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The surface of small glaciers as radioactive hotspots: Concentration of radioisotopes during predicted intensive melting in the Alps

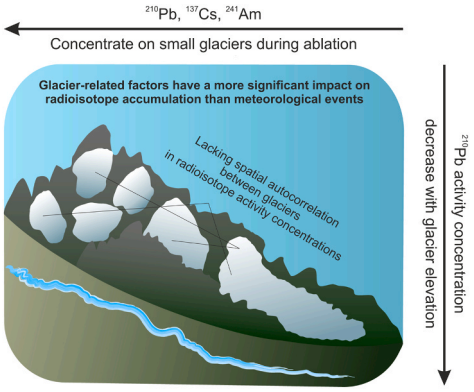
Jakub Buda [a,*], Sylwia Błażej [b], Roberto Ambrosini [c], Riccardo Scotti [d], Francesca Pittino [e], Dariusz Sala [f], Krzysztof Zawierucha [a], Edyta Łokas [f]

[a] Department of Animal Taxonomy and Ecology, Adam Mickiewicz University in Poznań, Poland
[b] Department of Nuclear Physical Chemistry, The Henryk Niewodniczański Institute of Nuclear Physics, Polish Academy of Sciences, Poland
[c] Department of Environmental Science and Policy, University of Milan, Italy
[d] Servizio Glaciologico Lombardo - Glaciological Service of Lombardy, Italy
[e] Department of Earth and Environmental Sciences (DISAT), Università degli Studi di Milano-Bicocca, Italy
[f] Department of Mass Spectrometry, The Henryk Niewodniczański Institute of Nuclear Physics, Polish Academy of Sciences, Poland

HIGHLIGHTS

- ^{210}Pb, ^{137}Cs and ^{241}Am concentrate on small glaciers during the melting of ice.
- Small glaciers that are predicted to melt are a threat to glacier-adjacent biota.
- Accumulation of radioisotopes on glaciers is not spatially related.
- ^{210}Pb accumulates stronger in lower-elevated glaciers.

GRAPHICAL ABSTRACT



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ABSTRACT

Glaciers are considered secondary sources of pollutants, including radioisotopes such as Cesium or Plutonium, with heightened concentrations compared to other ecosystems. The predicted melting of glaciers poses a substantial risk of releasing stored radioisotopes, yet understanding the glacier-specific factors influencing their concentration remains limited. This study investigates the relationship between glacier altitude, surface area, organic matter content in dark supraglacial sediment (cryoconite), precipitation, and activity concentrations of natural (^{210}Pb) and anthropogenic radionuclides (^{137}Cs and ^{241}Am) across 19 Alpine glaciers. Results indicate that radioisotope concentrations depend on organic matter content in the cryoconite, highlighting the role of biotic-abiotic interactions in pollutant accumulation on glaciers. Moreover, ^{210}Pb activity concentration decreases with glacier altitude, likely due to atmospheric variations in ^{222}Rn. Water precipitation events, such as

* Correspondence to: Department of Animal Taxonomy and Ecology, Adam Mickiewicz University in Poznan, Uniwersytetu Poznańskiego 6, 61-614 Poznań, Poland.
E-mail address: jakbud1@amu.edu.pl (J. Buda).

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during peaks in ^{137}Cs deposition and after the Chernobyl Nuclear Power Plant disaster, do not impact current activity concentrations. Importantly, radioisotope concentrations in cryoconite are higher on smaller glaciers. This directly supports the hypothesis that the cryoconite retains a significant share of radioisotopes stored in the ice during intensive melting. Since many small glaciers in the Alps are predicted to disappear within the next 50 years, we anticipate release of radioisotopes to mountain ecosystems might be higher than previously forecasted.

1. Introduction

The record of intensive anthropic pressure, such as the legacy of industry and military activities, is present in diverse habitats across Europe [1-3]. Among the high-risk pollutants, anthropogenic radioisotopes persist in different habitats at relatively high activity concentrations, even if the amount released has dropped significantly since the Limited Test Ban Treaty in 1963 [4,5,1,6-8]. The main sources of anthropogenic radioisotopes in Europe are local and global nuclear weapon tests (known as global fallout) and, to a large extent, the Chernobyl Nuclear Power Plant (NPP) accident [1,9]. Some radionuclides are harmful when ingested or inhaled in high amounts but pose low threats in terms of long-term effects on the environment overall. Two of them, ^{131}I and ^{89}Sr, are especially important for human health, but because of their very short half-life ($t_{½}$ = 8.04 and 50.52 days, respectively), the removal rate from the environment due to natural decay is fast. Therefore, the long-term effects are limited [10]. On the other hand, some radioisotopes persist in the environment and accumulate in higher trophic organisms due to their longer half-life and chemical properties [11-13]. Therefore, the most important in terms of biological threats are radioisotopes released in high amounts but also with relatively long half-lives such as ^{90}Sr ($t_{½}$ = 28.78 yrs.), ^{137}Cs ($t_{½}$ = 30.17 yrs.) or ^{239}Pu ($t_{½}$ = 24 110 yrs.) along with other Plutonium-related isotopes, such as ^{238,240,241}Pu (^{238}Pu: $t_{½}$ = 87.7 yrs., ^{240}Pu: $t_{½}$ = 6561 yrs., ^{241}Pu: $t_{½}$ = 14.3 yrs.) or ^{241}Am ($t_{½}$ = 432 yrs.). Cesium-137 was released by NPP accidents and global military actions, with a peak in deposition in 1963 [14]. Since ^{137}Cs reach high activity concentrations, its chemical properties are similar to those of potassium, and its decay produces beta particles, there is a high concern about its presence in the environment [15,16]. Its primary sources have significantly dropped after the Limited Test Ban Treaty, and its activity concentrations are progressively decreasing, with some peaks in activity due to NPP accidents [5]. However, other longer-lived anthropogenic radioisotopes, such as ^{241}Am or $^{239–240}$Pu, persist in amounts similar to those observed when released [17]. Their radioactivity's effects on ecosystems are still observed but only well investigated in areas close to NPP accidents, where elevated ionising radiation exerted broad effects on wildlife. For instance, close to the Fukushima Daichi NPP accident, increased aberrant cell frequencies were found in pine needles exposed to higher IR doses [18] and decreases in bird abundances were observed due to chronic exposure [19]. Furthermore, scots pines *Pinus sylvestris* exposed to elevated levels of ionising radiation in the vicinity of the Chernobyl NPP accident exhibit increased DNA damage [20]. Additionally, bank voles *Clethrionomys glareolus* have shown a response to this radiation by reducing overall gut health [21] along with genomic scale alterations [22].

Due to their position and relatively intense precipitation, the Alps have received high amounts of anthropogenic radionuclides from global fallout and the Chernobyl NPP accident [23,24]. Relatively high activity concentrations in the Alps were found on the surface of glaciers [25-27]. The dark sediment called cryoconite, which covers glaciers worldwide is a biogeochemical hotspot that adsorbs contaminants delivered with dry and wet deposition from the atmosphere [25,28,29,7,30]. Cryoconite is composed of minerals, microbial communities, and a high share of dead organic matter that originates on the glacier (autochthonous organic matter) or elsewhere (allochthonous organic matter) [31-34]. Due to its dark colour, the sediment absorbs solar radiation and melts into the ice, creating water-filled depressions called cryoconite holes [35,36,33]. This habitat is dominated by microorganisms such as bacteria, algae, fungi and other eukaryotic groups, including heterotrophic protozoa or microinvertebrates; no vascular plants or vertebrates live and reproduce there [37,38,32,39]. Organisms producing extracellular polymeric substances like Cyanobacteria, which are highly abundant in cryoconite, seem to play an important role in binding the mentioned cryoconite compounds, but potentially also the contaminants into spherical granules [33,34,40]. These bioaggregates were found to be important in the analysis of sources of radioactivity globally, even in remote areas, due to the capability of cryoconite to accumulate radionuclides which also provides a new substratum for environmental monitoring [26,37,29,41,42,27,43].

On Alpine glaciers, the main sources of anthropogenic radionuclides were global fallout (atmospheric nuclear blasts) with a peak in the '60 s and the Chernobyl NPP accident in 1986 [4,27,43]. The concentrations of anthropogenic isotopes estimated for 1986 reached 223 kBq kg^{-1} of ^{137}Cs and 129.1 kBq kg^{-1} of ^{134}Cs [43]. On the other hand, the activity concentration of the most abundant natural isotope found in cryoconite – ^{210}Pb – was equal to 57 kBq kg^{-1} when estimated for 1986 [43]. The overall comparison of mean activity concentrations between cryoconite and other habitats in the Alps has shown that both natural and anthropogenic radioisotopes are higher in the former by one to two orders of magnitude than in surrounding ecosystems [4]. These works are important for understanding pollution levels and radionuclide sources in the Alps but were limited to a few glaciers. The lack of large-scale data lowers our ability to infer spatial variation and factors that affected the accumulation in the Alps. Indeed, the high variation in activity concentrations observed between the investigated glaciers raises a question of whether the accumulation is mainly driven by spatial variation (e.g. distance from the source or variation in precipitation) or glacier-related features. Recognition of these processes might be important in estimating the effects of ongoing intense glacier melting for the release of radioactive contaminants accumulated over time, some of which, like ^{239}Pu or ^{241}Am, persist in the environment over an extended time.

