171 mg kg^{-1}) was notably lower than for cryoconite from Falljökull, Skaftafellsjökull and Virkisjökull (218; 229 and 241 mg kg^{-1} respectively).

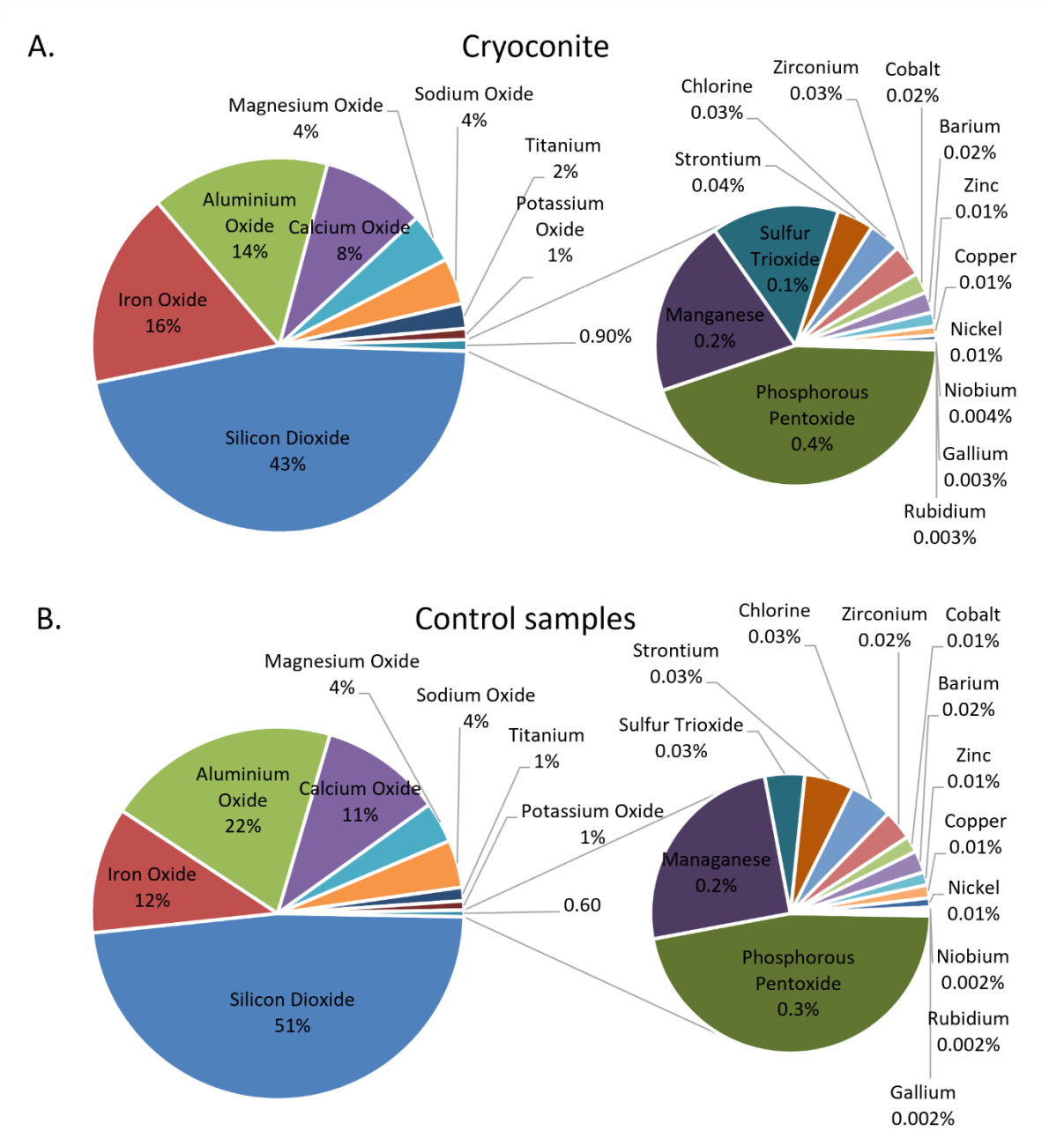

Figure 2. a) Mean composition of cryoconite samples from Iceland; b) mean composition of control samples.