In this research, we examine the accumulation and concentration of radioisotopes on glaciers at a regional scale. Our analysis is based on cryoconite sediment samples collected from 19 glaciers in the Alps over two years. We specifically focused on two anthropogenic radioisotopes (^{137}Cs and ^{241}Am) and one natural radioisotope (^{210}Pb). These radioisotopes are the most radioactive fallout isotopes on glaciers, widely detectable by gamma spectrometry, and known to concentrate well in cryoconite compared to other habitats [44,29,41]. Therefore, they are the most important isotopes for analysing the threats related to the biota under intensive melting and the potential release of stored radioisotopes. We measured radioisotopes' spatial variation in activity concentrations and examined their relationship with glacier features such as average altitude, surface area, and organic matter content in the cryoconite. Moreover, given that the concentration of ^{137}Cs and ^{241}Am from the global atmospheric fallout could be influenced by precipitation, we also analysed their activity concentrations in relation to past precipitation on the glaciers. This approach, combined with spatial autocorrelation analysis, allows us to determine whether the observed high radioactivity on some glaciers is due to past precipitations or related to glacier features that promote the concentration of pollutants on their surfaces. This comprehensive study focused on a single geographical area (the Alps). It thus controlled for unknown parameters such as large variations in ^{222}Rn sources (an indirect parental isotope of ^{210}Pb), large-scale air masses flow, and the composition of minerals and

organisms, thereby enhancing the power of our inferences. This could be beneficial for future predictive analyses of ongoing global changes and the potential release of secondary pollutant sources due to glacier melting.

2. Methods

2.1. Study area and sampling

In this study, our focus was on the most glaciated mountain groups in the Alps. We collected a total of 140 samples from 19 different glaciers during the 2020 and 2021 campaigns (Fig. 1). The altitude of the glacier sampling zones (ablation areas) ranged from 2015 to 3004 m a.s.l with a mean elevation of 2569 m a.s.l. A list of the investigated glaciers and their features is provided in Supplementary Table 1. We collected five to ten cryoconite samples from distinct cryoconite holes using a sterile clean pipette or a stainless-steel spoon from each glacier. We ensured that the cryoconite holes sampled were not interconnected. The collected samples were frozen at − 20 °C or preserved with ethyl alcohol. Upon returning to the laboratory, the samples were homogenised and dried in preparation for radiometric analysis. The average dry mass of the cryoconite samples used in the analysis varied from 0.462 g to 2.134 g, with a mean value of 0.948 g.

2.2. Radiometric analysis

Using gamma spectrometry, we estimated the activity concentrations of three fallout radioisotopes, namely ^{137}Cs, ^{210}Pb and ^{241}Am. These radioisotopes are known to be effectively concentrated in cryoconite compared to lithogenic radioisotopes such as ^{40}K, ^{238}U, ^{232}Th, ^{234}Th or ^{212}Bi [4]. These radionuclides are related to their contents in the source minerals [45] and are not influenced by the action of cryoconite. Lead-210 is also a lithogenic radioisotope as a product of the ^{238}U series. However, its presence in the environment might be from two different distribution pathways. The first one is the weathering processes of minerals containing ^{210}Pb (supported ^{210}Pb); this fraction is relatively low mobile and shows equilibrium with ^{238}U concentration in local minerals. The second fraction has atmospheric distribution, as the indirect parental isotope of ^{210}Pb, which is ^{222}Rn, a noble gas with a short half-life reaching the atmosphere from uranium-rich ores; this fraction is known as unsupported ^{210}Pb and can be transported in the atmosphere and then effectively captured. The vast majority of ^{210}Pb in cryoconite is concentrated from atmospheric deposition as unsupp. ^{210}Pb [4,46], therefore, in our analysis, we relayed on total ^{210}Pb activity

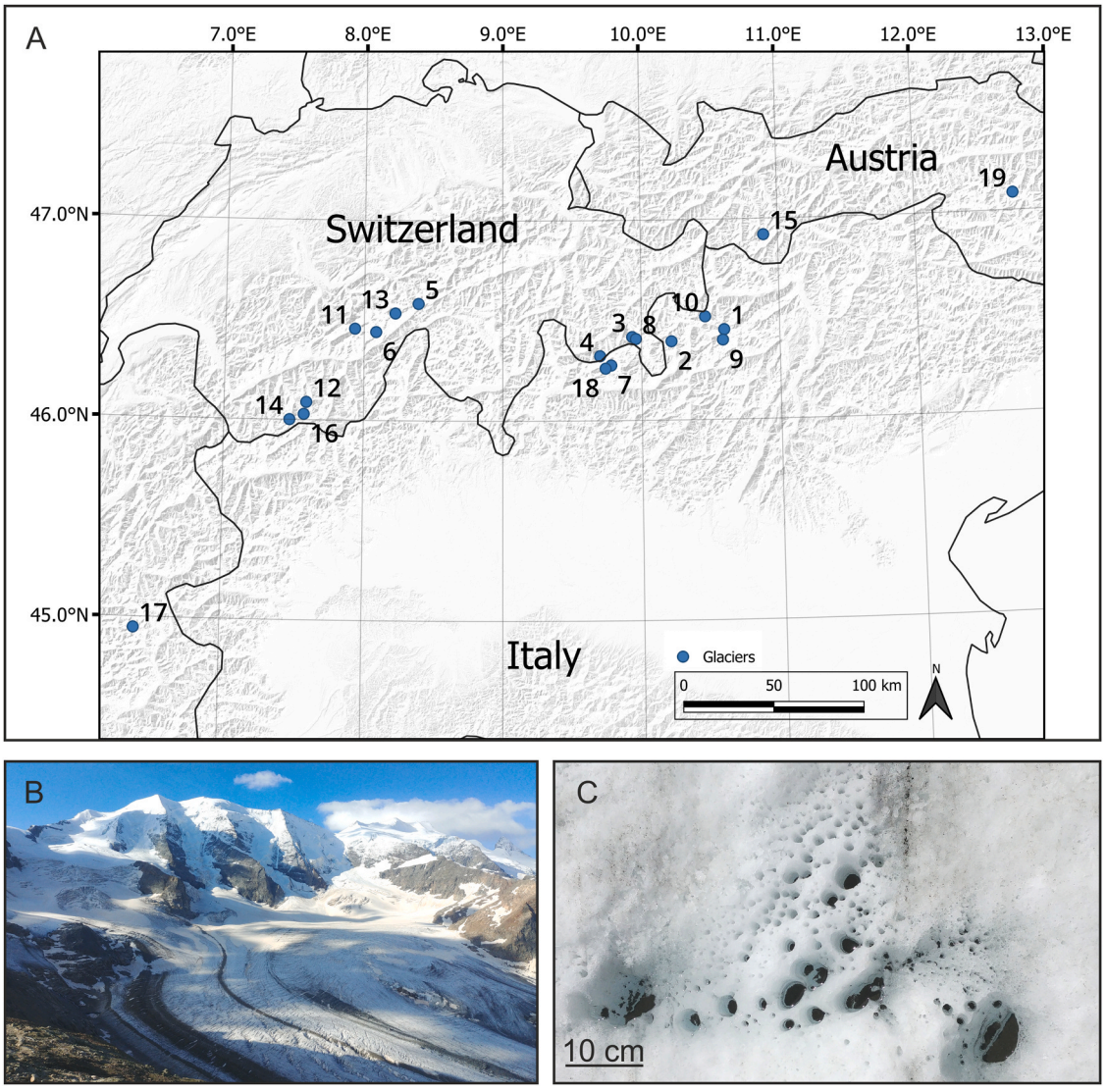

Fig. 1. Map of the Alps with sampled glaciers (blue points). Pers Glacier (B), cryoconite holes (C) on de Moiry Glacier surface. The ID corresponds to the numeration in Supplementary Table 1 and 2.

concentrations assuming that they represented mostly unsupp. ^{210}Pb. An example spectrum is presented in Supplementary Figure 1.

The samples were analysed with a low-background, digital gamma-ray spectrometer equipped with a Broad Energy Germanium (BEGe) detector BE5030 with a relative efficiency of about 48 % and a multi-layer passive shield surrounded by an active shield's detector [47]. For ^{137}Cs, activity was determined by measuring the 661.6 keV emission peak of ^{137m}Cs, for ^{210}Pb, it was the 46.5 keV peak, while for ^{241}Am, the 59.5 keV peak was used as an analytical signal. Efficiency calibration (including self-absorption correction) for used measurement geometry was determined using LabSOCS calibration software (Mirion Technologies). The spectra were collected for approximately 24 h. The activity concentrations were then calculated on the day of sampling. Data quality was evaluated by measuring of IAEA Reference Materials (IAEA 447). The results (Supplementary Table 3) agreed well with the recommended values.

2.3. Predictors

We quantified organic matter as the percentage weight loss after combustion at 550 °C for 6 h, following homogenisation and drying at 100 °C for 24 h [48]. The mass of the analysed samples was the same as that used for gamma spectroscopy, as the samples were combusted post-radiometric analysis.