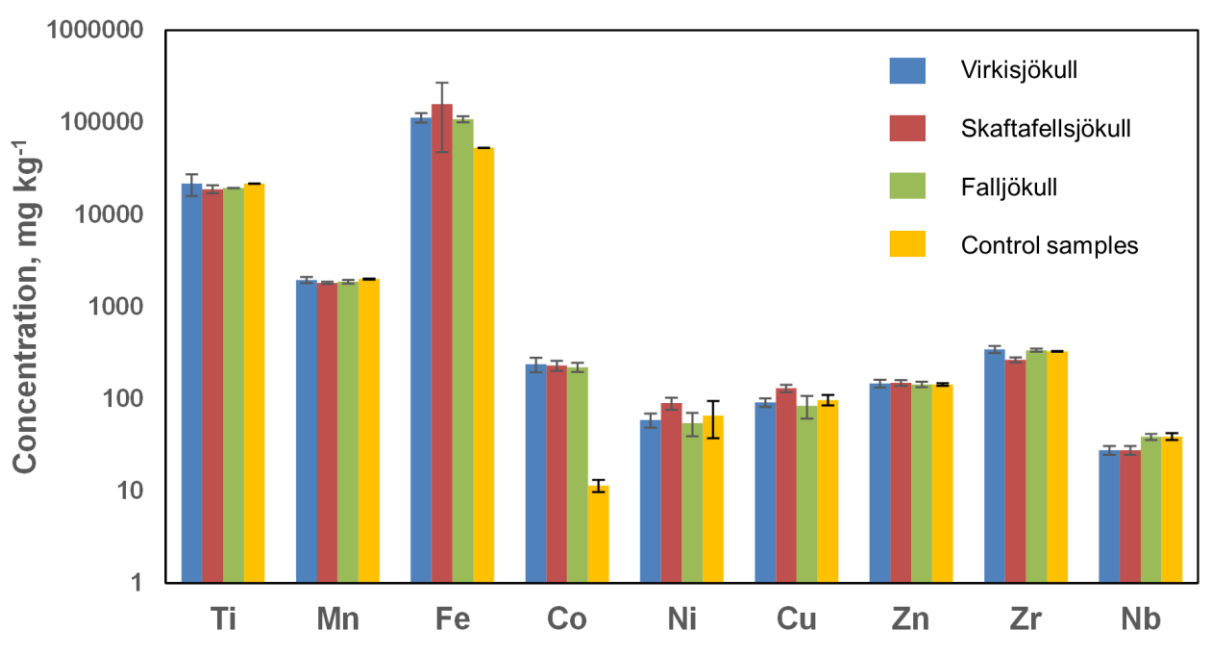

Figure 3. Concentrations of selected elements (mg kg^{-1}) determined in Icelandic cryoconite arranged in increasing atomic number: Virkisjökull (blue; n=10); Skaftafellsjökull (red; n=5); Falljökull (green; n=19); control samples (yellow; n=3).

The Enrichment Factor (EF) values for elements in cryoconite samples from Icelandic glaciers ranged from 1.3 to 5.3 – minor to moderate enrichment (Table 1). The highest EF values were found for Cu, followed by Ti and Ni, whereas lower EFs were obtained for Zn. Other elements, not shown, including Rb, Ba and Sr, had EFs<1. Similarly, control samples from ~280 m distal to the terminus of Falljökull had EF values that were similar to, or in some cases slightly greater than, the cryoconite values, especially for Ni and Cu. However, the enrichment of some metals in cryoconite from Iceland is significantly lower than the values for cryoconite from Sweden, as shown in Table 1 (Clason et al., 2021).

Table 1. Enrichment factors estimated using Eq. 1, based on the measured concentrations. The values for Icelandic cryoconite are compared to those obtained by Clason et al. (2021) for cryoconite from Sweden.

	Cr	**Cu**	**Fe***	**Ni**	**Pb**	**Ti**	**Zn**
Concentration (mg kg^{-1}) - Icelandic cryoconite	67±22	102±24	$1.1x10^5$	67±19	29±19	$20±1.5x10^3$	141±3
Enrichment Factors - Icelandic cryoconite	1.4±0.5	5.3±1.2	2.7±0.1	2.8±0.7	1.3±0.7	4.6±0.7	2.1±0.1
Enrichment Factors - Icelandic controls	1.7±0.2	6.4±2	1.3±0.0	3.4±0.2	ND	4.0±0.7	1.8±0.1
Enrichment Factors - Swedish cryoconite (Clason et al., 2021)	8.1 ± 0.9	15±5	3.2±0.3	9.2±1.3	17.1±4.4	3.2±0.1	4.0±0.5

*The full concentration value for Fe is $1.1x10^5 \pm 0.015x10^5$ mg kg^{-1}.

4.2 Radiological composition of Icelandic cryoconite

The cryoconite samples analysed here may be classed as having fine granules with a dark tone (Rozwalak et al., 2022; Figure A1). Total activity concentrations of radionuclides (Bq kg^{-1}) in cryoconite samples were highly variable (Figs. 4 and 5). The Virkisjökull samples had relatively low, close to background ^{137}Cs activity concentrations while the Skaftafellsjökull samples had significantly enhanced, but variable, ^{137}Cs activities (Figs. 4a; d). The activities of ^{137}Cs in the Falljökull samples had values intermediate to the other two sites (Figure 4g). The activity concentrations of excess ^{210}Pb ($^{210}Pb_{ex}$), where $^{210}Pb_{ex} = {}^{210}Pb_{total} - {}^{214}Pb$ (Figs 4b; e; h), were significantly higher than the ^{137}Cs activities where elevated values were obtained for the Skaftafellsjökull samples. Fewer samples had activities of ^{241}Am above the detection limit, with only ~50% of Falljökull samples having detectable ^{241}Am (Figure 4c; f; i). The maximum

activity concentrations for these radionuclides were coincident for some samples, for example in sample 2 from Virkisjökull, sample 1 from Skaftafellsjökull and sample 5 from Falljökull. This suggests that the activity concentrations of atmospheric fallout particulate matter were well mixed prior to deposition. The solids collected from the control site (n=3) had significantly lower activity concentrations for the key radionuclides compared to the cryoconite samples, such that the mean activity concentration was 2.9 ± 1.1 Bq kg^{-1} for ^{137}Cs; <12.4 ± 10.1 Bq kg^{-1} for ^{210}Pb, whereas ^{214}Pb and ^{241}Am were below detection in all control samples.