We estimated the precipitation related to the peak of global fallout as the total precipitation in the glacier region from January 1962 to December 1964. Additionally, we estimated the precipitation that could influence the deposition of ^{137}Cs following the Chernobyl NPP accident as the total precipitation in May 1986. The data was gathered from WorldClim v2.1 [49,50] and proceeded in qGIS software [51]. We sampled the total precipitation for each month based on raster data from WorldClim, with sampled values for the coordinates of the sampling area. The surface area of glaciers, their average and maximum altitude was estimated for 2015/16, with one extent due to lack of data for 2015/16 (Pers, 2022), based on the Global Land Ice Measurements from Space (GLIMS) database [52]. Detailed parameters of the glaciers and the results of the activity concentrations are presented in Supplementary Tables 1 and 2.

2.4. Statistical analysis

Radiometric data were averaged for each glacier, as our primary goal was to analyse factors that affect activity concentrations of radionuclides on an Alpine scale. First, we tested whether activity concentrations of radioisotopes are spatially autocorrelated between glaciers, which could be a proxy for weather-dependent deposition. This test was based on the k nearest neighbours estimate (k = 3, equal weights for each k) and the orthodromic distance between points, which are coordinates of sampling areas (one for each glacier). This spatial analysis was performed using the *sp* [53] and *spdep* [54] packages.

To determine which factors affect the activity concentrations of ^{210}Pb and ^{137}Cs, we used linear models with a Gaussian error distribution and log-scaled response variables. Based on previous studies, we assumed that organic matter content (OM) is the most important predictor for ^{210}Pb and ^{137}Cs activity concentrations. Therefore, we built a set of models with OM as a core predictor and additional variables specific to each isotope. In the case of ^{210}Pb models, we built models that contained OM as the only predictor, OM with an additive effect of a) the surface area of the glacier, b) the average altitude of the glacier, and c) both the surface area and the average altitude of the glacier. For ^{137}Cs, we built models that contained OM as the only predictor, OM with an additive effect of a) the surface area of the glacier, b) the sum of precipitation during the peak of global fallout (GF), c) the sum of precipitation a month after the Chernobyl NPP accident, d) all the above predictors. The above models were compared separately for each radionuclide using an F test based on a reduction of the Residual Sum of Squares (RSS). The best model was chosen based on RSS reduction and presence-absence of influential observations. A comparison of models is presented in Supplementary Table 4, while only the results of the best models are presented in the results, with the test for predictor significance based on the F-test using Type-II Anova. In the case of ^{241}Am activity concentrations, on 8 out of 19 glaciers, we could not reach the minimum detectable concentration (MDC). Therefore, we used Censored Regression to analyse these data [55]. This model contained the OM and the sum of precipitation during the peak of global fallout (GF) as predictors.

In the above models, the surface area of the glaciers and precipitation data were log-scaled to reduce the effect of influential observations; the Aletsch Glacier is extreme in size (~83.4 km^2) compared to the second largest glacier, Pasterze (16.2 km^2), while the precipitation on the Pasterze Glacier was four-fold higher than those on the second in order (see Supplementary Table 1).

Models were implemented in the R software v4.3.2 [56] using the *lm* function and the *censReg* [55] package. Linear models were checked for violation of assumptions based on diagnostic plots, and outliers' presence tested based on Cook's distances using *performance* package [57]. A project with raw data and R code is presented in the GitHub repository under the link: https://github.com/jakbud1/Small-glaciers-as-radioisotopes-hotspots-in-the-Alps.git.

3. Results and discussion

3.1. Radioisotopes pollution levels

The activity concentrations of ^{210}Pb, ^{137}Cs, and ^{241}Am in cryoconite were within the range of those previously reported on glaciers in the Alps [26,43], with an overall mean of 4 655 Bq kg^{-1}, 1 732 Bq kg^{-1} and 15 Bq kg^{-1}, respectively (Fig. 2). However, these activity concentrations are higher than those on glaciers in South America or Antarctica, but within the range of those found in the Northern Hemisphere [29]. The lowest average activity concentration of ^{210}Pb was found on the Lang Glacier (661 Bq kg^{-1}), while for ^{137}Cs, it was on the Oberaar Glacier (36 Bq kg^{-1}). The activity concentration of ^{241}Am was not detectable in any sample for 8 of the 19 glaciers, with a minimum average of 11 Bq kg^{-1} for glaciers that were above detection limits (which varied between 2.2 to 26 Bq kg^{-1}). In contrast, the highest activity concentration of ^{210}Pb was found on the Ebenferner (10 968 Bq kg^{-1}), ^{137}Cs on the Ventina Glacier (5 785 Bq kg^{-1}), and ^{241}Am on the Dosdé Glacier (40 Bq kg^{-1}). Average data are presented in Fig. 2A and Supplementary Table 3. The high variation in activity concentrations between glaciers, even those closely located, without a clear spatial pattern, suggests that glacier-related processes are involved in accumulating natural and anthropogenic radioactivity on glaciers. While the correlations between the activity concentrations of each pair of radionuclides are positive (Fig. 2B), there is still a high proportion of unexplained variation, indicating that different mechanisms are involved in their accumulation on glaciers, possibly related to both sources and glacier features.

3.2. Effects of glacier-related features on activity concentration of radionuclides

Contrary to our expectation, neither the sum of precipitation during the GF peak nor the month following the Chernobyl NPP accident explained the current activity concentrations of ^{137}Cs (the best model based on the reduction of RSS did not contain these predictors) and ^{241}Am ($F_{1,14} = 1.495$, $p = 0.242$; Table 1). The total precipitation varied, from 1840 mm to 5063.3 mm for the 3-years GF peak, and from 68.9 mm to 154 mm one month after Chernobyl NPP accident. This contradicts our hypothesis of an increase in the activity concentrations of ^{137}Cs with the sum of precipitation during the GF peak and in May 1986. The activity concentrations are calculated per 2020/21. However, this should have only implied lower values that vary linearly with

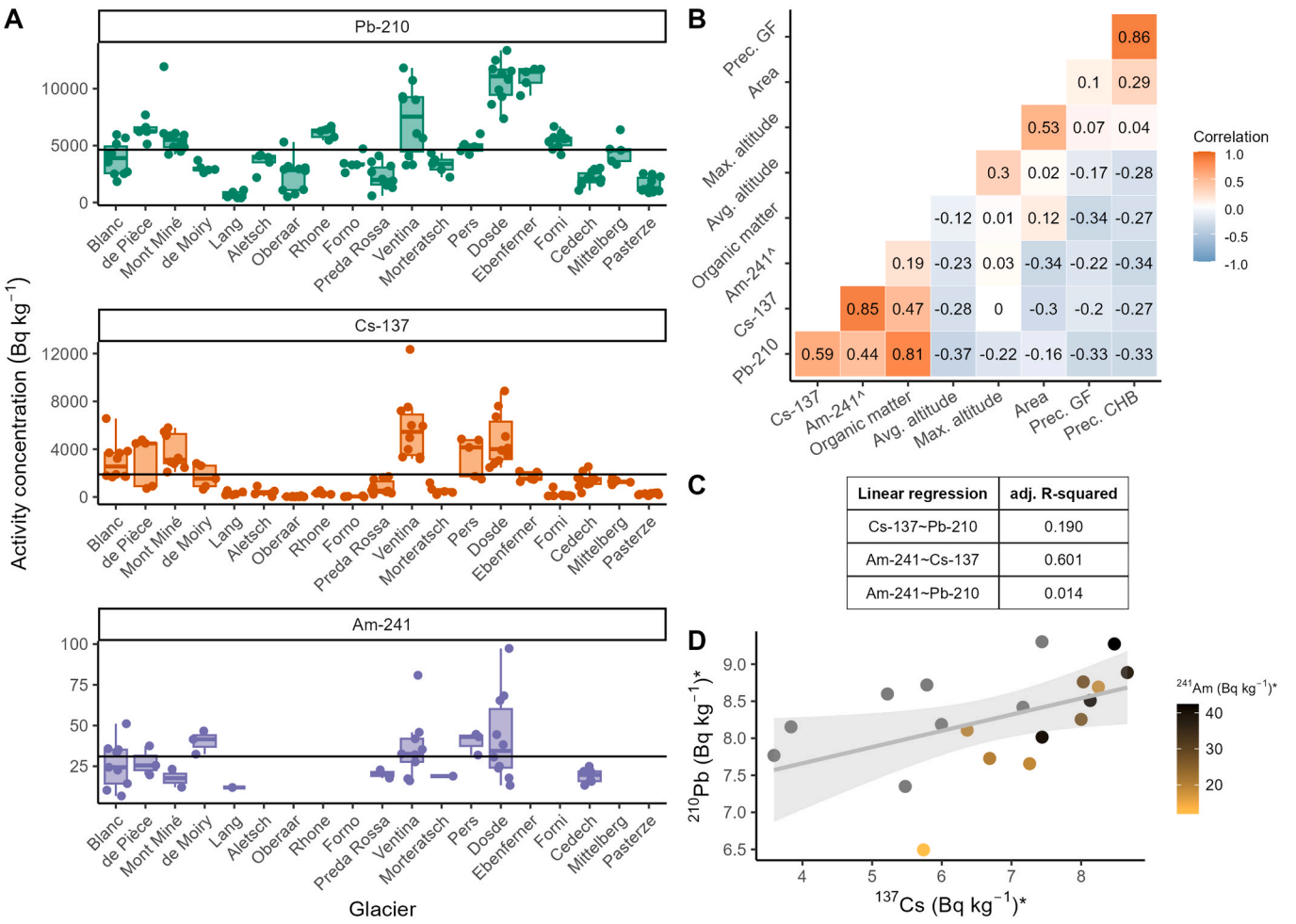

Fig. 2. Inter- and intra-glacier variation of ^{210}Pb, ^{137}Cs, and ^{241}Am activity concentrations across the Alpine glaciers ordered from West to East, solid lines on plots represent overall means (A). Correlation of variables included in the analysis, ˆValues of ^{241}Am that were below MDC assigned as ½ MDC (B). Adjusted R^2 of linear relations between each pair of radioisotopes (C). Linear relation of ^{210}Pb on ^{137}Cs with ^{241}Am activity concentrations represented by colour as the third dimension, grey dots are observations below MDC; *log-scaled vectors (D). Correlations between precipitations after GF and Chernobyl NPP accidents are meaningful only for ^{137}Cs and ^{241}Am activity concentrations.

Table 1
Results of linear models (censored regression for ^{241}Am). *log-scaled vectors. P-values under 0.05 are bolded. For the censored regression, the "log(sigma)" refers to the proportion of unexplained variation by fixed effects, in this model 8 out of 19 observations were left-censored. Comparison of models are presented in the Supplementary Table 4. Model summary for ^{210}Pb: adj. $R^2 = 0.83$, $F_{3,15} = 29.82$, $P < 0.001$; ^{137}Cs: adj. $R^2 = 0.32$, $F_{2,16} = 5.28$, $P = 0.017$.

Response	Parameter	Estimate	SE	t-value	p
^{210}Pb*	Intercept	10.727	1.2965	8.274	**< 0.001**
	Organic matter content	0.144	0.0172	8.364	**< 0.001**
	Average altitude	-0.001	0.0004	-2.708	**0.016**
	Surface area*	-0.125	0.0548	-2.283	**0.038**
^{137}Cs*	Intercept	5.974	0.761	7.856	**< 0.001**
	Organic matter content	0.189	0.075	2.530	**0.026**
	Surface area*	-0.522	0.237	-2.209	**0.042**
^{241}Am*	Intercept	29.505	21.4497	1.376	0.169
	Organic matter content	-0.076	0.1398	-0.540	0.589
	Surface area*	-1.101	0.4823	-2.282	**0.022**
	Precipitation GF*	-3.328	2.7216	-1.223	0.221
	log (sigma)	*0.720*	*0.2384*	*3.019*	***0.003***

precipitations. Indeed, a significant amount of ^{137}Cs released during the Chernobyl NPP accident was subject to wet deposition, strongly correlated with precipitation [58]. Although precipitation was found to be the strongest predictor of ^{137}Cs deposition from global fallout [59], the lack of precipitation effects is evident from our study by the absence of spatial autocorrelation (Table 2, Fig. 3). In a scenario of precipitation effect, we should observe similarities in activity concentrations of isotopes between spatially close glaciers. Similarly, based on Pearson's correlation, Clason et al., [29] also did not find a relation between the activity concentration of ^{137}Cs and the distance from Chernobyl in the Northern Hemisphere. This suggests that in the case of ^{137}Cs and ^{241}Am, factors related to glacier features are more important for predicting current activity concentrations than the sources, distance and potentially received amounts of radionuclides in past. Therefore, the observed activity concentrations probably reflect the ability of specific glaciers to concentrate fallout radioisotopes more efficiently than others.

Table 2
Results of spatial autocorrelation analysis based on Moran's I test.

Isotope	Moran's I statistic	Z	P
^{210}Pb	0.062	0.432	0.433
^{137}Cs	0.098	0.462	0.315
^{241}Am	0.147	0.512	0.184

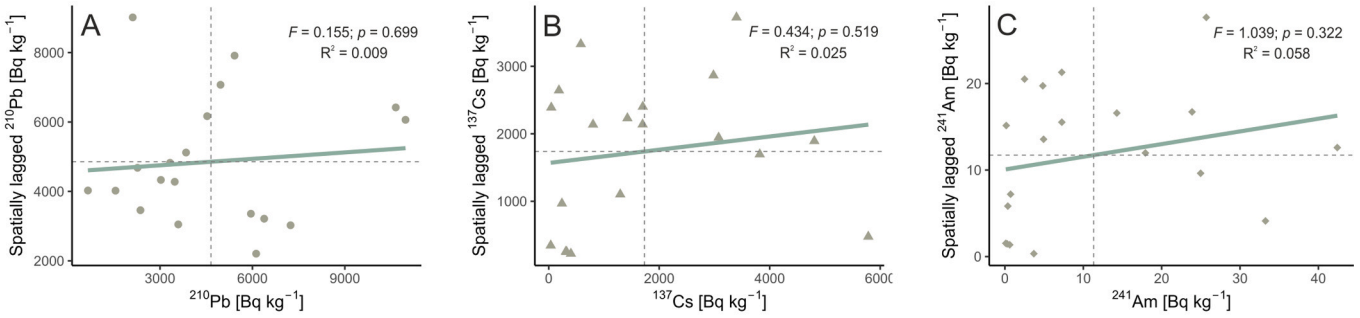

Fig. 3. Lack of spatial autocorrelation of ^{210}Pb (A), ^{137}Cs (B) and ^{241}Am (C) activity concentrations. Dots represent observations for glaciers, a solid-coloured line is a regression line while dashed horizontal and vertical lines are means of observations (x-axis) and spatially lagged observations (y-axis).

We observed a negative relation between the activity concentration of ^{210}Pb in cryoconite and the average altitude of a glacier ($F_{1,14}$ = 1.495, p = 0.039; Table 1, Fig. 4). The natural isotope ^{210}Pb on glaciers precipitates primarily as a product of ^{222}Rn decay, with short-lived nuclides between them in a decay chain. The unsupported fraction of ^{210}Pb is a majority in alpine cryoconite, consisting of 99.1 % of total ^{210}Pb for Forni Glacier and 97.2 % on Morteratsch Glacier [4]. As ^{222}Rn is a noble gas with a short half-life ($t_{½}$ = 3.6 days) originating from uranium-reach ores underground, its concentrations in the air decrease with the altitude [60,61], thus potentially explaining our observed negative relation of ^{210}Pb activity concentration with altitude. ^{210}Pb, an indirect daughter isotope (with other short-lived isotopes in between), also follows this linear pattern with altitude, as found in our case for cryoconite [62]. Results obtained in this study are not consistent with the reports of Clason et al., [29], who found a positive correlation between altitude and ^{210}Pb activity concentrations in the Northern Hemisphere, while no other predictors were taken into account in that analysis (based on Pearson's correlation only). These studies differ also by the scale. Data from Clason et al., [29] was collected globally, covering two groups of glaciers: the low-elevation glaciers in polar regions and high-elevation mountain glaciers. The low-elevated polar glaciers are located close to the seas, while the mountain glaciers are mainly on the continents. ^{222}Rn varies not only altitudinaly but also between lands and oceans; the estimated contribution of the oceans in global ^{222}Rn release was lower than 5 %, being significantly higher above grounds than the oceans [60,63]. Therefore, the results of Clason et al., [29] show variation between altitudes, which could also be interpreted as the variation between two types of glacier localities. Indeed, the authors observed an effect of the distance from the ocean on ^{210}Pb activity concentrations. However, this variable was not included in their models, while, according to the results of a PCA reported in that study, distance from the ocean was also positively correlated with mean elevation. It is therefore possible that the observed effects were the results of collinearity. In this study, we controlled for this variation by analysing glaciers in one region, and we assessed a lack of spatial autocorrelation in ^{137}Cs, ^{241}Am and ^{210}Pb activities concentrations (Table 2; Fig. 3). This suggests a lack of local effects related to ^{222}Rn sources on ^{210}Pb activity concentrations. It is worth mentioning that even if ^{222}Rn shows a general trend of decrease with altitude on regional scales [61], the activity concentrations of ^{210}Pb do not always follow this trend [64,65]. This might be due to the influence of local wind circulation, foehn wind, random wind gusts and the concentration of aerosols on which ^{210}Pb quickly adsorbs [66]. Moreover, it was shown that the ground-level of ^{210}Pb activity concentration should show some correlation with rain and snow data for the same region [67]. However, glaciers located close to one another and subject to similar precipitations had no similar ^{210}Pb activity concentrations (Table 2; Fig. 3). It cannot be excluded that the observed increase of ^{210}Pb activity concentrations on high elevation glaciers might be related to other factors not related to glacier size, organic matter content, or geographical position.