The activity concentrations for ^{7}Be were highly variable, with relatively low values for Skaftafellsjökull and marginally higher activities for Virkisjökull and Falljökull (Figure 5a; d; g). The mean activities for ^{40}K in Falljökull and Virkisjökull were identical, statistically, whereas the mean of the Skaftafellsjökull samples was only 1.1% lower (Figure 5b; e; h). The mean LOI values for Virkisjökull, Skaftafellsjökull and Falljökull were 0.86 ± 1.12%, 1.51 ± 0.60 % and 0.96 ± 0.20%, respectively (Figure 5c; f; i). These values are relatively low compared to those found elsewhere (Łokas *et al*., 2022; Clason et al., 2023a).

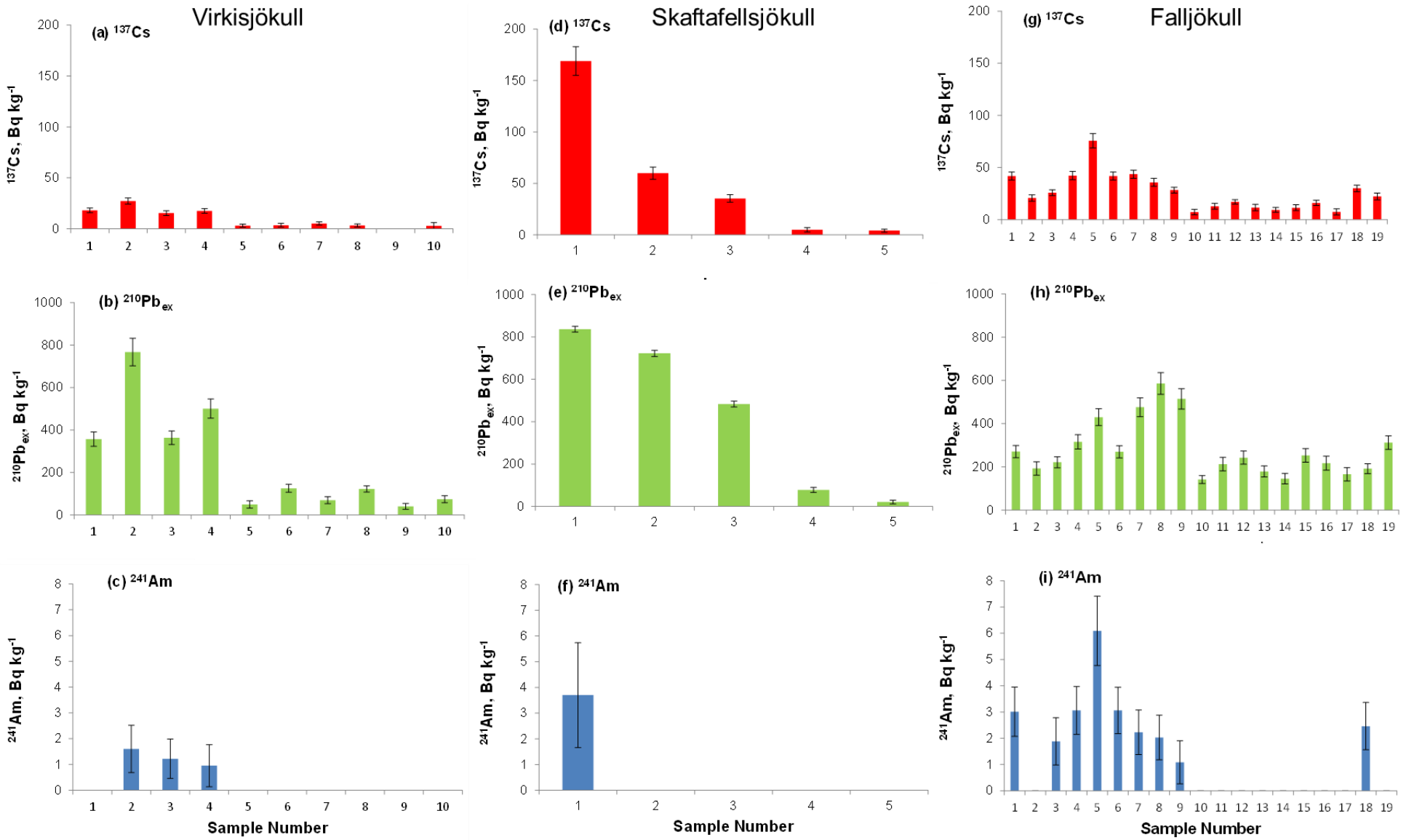

Figure 4. Activity concentrations (Bq kg^{-1}) and counting errors (±2σ) for ^{137}Cs (red), ^{210}Pb$_{ex}$ (green) and ^{241}Am (blue) in cryoconite samples from glaciers in Iceland: a, b, c - Virkisjökull; d, e, f - Skaftafellsjölkull; g, h, I - Falljökull.