The activity concentration of ^{210}Pb was positively related to the average organic matter content in cryoconite ($F_{1,15}$ = 69.962, p < 0.001; Table 1, Fig. 4). This inter-glacier relation is consistent with those observed in previous studies relating ^{210}Pb concentrations to OM contents of individual cryoconite samples within single glaciers [28,29]. It was well-documented that negatively charged bivalent ^{210}Pb binds effectively to organic matter in various habitats [68,69]. The activity concentration of ^{137}Cs was also positively related to the organic matter content of cryoconite ($F_{1,16}$ = 6.4, p = 0.022; Table 1, Fig. 5), but this relationship was weaker, and the estimate error was higher (Table 1, Figs. 4 and 5). This aligns with the results of Buda et al., [28] and Davidson et al., [46], who, based on different approaches, confirmed that a significant proportion of ^{137}Cs in cryoconite is bound to mineral fractions rather than to organic matter itself, while ^{210}Pb binds more strongly with the organic matter surfaces. This finding is interesting as it suggests that the OM-minerals relationship may also influence the relationship between ^{137}Cs and OM in cryoconite. For instance, Buda et al., [28] observed that cryoconite with higher OM is also

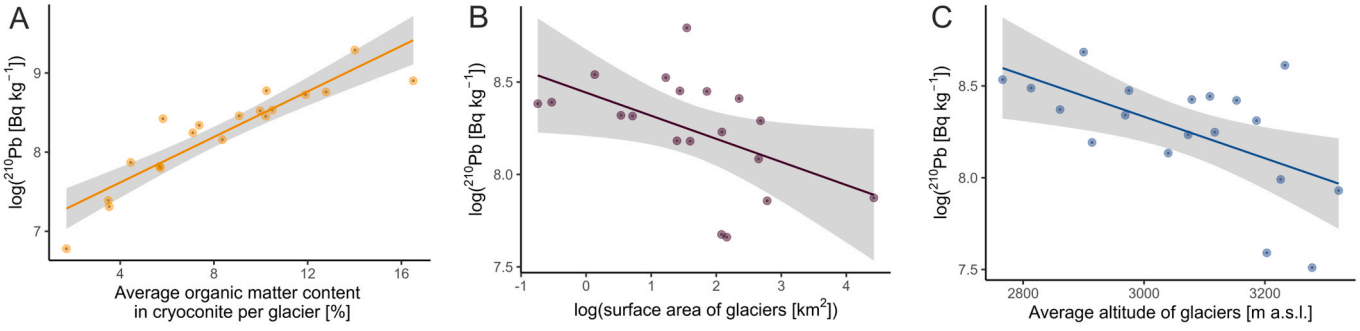

Fig. 4. Relationship between ^{210}Pb activity concentration (log-scaled) and glacier-specific variables such as organic matter content in cryoconite (A), surface area of glaciers (B), and average altitude of a glacier (C). Plots represent selected model visualisation with partial residuals (dots). Therefore, each graph shows the relationship between a given independent variable and the ^{210}Pb while accounting for the effects of the other independent variables in the model.

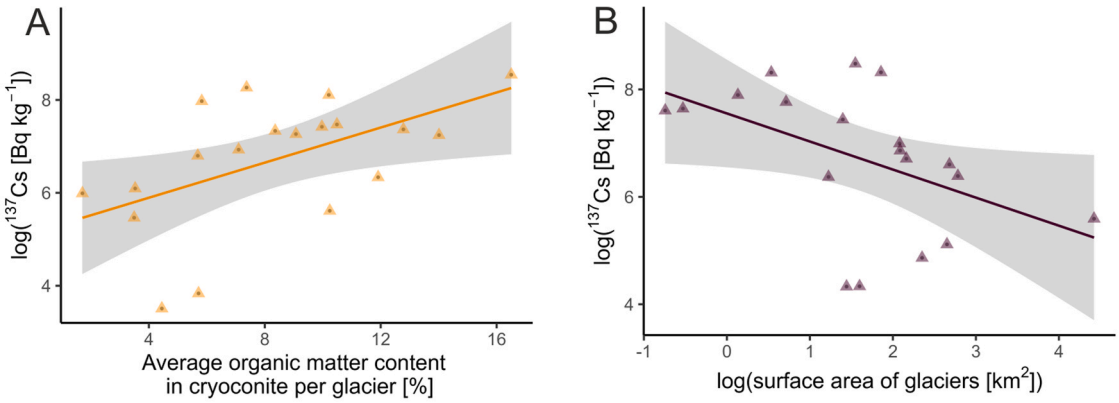

Fig. 5. Relationship between ^{137}Cs activity concentration (log-scaled) and glacier-specific variables such as organic matter content in cryoconite (A) and surface area of glaciers (B). Plots represent selected models visualisation with partial residuals (dots). Therefore, each graph shows the relationship between a given independent variable and the ^{210}Pb while accounting for the effects of the other independent variables in the model.

characterised by a smaller fraction of minerals, leading to a larger surface area of minerals. Moreover, for ^{241}Am, the organic matter content was not related to its activity concentration ($F_{1,14} = 0.292$, $p = 0.598$; Table 1). Overall, the importance of organic matter in binding radioisotopes depends on the chemical properties of the radioisotope. However, as the measurement of organic matter is straightforward and the explained variation is relatively large, it is a good predictor of the activity concentration of some radionuclides on glaciers.

3.3. *Cryoconite on small disappearing glaciers as hotspots of radioisotopes*

Understanding the fate of pollutants stored on glaciers upon melting and which glaciers are the most contaminated is crucial for assessing potential threats to downstream ecosystems. In this study, we observed a negative relationship between the activity concentrations in cryoconite and the surface area of glaciers for all subject radionuclides (^{210}Pb: $F_{1,15} = 5.21$, $p = 0.037$; ^{137}Cs: $F_{1,16} = 4.878$, $p = 0.042$; ^{241}Am: $F_{1,14} = 5.208$, $p = 0.039$; Table 1, Figs. 4 and 5). This relationship might be due to the concentration of radioisotopes by cryoconite through intense glacier melting in the past, as well as long-term deposition and accumulation of organic matter along with other components of cryoconite on the glacier surface. During ablation, the surface of a glacier becomes a dynamic habitat [70,71] where cryoconite is redistributed and can effectively capture radioisotopes [28,41,72].

The observed high activity concentrations of radionuclides on smaller glaciers might be related to the release of radioisotopes during the intensive melting of accumulation zones in the last decades. The accumulation zone of glaciers in the Alps moved upward by about 240 m since the 1960s and 1970s compared to 2009 despite their average altitude remained approximatly the same [73]. This suggests that a significant amount of relatively young snow and firn from the lower parts of accumulation zones melted and released stored radioisotopes over this period. In the case of small glaciers, we could expect lower dilution of those radioisotopes than in the case of big glaciers. Another possible explanation is related to variation in surface ice age. Ice that melts currently in the ablation zone of bigger glaciers can be older than that in the ablation zone of smaller glaciers. Therefore, cryoconite on bigger glaciers in the last decades was in contact with ice that formed hundreds or thousands of years in the past, significantly before the nuclear era and the release of anthropogenic radioisotopes, but also old enough to decay ^{210}Pb ($t_{1/2} = 20.3$ yrs.). In such a scenario, only the accumulation of radioactivity from the atmosphere affected the concentration of fallout radionuclides in cryoconite. This is also valid for small glaciers, where radioisotopes bind to cryoconite continuously from the atmosphere (^{210}Pb). Taking into account that the age of surface ice in the border between accumulation and ablation zones of smaller glaciers can be relatively young (even below 50 yrs.; [74]), progressive melting releases radioisotopes stored in the ice of the accumulation zone (^{210}Pb, ^{137}Cs and ^{241}Am), which are then concentrated in the cryoconite on the surface of ablation zone. However, to fully understand these potential reasons, more detailed data about the age of surface ice in the cryoconite zone on different glaciers in the Alps is needed.

Globally, glaciers are melting rapidly, a trend that has accelerated since the end of the 19th century [75-77]. Anthropogenic radioisotopes accumulated in large glaciers, especially those with a half-life too short compared to the persistence of ice within a glacier, do not pose a real threat to downstream ecosystems due to radioactivity. Our results show that many anthropogenic radioisotopes persist on glaciers and concentrate there during melting. This is consistent with Cao et al., [78] and Clason et al., [79], who demonstrated a high disproportion in the concentrations of radioisotope activities on glaciers compared to downstream habitats. Therefore, when considering large glaciers, we could observe a decrease in the radioactivity of these short-lived radionuclides before the ice melts. However, we think that small glaciers, which are the most numerous in low and mid-latitude mountains and are highly threatened with disappearance [80], are a special case. As radionuclides concentrate in the supraglacial sediment as melting progresses (Figs. 4 and 5), we could observe a relatively high peak of released radioactivity at the complete melt of glaciers. This is possible even for short-lived radionuclides, as around half of the smallest glaciers in the Swiss Alps are expected to melt within the next 25 years [80], which is within the range of the half-life of the radioisotopes considered in this study. Such a scenario also seems to hold for Scandinavian glaciers that store a high amount of radioisotopes [7,79] and 98 % of them are predicted to disappear by 2100 [81]. On the one hand, ^{210}Pb – a natural isotope with constant deposition – will concentrate on glaciers during melting and then be released to downstream ecosystems from glaciers. A current comparison of activity concentration of fallout radionuclides shows that still some share of radioisotopes passes through glacier and reaches the proglacial zone, suggesting downstream accumulation of them with localised off-ice hotspots [79,82]. In a study in British Columbia, Canada, Owens et al., [72] documented declining activity concentration of fallout radionuclides in fluvial sediments with increasing distance from the glacier terminus, while studies in Arctic Sweden [79] and Svalbard [82] found that the activity concentration of some radionuclides were higher in sediments from southern proglacial outlets than in northern outlets, highlighting the importance of local variability in glacier topography and hydrology. However, given that a significant proportion of radioisotopes such as ^{137}Cs is bound to minerals [28], their mobility in downstream ecosystems might be limited, posing a real threat to glacier biota and strictly glacier-adjacent ecosystems which should be extensively monitored during ongoing glaciers melting. Moreover, already

reported activity concentrations of radioisotopes in proglacial sediments also suggest that the sediment from glaciers might be diluted by the debris, lowering the negative effect on biota.

Following Beard et al., [83], we emphasise the importance of monitoring radioactive contamination on glaciers and glacier-adjacent ecosystems as the radiative pressure seems to increase in cryoconite and glaciers melting. In our perspective, a particular case are mountain glaciers. On the one side, their are located in areas with high accumulation of anthropogenic radioisotopes in the past. However, on the other hand, they terminate on the lands, providing water for people living down in valleys. Knowing that mountain glaciers in the northern hemisphere [29] show the highest radioactivity levels, we underline the need to monitor radioactivity pressure on glaciers and downstream ecosystems in the Alps, the Caucasus, central-Asian and North American mountain ranges.

4. Conclusion

The discussion on the glacier-related pollution increases in the literature. However, the debate mainly focuses on the direct measurements of the contaminants on glaciers, while the anticipation of hazardous glacial hotspots still needs to be included. In the context of intensive glacier melting, assessing the potential threats of stored pollutants to understand their impact on downstream ecosystems is crucial. Our comprehensive sampling across the Alps has demonstrated that glacier features such as surface area, altitude (^{210}Pb), and the amount of OM in the cryoconite play a more significant role in the accumulation of radioisotopes than geographic factors. Apart from the organic matter content in cryoconite and elevation (^{210}Pb), the surface area emerges as the most critical predictor of radioisotope activity concentrations on Alpine glaciers. The activity concentration of both natural (^{210}Pb) and anthropogenic (^{137}Cs, ^{241}Am) radioisotopes increases as the surface area decreases. This suggests that a significant proportion of radioactivity is bound to the supraglacial sediment, known as cryoconite, during the melting of ice, which contains natural and anthropogenic fallout radionuclides. This might be important, as the small glaciers in the Alps are predicted to disappear within the next 50 years, releasing stored radioisotopes. If so, we could witness intense abiotic pressure on glacier-adjacent ecosystems as glaciers disappear. Multidimensional studies are required to understand the potential effects of increasing radioactivity, seemingly linked to ongoing glacier melting.

Environmental implication

In the proposed article, we aimed to determine what factors affect the accumulation of radioisotopes on glaciers in the Alps which led us to understand whether predicted global climate change can increase pressure on organisms related to glaciers due to the concentration and release of anthropogenic and natural radioisotopes stored in the ice. Our study based on wide sampling in the Alps revealed that cryoconite on small glaciers concentrate anthropogenic radioisotopes such as ^{137}Cs and ^{241}Am during melting. Given the near disappearance of numerous small alpine glaciers over the next five decades, it is anticipated that the release of anthropogenic radioisotopes into mountain ecosystems may surpass excess predictions. This poses a significant threat to the ecosystems adjacent to glaciers.

CRediT authorship contribution statement

Jakub Buda: Writing – review & editing, Writing – original draft, Visualization, Validation, Resources, Project administration, Methodology, Investigation, Funding acquisition, Formal analysis, Data curation, Conceptualization. **Sylwia Błażej:** Investigation. **Roberto Ambrosini:** Writing – review & editing, Resources, Investigation. **Riccardo Scotti:** Writing – review & editing, Investigation. **Francesca Pittino:** Writing – review & editing, Investigation. **Dariusz Sala:** Writing – review & editing, Investigation. **Krzysztof Zawierucha:** Writing – review & editing, Supervision, Resources, Investigation, Funding acquisition. **Edyta Łokas:** Writing – review & editing, Supervision, Resources, Investigation, Funding acquisition, Conceptualization.

Declaration of Competing Interest

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

Data Availability

The data and the R code are shared as link in the Statistical Analysis Section.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jhazmat.2024.135083.

References

[1] Gabrieli, J., Cozzi, G., Vallelonga, P., Schwikowski, M., Sigl, M., Eickenberg, J., et al., 2011. Contamination of Alpine snow and ice at Colle Gnifetti, Swiss/Italian Alps, from nuclear weapons tests. Atmos Environ 45 (3), 587–593. https://doi.org/10.1016/J.ATMOSENV.2010.10.039.

[2] Gallini, L., Ajmone-Marsan, F., Scalenghe, R., 2018. The contamination legacy of a decommissioned iron smelter in the Italian Alps. J Geochem Explor 186, 121–128. https://doi.org/10.1016/J.GEXPLO.2017.12.013.

[3] Panagos, P., Van Liedekerke, M., Yigini, Y., Montanarella, L., 2013. Contaminated sites in Europe: Review of the current situation based on data collected through a European network. J Environ Public Health 2013. https://doi.org/10.1155/2013/158764.

[4] Baccolo, G., Łokas, E., Gaca, P., Massabò, D., Ambrosini, R., Azzoni, R.S., et al., 2020. Cryoconite: an efficient accumulator of radioactive fallout in glacial environments. Cryosphere 14 (2), 657–672. https://doi.org/10.5194/TC-14-657-2020.

[5] Foucher, A., Chaboche, P.A., Sabatier, P., Evrard, O., 2021. A worldwide meta-analysis (1977-2020) of sediment core dating using fallout radionuclides including 137Cs and 210Pbxs. Earth Syst Sci Data 13 (10), 4951–4966. https://doi.org/10.5194/ESSD-13-4951-2021.

[6] Krouglov, S.V., Kurinov, A.D., Alexakhin, R.M., 1998. Chemical fractionation of 90Sr, 106Ru, 137Cs and 144Ce in Chernobyl-contaminated soils: an evolution in the course of time. J Environ Radioact 38 (1), 59–76. https://doi.org/10.1016/S0265-931X(97)00022-2.

[7] Łokas, E., Wachniew, P., Baccolo, G., Gaca, P., Janko, K., Milton, A., et al., 2022. Unveiling the extreme environmental radioactivity of cryoconite from a Norwegian glacier. Sci Total Environ 814. https://doi.org/10.1016/j.scitotenv.2021.152656.

[8] Mietelski, J.W., Maksimova, S., Szwałko, P., Wnuk, K., Zagrodzki, P., Błazej, S., et al., 2010. Plutonium, 137Cs and 90Sr in selected invertebrates from some areas around Chernobyl nuclear power plant. J Environ Radioact 101 (6), 488–493. https://doi.org/10.1016/J.JENVRAD.2008.04.009.

[9] Łokas, E., Zawierucha, K., Cwanek, A., Szufa, K., Gaca, P., Mietelski, J.W., et al., 2018. The sources of high airborne radioactivity in cryoconite holes from the Caucasus (Georgia). Sci Rep 2018 8:1 8 (1), 1–10. https://doi.org/10.1038/s41598-018-29076-4.

[10] Minenko, V., Kukhta, T., Trofimik, S., Zhukova, O., Podgaiskaya, M., Viarenich, K., et al., 2022. Evaluation of 131I transfer in the environment based on the available measurements made in Belarus after the Chernobyl accident. J Environ Radioact 250, 106928. https://doi.org/10.1016/J.JENVRAD.2022.106928.

[11] Copplestone, D., Johnson, M.S., Jones, S.R., Toal, M.E., Jackson, D., 1999. Radionuclide behaviour and transport in a coniferous woodland ecosystem: vegetation, invertebrates and wood mice, Apodemus sylvaticus. Sci Total Environ 239 (1–3), 95–109. https://doi.org/10.1016/S0048-9697(99)00294-6.

[12] Dragović, S., Mandić, L.J., 2010. Transfer of radionuclides to ants, mosses and lichens in semi-natural ecosystems. Radiat Environ Biophys 49 (4), 625–634. https://doi.org/10.1007/S00411-010-0319-8/TABLES/8.

[13] Mietelski, J.W., Dubchak, S., Błazej, S., Anielska, T., Turnau, K., 2010. 137Cs and 40K in fruiting bodies of different fungal species collected in a single forest in southern Poland. J Environ Radioact 101 (9), 706–711. https://doi.org/10.1016/J.JENVRAD.2010.04.010.

[14] Hu, Q.H., Weng, J.Q., Wang, J.S., 2010. Sources of anthropogenic radionuclides in the environment: a review. J Environ Radioact 101 (6), 426–437. https://doi.org/10.1016/J.JENVRAD.2008.08.004.

[15] Ashraf, M.A., Akib, S., Maah, M.J., Yusoff, I., Balkhair, K.S., 2014. Cesium-137: radio-chemistry, fate, and transport, remediation, and future concerns. Crit Rev Environ Sci Technol 44 (15), 1740–1793. https://doi.org/10.1080/10643389.2013.790753.

[16] Singh, B.S.M., Dhal, N.K., Kumar, M., Mohapatra, D., Seshadri, H., Rout, N.C., et al., 2022. Phytoremediation of 137Cs: factors and consequences in the environment. *Radiat Environ Biophys 2022 61*:3 61 (3), 341–359. https://doi.org/10.1007/S00411-022-00985-3.

[17] Liao, H., Bu, W., Zheng, J., Wu, F., Yamada, M., 2014. Vertical distributions of radionuclides (239+240Pu, 240Pu/239Pu, and 137Cs) in sediment cores of lake bosten in Northwestern China. Environ Sci Technol 48 (7), 3840–3846. https://doi.org/10.1021/ES405364M/SUPPL_FILE/ES405364M_SI_001.PDF.