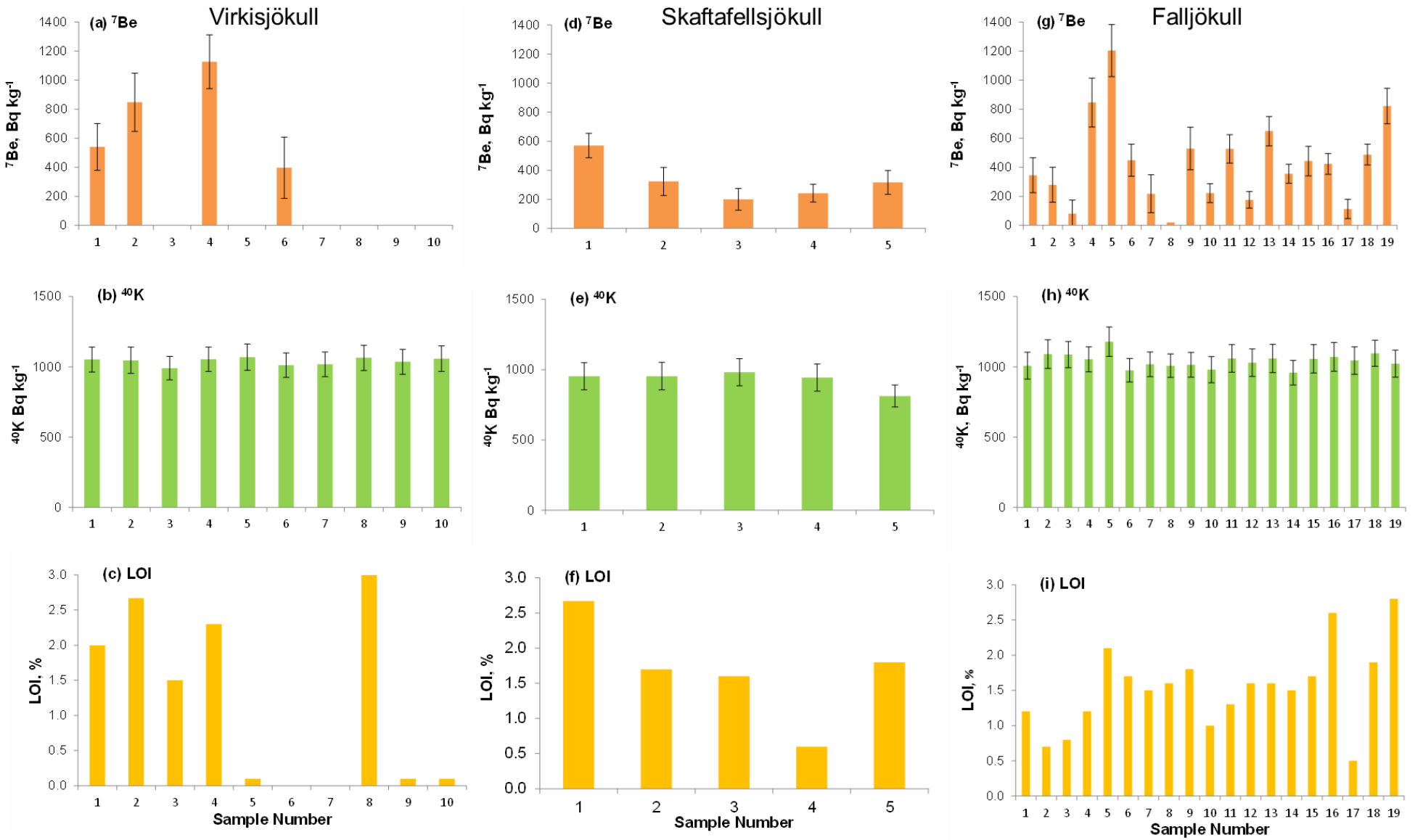

Figure 5. Activity concentrations (Bq kg^{-1}) and counting errors (±2σ) for ^{7}Be (orange), ^{40}K (green) and LOI (yellow) in cryoconite samples from glaciers in Iceland: a, b, c - Virkisjökull; d, e, f - Skaftafellsjölkull; g, h, i - Falljökull.

4.3 Radioactivity in glacier mice

Three samples of glacier mice (moss balls) were retrieved from the surface of Falljökull, an example of which is shown in Figure 6a. The samples were dissected to allow access to their interiors, which were filled with a solid phase similar to the cryoconite particles (Figure 6b). Whole samples of glacier mice, and the internalised lithogenic material, were subjected to gamma counting and XRF spectrometry as described above, and the results are shown in Table 2 and Figure 7. ^{137}Ca and ^{210}Pb$_{ex}$ activities were higher in the whole moss balls than the internalised dusts, and comparable to the activity concentrations reported for cryoconite in the Falljökull-Virkisjökull glacier complex. ^{241}Am was below the minimum detectable activity for all samples. Concentrations of selected elements are also comparable between glacier mice and Icelandic cryoconite (c.f. Fig 3).