[18] Geras'kin, S.A., Yoschenko, V., Bitarishvili, S., Makarenko, E., Vasiliev, D., Prazyan, A., et al., 2021. Multifaceted effects of chronic radiation exposure in Japanese red pines from Fukushima prefecture. Sci Total Environ 763, 142946. https://doi.org/10.1016/J.SCITOTENV.2020.142946.

[19] Garnier-Laplace, J., Beaugelin-Seiller, K., Della-Vedova, C., Métivier, J.M., Ritz, C., Mousseau, T.A., et al., 2015. Radiological dose reconstruction for birds reconciles outcomes of Fukushima with knowledge of dose-effect relationships. Sci Rep 2015 5:1 5 (1), 1–13. https://doi.org/10.1038/srep16594.

[20] Nybakken, L., Lee, Y.K., Brede, D.A., Mageroy, M.H., Lind, O.C., Salbu, B., et al., 2023. Long term effects of ionising radiation in the Chernobyl Exclusion zone on DNA integrity and chemical defence systems of Scots pine (Pinus sylvestris). Sci Total Environ 904, 166844. https://doi.org/10.1016/J.SCITOTENV.2023.166844.

[21] Jernfors, T., Lavrinienko, A., Vareniuk, I., Landberg, R., Fristedt, R., Tkachenko, O., et al., 2024. Association between gut health and gut microbiota in a polluted environment. Sci Total Environ 914, 169804. https://doi.org/10.1016/J.SCITOTENV.2023.169804.

[22] Jernfors, T., Danforth, J., Kesäniemi, J., Lavrinienko, A., Tukalenko, E., Fajkus, J., et al., 2021. Expansion of rDNA and pericentromere satellite repeats in the genomes of bank voles Myodes glareolus exposed to environmental radionuclides. Ecol Evol 11 (13), 8754–8767. https://doi.org/10.1002/ECE3.7684.

[23] Erlinger, C., Lettner, H., Hubmer, A., Hofmann, W., Steinhäusler, F., 2008. Determining the Chernobyl impact on sediments of a pre-Alpine lake with a very comprehensive set of data. J Environ Radioact 99 (8), 1294–1301. https://doi.org/10.1016/J.JENVRAD.2008.03.012.

[24] Gerdol, R., Degetto, S., Mazzotta, D., Vecchiati, G., 1994. The vertical distribution of the Cs-137 derived from Chernobyl fall-out in the uppermost Sphagnum layer of two peatlands in the southern Alps (Italy). Water Air Soil Pollut 75 (1–2), 93–106. https://doi.org/10.1007/BF01100402/METRICS.

[25] Baccolo, G., Di Mauro, B., Massabò, D., Clemenza, M., Nastasi, M., Delmonte, B., et al., 2017. Cryoconite as a temporary sink for anthropogenic species stored in glaciers. Sci Rep 2017 7:1 7 (1), 1–11. https://doi.org/10.1038/s41598-017-10220-5.

[26] Baccolo, G., Nastasi, M., Massabò, D., Clason, C., Di Mauro, B., Di Stefano, E., et al., 2020. Artificial and natural radionuclides in cryoconite as tracers of supraglacial dynamics: Insights from the Morteratsch glacier (Swiss Alps). CATENA 191, 104577. https://doi.org/10.1016/J.CATENA.2020.104577.

[27] Tieber, A., Lettner, H., Bossew, P., Hubmer, A., Sattler, B., Hofmann, W., 2009. Accumulation of anthropogenic radionuclides in cryoconites on Alpine glaciers. J Environ Radioact 100 (7), 590–598. https://doi.org/10.1016/J.JENVRAD.2009.04.008.

[28] Buda, J., Łokas, E., Błażej, S., Gorzkiewicz, K., Buda, K., Ambrosini, R., et al., 2024. Unveiling threats to glacier biota: bioaccumulation, mobility, and interactions of radioisotopes with key biological components. Chemosphere 348, 140738. https://doi.org/10.1016/J.CHEMOSPHERE.2023.140738.

[29] Clason, C.C., Baccolo, G., Łokas, E., Owens, P.N., Wachniew, P., Millward, G.E., et al., 2023. Global variability and controls on the accumulation of fallout radionuclides in cryoconite. Sci Total Environ 894, 164902. https://doi.org/10.1016/J.SCITOTENV.2023.164902.

[30] Pittino, F., Buda, J., Ambrosini, R., Parolini, M., Crosta, A., Zawierucha, K., et al., 2023. Impact of anthropogenic contamination on glacier surface biota. Curr Opin Biotechnol 80. https://doi.org/10.1016/j.copbio.2023.102900.

[31] Pautler, B.G., Dubnick, A., Sharp, M.J., Simpson, A.J., Simpson, M.J., 2013. Comparison of cryoconite organic matter composition from Arctic and Antarctic glaciers at the molecular-level. Geochim Et Cosmochim Acta 104, 1–18. https://doi.org/10.1016/J.GCA.2012.11.029.

[32] Pittino, F., Maglio, M., Gandolfi, I., Azzoni, R.S., Diolaiuti, G., Ambrosini, R., et al., 2018. Bacterial communities of cryoconite holes of a temperate alpine glacier show both seasonal trends and year-to-year variability. Ann Glaciol 59 (77), 1–9. https://doi.org/10.1017/AOG.2018.16.

[33] Rozwalak, P., Podkowa, P., Buda, J., Niedzielski, P., Kawecki, S., Ambrosini, R., et al., 2022. Cryoconite – From minerals and organic matter to bioengineered sediments on glacier's surfaces. Sci Total Environ 807. https://doi.org/10.1016/j.scitotenv.2021.150874.

[34] 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 (56), 9–14. https://doi.org/10.3189/172756411795932010.

[35] Cook, J.M., Hodson, A.J., Irvine-Fynn, T.D.L., 2016. Supraglacial weathering crust dynamics inferred from cryoconite hole hydrology. Hydrol Process 30 (3), 433–446. https://doi.org/10.1002/HYP.10602.

[36] McIntyre, N.F. (2011). Cryoconite hole thermodynamics. ⟨Https://Doi.Org/10.1139/E84–016⟩, 21(2), 152–156. ⟨https://doi.org/10.1139/E84–016⟩.

[37] Buda, J., Łokas, E., Pietryka, M., Richter, D., Magowski, W., Iakovenko, N.S., et al., 2020. Biotope and biocenosis of cryoconite hole ecosystems on ecology glacier in the maritime Antarctic. Sci Total Environ 724. https://doi.org/10.1016/j.scitotenv.2020.138112.

[38] McCrimmon, D.O., Bizimis, M., Holland, A., Ziolkowski, L.A., 2018. Supraglacial microbes use young carbon and not aged cryoconite carbon. Org Geochem 118, 63–72. https://doi.org/10.1016/J.ORGGEOCHEM.2017.12.002.

[39] Zawierucha, K., Trzebny, A., Buda, J., Bagshaw, E., Franzetti, A., Dabert, M., et al., 2022. Trophic and symbiotic links between obligate-glacier water bears (Tardigrada) and cryoconite microorganisms. PLoS ONE 17 (1 January). https://doi.org/10.1371/journal.pone.0262039.

[40] Wejnerowski, Ł., Poniecka, E., Buda, J., Klimaszyk, P., Piasecka, A., Dziuba, M.K., et al., 2023. Empirical testing of cryoconite granulation: Role of cyanobacteria in the formation of key biogenic structure darkening glaciers in polar regions. J Phycol. https://doi.org/10.1111/JPY.13372.

[41] 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. https://doi.org/10.1016/J.CHEMOSPHERE.2016.06.051.

[42] 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. https://doi.org/10.1016/J.JENVRAD.2023.107206.

[43] 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 210Pb. J Environ Radioact 186, 90–100. https://doi.org/10.1016/J.JENVRAD.2017.06.020.

[44] Baccolo, G. , Łokas, E. , Gaca, P. , Massabò, D. , Ambrosini, R. , Azzoni, R.S. et al. (2019). Cryoconite as an efficient monitor for the deposition of radioactive fallout in glacial environments. ⟨https://doi.org/10.5194/TC-2019–176⟩.

[45] Megumi, K., Oka, T., Doi, M., Kimura, S., Tsujimoto, T., Ishiyama, T., et al., 1988. Relationships Between the Concentrations of Natural Radionuclides and the Mineral Composition of the Surface Soil. Radiat Prot Dosim 24 (1–4), 69–72. https://doi.org/10.1093/OXFORDJOURNALS.RPD.A080244.

[46] Davidson, H., Millward, G.E., Clason, C.C., Fisher, A., Taylor, A., 2023. Chemical availability of fallout radionuclides in cryoconite. J Environ Radioact 268–269, 107260. https://doi.org/10.1016/J.JENVRAD.2023.107260.

[47] Gorzkiewicz, K., Mietelski, J.W., Kierepko, R., Brudecki, K., 2019. Low-background, digital gamma-ray spectrometer with BEGe detector and active shield: commissioning, optimisation and software development. J Radioanal Nucl Chem 322 (3), 1311–1321. https://doi.org/10.1007/S10967-019-06853-7/FIGURES/10.

[48] Hoogsteen, M.J.J., Lantinga, E.A., Bakker, E.J., Tittonell, P.A. (2018). An Evaluation of the Loss-on-Ignition Method for Determining the Soil Organic Matter Content of Calcareous Soils. ⟨Https://Doi.Org/10.1080/00103624.2018.1474475⟩, 49(13), 1541–1552. ⟨https://doi.org/10.1080/00103624.2018.1474475⟩.

[49] Fick, S.E., Hijmans, R.J., 2017. WorldClim 2: new 1-km spatial resolution climate surfaces for global land areas. Int J Climatol 37 (12), 4302–4315. https://doi.org/10.1002/JOC.5086.

[50] Harris, I., Osborn, T.J., Jones, P., Lister, D., 2020. Version 4 of the CRU TS monthly high-resolution gridded multivariate climate dataset. Sci Data 2020 7:1 7 (1), 1–18. https://doi.org/10.1038/s41597-020-0453-3.

[51] QGIS. (2023). QGIS.org. QGIS Geographic Information System. Open Source Geospatial Foundation Project. ⟨http://qgis.org⟩. ⟨http://qgis.org⟩.

[52] Raup, B., Racoviteanu, A., Khalsa, S.J.S., Helm, C., Armstrong, R., Arnaud, Y., 2007. The GLIMS geospatial glacier database: A new tool for studying glacier change. Glob Planet Change 56 (1–2), 101–110. https://doi.org/10.1016/J.GLOPLACHA.2006.07.018.

[53] Pebesma, E., & Bivand, R. (2024). Classes and Methods for Spatial Data. *CRAN*. ⟨https://github.com/edzer/sp/⟩.

[54] Bivand, R., 2024. The analysis of spatial association by use of distance statistics. CRAN. https://doi.org/10.1111/J.1538-4632.1992.TB00261.X.

[55] Henningsen, A. (2022). Estimating Censored Regression Models in R using the censReg Package. In CRAN. CRAN.R-project.org.

[56] R Core Team. (2024). R: A language and environment for statistical computing (4.3.2). R Foundation for Statistical Computing.

[57] Lüdecke, D., Ben-Shachar, M., Patil, I., Waggoner, P., Makowski, D., 2021. performance: An R Package for Assessment, Comparison and Testing of Statistical Models. J Open Source Softw 6 (60), 3139. https://doi.org/10.21105/JOSS.03139.

[58] Clark, M.J., Smith, F.B., 1988. Wet and dry deposition of Chernobyl releases. Nat 1988 332:6161 332 (6161), 245–249. https://doi.org/10.1038/332245a0.

[59] Pálsson, S.E., Howard, B.J., Wright, S.M., 2006. Prediction of spatial variation in global fallout of 137Cs using precipitation. Sci Total Environ 367 (2–3), 745–756. https://doi.org/10.1016/J.SCITOTENV.2006.01.011.

[60] Baldoncini, M., Albéri, M., Bottardi, C., Minty, B., Raptis, K.G.C., Strati, V., et al., 2017. Exploring atmospheric radon with airborne gamma-ray spectroscopy. Atmos Environ 170, 259–268. https://doi.org/10.1016/J.ATMOSENV.2017.09.048.
[61] Feichter, J., Crutzen, P.J., 1990. Parameterization of vertical tracer transport due to deep cumulus convection in a global transport model and its evaluation with 222Radon measurements. Tellus B 42 (1), 100–117. https://doi.org/10.1034/J.1600-0889.1990.00011.X.
[62] Piliposian, G.T., Appleby, P.G., 2003. A simple model of the origin and transport of 222Rn and 210Pb in the atmosphere. Contin Mech Thermodyn 15 (5), 503–518. https://doi.org/10.1007/S00161-003-0129-1/METRICS.
[63] Wilkening, M.H., Clements, W.E., 1975. Radon 222 from the ocean surface. J Geophys Res 80 (27), 3828–3830. https://doi.org/10.1029/JC080I027P03828.
[64] Bourcier, L., Masson, O., Laj, P., Pichon, J.M., Paulat, P., Freney, E., et al., 2011. Comparative trends and seasonal variation of 7Be, 210Pb and 137Cs at two altitude sites in the central part of France. J Environ Radioact 102 (3), 294–301. https://doi.org/10.1016/J.JENVRAD.2010.12.005.
[65] Rastogi, N., Sarin, M.M., 2008. Atmospheric 210Pb and 7Be in ambient aerosols over low- and high-altitude sites in semiarid region: Temporal variability and transport processes. J Geophys Res Atmosph 113 (D11), 11103. https://doi.org/10.1029/2007JD009298.
[66] Isakar, K., Kiisk, M., Realo, E., Suursoo, S., 2015. Lead-210 in the atmospheric air of North and South Estonia: long-term monitoring and back-trajectory calculations. Proc Est Acad Sci 64, 442–451. https://doi.org/10.3176/proc.2016.4.11.
[67] Sykora, I., Froehlich, K., 2009. Chapter 3 Radionuclides as Tracers of Atmospheric Processes. Radioact Environ 16, 51–88. https://doi.org/10.1016/S1569-4860(09)01603-9.
[68] El-Daoushy, F., Garcia-Tenorio, R., 1988. Speciation of Pb-210/Po-210 in aquatic systems and their deposits. Sci Total Environ 69 (C), 191–209. https://doi.org/10.1016/0048-9697(88)90343-9.
[69] Mihailović, A., Vučinić Vasić, M., Todorović, N., Hansman, J., Vasin, J., Krmar, M., 2014. Potential factors affecting accumulation of unsupported 210Pb in soil. Radiat Phys Chem 99, 74–78. https://doi.org/10.1016/J.RADPHYSCHEM.2014.02.020.
[70] Takeuchi, N., Sakaki, R., Uetake, J., Nagatsuka, N., Shimada, R., Niwano, M., et al., 2018. Temporal variations of cryoconite holes and cryoconite coverage on the ablation ice surface of Qaanaaq Glacier in northwest Greenland. Ann Glaciol 59 (77), 21–30. https://doi.org/10.1017/AOG.2018.19.
[71] Zawierucha, K., Buda, J., Nawrot, A., 2019. Extreme weather event results in the removal of invertebrates from cryoconite holes on an Arctic valley glacier (Longyearbreen, Svalbard). Ecol Res 34 (3). https://doi.org/10.1111/1440-1703.1276.
[72] 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 2019 9:1 9 (1), 1–9. https://doi.org/10.1038/s41598-019-48873-z.
[73] Carturan, L., Filippi, R., Seppi, R., Gabrielli, P., Notarnicola, C., Bertoldi, L., et al., 2013. Area and volume loss of the glaciers in the Ortles-Cevedale group (Eastern Italian Alps): controls and imbalance of the remaining glaciers. Cryosphere 7 (5), 1339–1359. https://doi.org/10.5194/TC-7-1339-2013.
[74] Festi, D., Schwikowski, M., Maggi, V., Oeggl, K., Jenk, T.M., 2021. Significant mass loss in the accumulation area of the Adamello glacier indicated by the chronology of a 46m ice core. Cryosphere 15 (8), 4135–4143. https://doi.org/10.5194/TC-15-4135-2021.
[75] Marta, S., Azzoni, R.S., Fugazza, D., Tielidze, L., Chand, P., Sieron, K., et al., 2021. The retreat of mountain glaciers since the little ice age: a spatially explicit database. Data 6 (10), 107. https://doi.org/10.3390/DATA6100107.
[76] Pörtner, H.O., Roberts, D.C., Masson-Delmotte, V., Zhain, P., Tignor, E., Poloczanska, E., et al., 2019. The Ocean and Cryosphere in a Changing Climate. In The Ocean and Cryosphere in a Changing Climate. Cambridge University Press. ⟨https://doi.org/10.1017/9781009157964⟩.
[77] Zemp, M., Nussbaumer, S.U., Naegeli, K., Gärtner-Roer, I., Paul, F., Hoelzle, M., et al., 2013. Glacier mass balance bulletin no. 12 (2010-2011). Glacier Mass Balance Bull 12.
[78] 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. https://doi.org/10.1016/J.SCITOTENV.2023.164140.
[79] Clason, C.C., Blake, W.H., Selmes, N., Taylor, A., Boeckx, P., Kitch, J., et al., 2021. Accumulation of legacy fallout radionuclides in cryoconite on Isfallsglaciären (Arctic Sweden) and their downstream spatial distribution. Cryosphere 15 (11), 5151–5168. https://doi.org/10.5194/TC-15-5151-2021.
[80] Huss, M., Fischer, M., 2016. Sensitivity of very small glaciers in the swiss alps to future climate change. Front Earth Sci 4, 180679. https://doi.org/10.3389/FEART.2016.00034/BIBTEX.
[81] Nesje, A., Bakke, J., Dahl, S.O., Lie, Ø., Matthews, J.A., 2008. Norwegian mountain glaciers in the past, present and future. Glob Planet Change 60 (1–2), 10–27. https://doi.org/10.1016/J.GLOPLACHA.2006.08.004.
[82] 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. https://doi.org/10.1016/J.JENVRAD.2017.08.018.
[83] Beard, D.B., Clason, C.C., Rangecroft, S., Poniecka, E., Ward, K.J., Blake, W.H., 2022. Anthropogenic contaminants in glacial environments II: Release and downstream consequences. Prog Phys Geogr 46 (5), 790–808. https://doi.org/10.1177/03091333221127342/ASSET/IMAGES/LARGE/10.1177_03091333221127342-FIG2.JPEG.