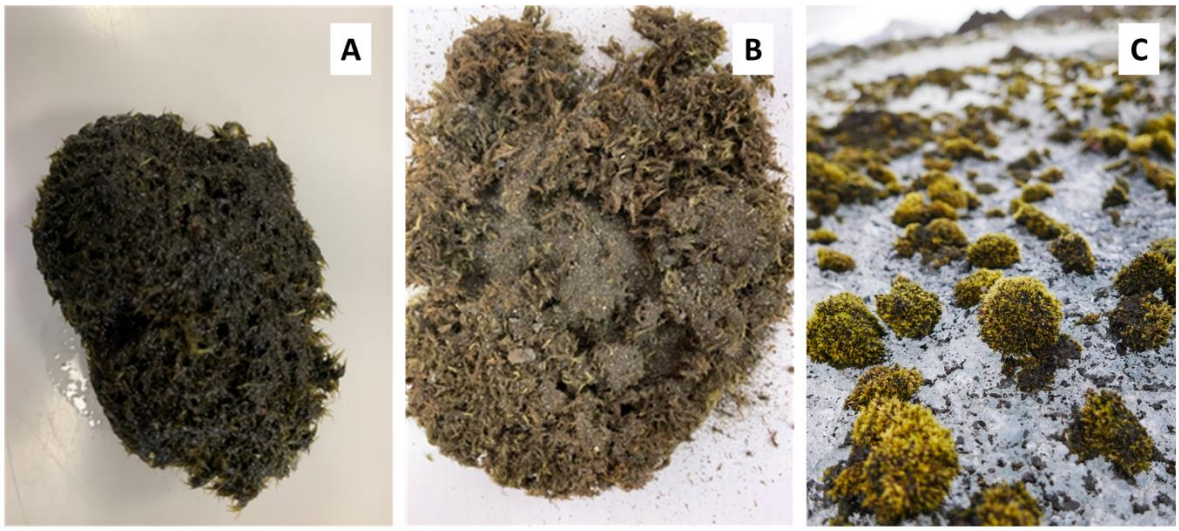

Figure 6. (a) an oblong glacier mouse as collected from Falljökull with any loose surface particulate matter shaken out; (b) a cross section of a glacier mouse illustrating the lithogenic material (moss ball dust) at the centre and the organic exterior (moss); (c) moss balls on the surface of Falljökull.

Table 2. Radionuclide activity concentrations (Bq kg^{-1}) in glacial moss balls and their dusts.

Sample	Shape	Total dry Weight, g	Activity Concentration, Bq kg^{-1}			
			$^{210}Pb_{ex}$	^{137}Cs	^{241}Am	^{40}K
Moss Ball 1	Circular	44	638 ± 30	18.3 ± 0.9	MDA<2.4	681 ± 28
Moss Ball 2	Oblong	70	627 ± 28	17.1 ± 0.8	MDA<1.4	524 ± 21
Moss Ball 3	Oblong	52	672 ± 31	17.8 ± 0.9	MDA<1.9	631 ± 26
Moss Ball Dust 1	---	5.8	186 ± 36	8.9 ± 1.7	MDA<1.7	1067 ± 143
Moss Ball Dust 2	---	4.5	386 ± 5.3	15.1 ± 3.2	MDA<2.7	1486 ± 193
Moss Ball Dust 3	---	6.5	235 ± 4.5	10.6 ± 3.0	MDA<1.50	1035 ± 135

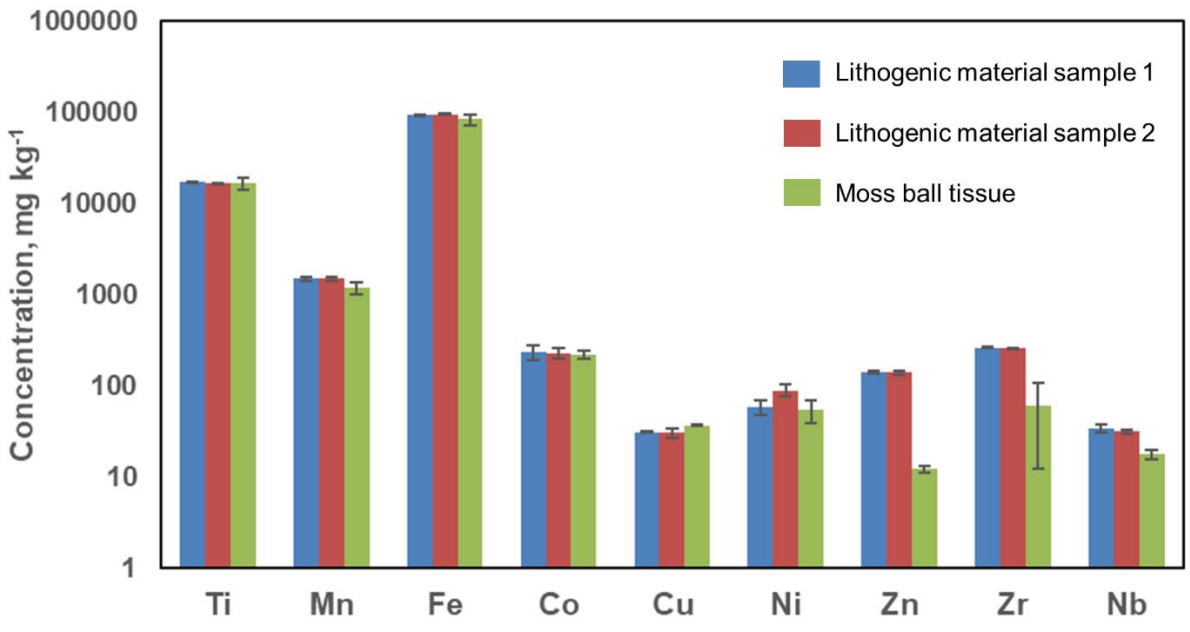

Figure 7. Concentrations of elements (mg kg^{-1}) determined in glacier mice from Falljökull (n=3) arranged in increasing atomic number: lithogenic material sample 1 (blue); lithogenic material sample 2 (red); moss ball tissue (green).

5. Discussion

5.1 Natural and anthropogenic sources of contaminants in Icelandic cryoconite

The mean concentrations of SiO_2, Al_2O_3, CaO and K_2O in cryoconite, and the local control site, were lower than values for the upper continental crust (Rudnick and Goa, 2014). The major geochemical composition of cryoconite may thus be linked to the local geology and other natural local sources (Singh *et al*., 2013). Volcanic ash samples comprised of mainly of basalt or andesite have been shown to contain relatively high SiO_2 contents (Table 3), whereas dust samples from near Skeiðarársandur and Mælifellssandur contain lower SiO_2 values and have other major element contents that cohere more closely with cryoconite samples analysed here. The elements Fe_2O_3, MgO and Na_2O are higher in volcanic ashes and dusts than the cryoconite samples, therefore natural sources may explain the higher concentrations. Minor elements in the cryoconite samples, Co, Ni, Cu, Zn, Zr and Nb, were found in higher concentrations than for both the upper continental crust values and those from the control site. The heavy metals found in the cryoconite samples may derive from industrial sources in Iceland discussed above, or from other, more distal sources by atmospheric transport (He et al., 2023).

Table 3. Ranges of selected elemental compositions of volcanic ash from Grimsvötn and Eyjafjallajökull (Vogel et al., 2017) and dust samples from Skeiðarársandur and Mælifellsandur (Arnalds et al., 2016) compared to the mean cryoconite and control sample compositions from this study.

Element	Volcanic Ash[a], %	Volcanic Dust[b], %	Mean Icelandic Cryoconite[c], %	Controls[d], %
SiO_2	51.7-61.2	45.0-42.5	43.1 ± 1.8	51.3 ± 0.4
Al_2O_3	13.6-14.7	14.5-14.2	14.4 ± 0.53	21.5 ± 0.4
Fe_2O_3	14.8-9.2	16.1-18.9	15.8 ± 1.3	11.6 ± 0.02
MgO	5.83-2.13	6.2-4.9	4.3 ± 0.5	3.8 ± 0.09
CaO	9.54-4.92	12.0-11.6	8.5 ± 0.7	11.2 ± 0.3
Na_2O	3.01-5.26	4.0-4.1	3.6 ± 0.2	4.36 ± 0.2
K_2O	0.15-1.66	---	0.58 ± 0.08	0.084 ± 0.002
Ti	0.016-0.010	2.1-3.4	2.0 ± 0.4	0.12 ± 0.03

[a]Vogel et al., 2017; [b]Arnalds et al., 2016; [c]This study (n=34); [d]This study (n=3).

5.2. Radionuclides in Icelandic cryoconite

Nuclear weapons testing and nuclear accidents, including Chernobyl in 1986 and Fukashima in 2011, have led to contamination of regions across the Northern Hemisphere (Steinhauser *et al*., 2014). For example, ^{137}Cs was introduced into the environment as early as 1945 and beyond during periods of nuclear weapons testing (UNSCEAR, 2000). The anticlockwise circulation of the atmosphere has allowed a global redistribution of 545 Mt of radioactive waste products generated by atmospheric testing and nuclear accidents through aerial transport. The potential for long-term fallout of waste from nuclear tests has been emphasised in research which shows that ^{137}Cs and Pu radionuclides can persist in the stratosphere for 2.5-5 years, and longer than previously estimated (Corcho Alvarado et al., 2014). However, Iceland received comparatively

less fallout from Chernobyl than other regions of the Northern Hemisphere, which is a likely contributing factor to the lower activities of ^{137}Cs found in Icelandic cryoconite compared to other areas such as central Europe and Scandinavia (Clason et al., 2023a). Strong spatial differences in local surface topography, winds, and precipitation also contribute to the considerable variability observed in the ^{137}Cs deposited in soils and vegetation across Iceland by historical nuclear activities (Palsson et al., 2002). Furthermore, Iceland experiences up to 135 dust storms annually, which could potentially dilute atmospheric FRNs before being deposited onto glacier surfaces, contributing to the relatively low activity concentrations in cryoconite in this region. Vatnajökull is subject to frequent and strong katabatic winds and the ensuing dust storms are highly effective at eroding and redistributing particulate matter deposited on glacier surfaces, including dry cryoconite granules (Hannesdóttir et al., 2013).

Owens *et al* (2019) and Łokas *et al* (2022) have highlighted the importance of organic matter in the accumulation of radionuclides due to its chemical complexation properties. Thus, the variability in activity concentrations of radionuclides between cryoconite samples from Iceland and other locations in the Northern Hemisphere may also be explained by the organic matter content of the solids (Clason et al., 2023a). The cryoconite samples analysed here have a relatively low organic content, in the range 0.0-2.9% (c.f. 0.0-38.5% for cryoconite from sites globally; Clason et al., 2023), and a potential explanation for this is the influence of active volcanoes in Iceland contributing to the deposition of volcanic ash onto glacier surfaces (Lutz *et al*., 2015). The prevalence of volcanic ash and dust influences the mineral content of cryoconite while also providing various nutrients (Lutz *et al*., 2015; Meinander *et al.*, 2016) to the potential benefit of the microbial community in cryoconite (Nagatsuka *et al*, 2014). The precise role of the glacier microbiome in accumulating contaminants, and how this will change under future increased meltwater production, however, is yet to be explored in detail.

Icelandic cryoconite samples also contain ^{7}Be, a cosmogenic radionuclide that forms when high energy cosmic radiation interacts with oxygen and nitrogen in the upper atmosphere. The activities of ^{7}Be in the Icelandic samples may be a result of its geographical location in the higher latitudes (Denk *et al.*, 2011), where a hole in the ozone layer over the Arctic could allow the access of fallout of atmospheric ^{7}Be. Since ^{7}Be has a short half-life and the Icelandic samples show relatively high activity concentrations, this indicates that ^{7}Be was deposited with the cryoconite samples soon before sample collection. This is in line with the findings of Baccolo *et al* (2020a) for cryoconite in the European Alps.

5.3. Radionuclide activity in glacier mice

Moss has often been used as a bioindicator of environmental pollution due to its ability to accumulate atmospheric radionuclides (Baccolo *et al*., 2020b). Moss balls or 'glacier mice' are found on Icelandic glaciers and have also been described for a limited number of other glacier sites around the world (Hotaling et al., 2020), with glacier mice on Falljökull previously having been shown to be a habitat for a range of invertebrate fauna (Coulson and Midgley, 2012). Glacier mice have high organic matter content and can accumulate significant amounts of silt by trapping fine dust, and also have a capacity to hold water, supporting a moist organic habitat of importance to a range of invertebrates, including Collembola, Nematoda, and Tardigrada (Coulson and Midgley, 2012). Our analyses of moss balls (n=3) gave a mean ^{137}Cs activity concentration of 17.7 ± 0.9 Bq kg^{-1} (Table 2) which fits into to the range of 7.3-75.7 Bq kg^{-1} for values of ^{137}Cs in the Falljökull cryoconite samples (Fig. 4g). Similarly, $^{210}Pb_{ex}$ in the moss balls had a mean value of 646 ± 23 Bq kg^{-1} compared to a range of 142-586 Bq kg^{-1} for Falljökull cryoconite (Fig. 4h). Consequently, the activity concentrations in whole glacier mice, and in the internalised solid phase (Table 2), are similar to those found in cryoconite on

the same glacier. Kołtonik et al. (2024) collected samples of glaciers mice from Austerdalsbreen in Norway in 2021, reporting a median of 177 Bq kg^{-1} for ^{137}Cs and 974 Bq kg^{-1} for ^{210}Pb for moss balls sampled from the ice surface. While ^{210}Pb concentrations are comparable to those reported here, ^{137}Cs is an order of magnitude higher in the Norwegian samples, and in line with reported regional variability in anthropogenic FRN concentrations in cryoconite (Clason et al., 2023). Further studies of the processes involved in the uptake of radionuclides into glacier mice are required to improve understanding of contaminant acquisition mechanisms.

5.4 Impact on the downstream environment

The impact of heavy metals on organisms, including humans, depends on their mobility in the environment, the chemical species, and the dose taken up (Tchounwou *et al.*, 2012). Some heavy metals are essential micronutrients for humans and animals but can also be harmful in excess quantities (Dietz *et al.*, 1998). Heavy metals of concern in Icelandic cryoconite include Co, Cu, Ni, Zn (Emsley, 2001). Canadian Sediment Quality Guidelines (SQG) for Cu, Zn and Ni are shown in Table 4 (CCME, 1995), and comparison against concentrations found in cryoconite suggests that levels of these potentially toxic elements are not concerning. Nickel is found in cryoconite at levels exceeding the probable effects level (PEL), however, the level of Ni is lower in cryoconite when compared with the Norwegian Moderate values.

410 **Table 4.** Current Sediment Quality Guidelines (SQG) for Cr, Cu, Ni, Pb, and Zn (mg kg^{-1}) and
411 comparison with the maximum levels found in Icelandic cryoconite samples. TEL = minimum adverse
412 effects level; PEL = probable effects level; CB PEC = Consensus Based probable effect concentrations;
413 and Norwegian Moderate.

Guideline Parameters, mg kg^{-1}	**Cr**	**Cu**	**Ni**	**Pb**	**Zn**
TEL	52.3	18.7	48	30.2	124
PEL	160	108	36	112	271
CB PEC	43.4	31.6	22.7	25.8	121
Norwegian Moderate	100	150	130	120	700
Maximum level in cryoconite	143[1]	143[1]	71.5[1]	112[1]	174[2]

414 [1]Sample from Skaftafellsjökull; [2]Sample from Virkisjökull.

415

416 In terms of radioactivity, ^{137}Cs, ^{210}Pb, and ^{241}Am are the radionuclides of most concern when
417 deposited into the environment where the biological damage caused by these radionuclides
418 depends on the radioactive half-life (i.e. persistence and activity in the environment), the
419 chemical form and mobility in the environment, route of exposure, and the type of radiation
420 emitted during decay. Activity concentrations of ^{137}Cs, ^{210}Pb and ^{241}Am in cryoconite samples
421 from Iceland are much lower than the activity concentrations measured on other glaciers in the
422 Northern Hemisphere (Table 5), however they still exhibit higher activity concentrations of
423 radionuclides compared to the off-ice control site. This highlights the ability of cryoconite to
424 effectively accumulate radionuclides even in regions with lower levels of atmospheric
425 contaminant deposition. Davidson et al. (2023) assessed the potential chemical lability of these
426 radionuclides when subject to various chemical environments for cryoconite samples from
427 Isfallsglaciären, Sweden (Clason *et al.*,2021) and Skaftafellsjökull, Iceland. A three-stage
428 sequential chemical extraction comprising fractions of radionuclides that are (1) exchangeable,

(2) reducible, (3) oxidisable and (4) residual was used to assess the radionuclide accessibility. ^{137}Cs was strongly bound to the cryoconite matrix resulting in <20% removal after three extraction steps. In contrast, $^{210}Pb_{ex}$ and ^{241}Am were more chemically mobile and ~90% and 70% respectively were removed by stage 3. These releases indicate the chemical mobility of radionuclides in cryoconite, and hence their potential for human, animal and environmental impact in glacial environments.

Overall, it is unlikely that the activity concentrations, and contaminant concentrations, we present for cryoconite and glacier mice in Iceland will have serious consequences for life in the downstream environment. It has previously been shown in a study of Castle Creek glacier in Canada (Owens *et al*., 2019) that clastic materials in the proglacial zone exposed following recent glacier retreat and sediments from a proglacial stream did not contain high levels of radioactive or stable contaminants in comparison to cryoconite at this site. This is likely a result of the dilution effect from the surrounding environmental media (Hasholt et al., 2000). However, the accumulation of contaminated sediments, including remobilised cryoconite, in proglacial lakes, could pose a risk to aquatic ecosystems in some glacial systems, particularly where contaminant levels in supraglacial materials are orders of magnitude higher than those found in Icelandic samples (Owens *et al.*, 2019; Clason et al., 2021).

Table 5. Comparison of activity concentrations ranges (Bq kg^{-1}) for ^{137}Cs, ^{210}Pb and ^{241}Am for selected glaciers in the northern and southern hemispheres.

Glacier and location	Range or mean activity concentrations (Bq kg^{-1})			Reference
	^{137}Cs	^{210}Pb	^{241}Am	
Falljökull, Virkisjökull, Skaftafellsjökull, Iceland	1.8 to 169	29.5 to 850	0.6 to 6.1	This study
Adishi Glacier, Georgia	580 to 4,940	1,400 to 12,000	8.1 to 68.3	Łokas *et al.* (2018)
Morteratsch Glacier, Switzerland	325 to 13558	1581 to 4143	2.9 to 120	Baccolo *et al.* (2020a)
Ecology Glacier, Antarctica	185 to 350	1400 to 2600	1.69 to 5.02	Buda *et al.* (2020)
Isfallsglaciären, Sweden	1010 to 4530	5760 to 14700	6.1 to 74	Clason *et al.* (2021)
Orwell Glacier, Antarctica	0.48 to 54.4	5.5 to 249.1	MDA to 1.61	Owens *et al.* (2023)

5. Conclusions

This investigation has identified, for the first time, the spatial variability of radioactive and trace elements in cryoconite samples and glacier mice from Icelandic glaciers. Local geology and natural particulate sources such as volcanic ash and dust likely contribute to the main geochemical composition of the Icelandic cryoconite. The radionuclides and many of the trace elements were higher in cryoconite than the proglacial sediments from the control site. The activity concentrations are, however, relatively low compared to published values from other glaciers in the northern hemisphere, and these already low values would likely be further diluted during transport within the downstream proglacial system. We also reported, for the first time, the trace element and radioactive content of Icelandic glacier mice, which are a prominent feature of the Falljökull supraglacial ecosystem. The capacity of glacier mice to acquire elevated radionuclide activities, in line with those found in cryoconite, suggests they require further investigation to understand their role in sequestering both radioactive and stable elements. While the efficient accumulation of radionuclides in both cryoconite and glacier mice

is elevated over background values, it is still below levels of contamination that would be likely to pose a risk to the downstream environment in this region. Nevertheless, as a ubiquitous component of the supraglacial ecosystem, cryoconite offers an opportunity to monitor environmental radioactivity in areas where flora typically used for this purpose are rare or absent.

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Author contributions

CCC devised the study, CCC and RF conducted field sampling, ES, GM, CCC and AT conducted sample analysis, and ES, GM and CCC conducted interpretation of the data. All authors contributed to preparation of the manuscript, led by ES.

Data availability

Data presented in this manuscript are available within the manuscript and its appendices.

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