

Artificial and natural radionuclides in cryoconite as tracers of supraglacial dynamics

Giovanni Baccolo^{1,2*}, Massimiliano Nastasi^{2,3}, Dario Massabò^{4,5}, Caroline Clason⁶, Biagio Di Mauro¹, Elena Di Stefano^{1,2,7}, Edyta Łokas⁸, Paolo Prati^{4,5}, Ezio Previtali^{2,3}, Nozomu Takeuchi⁹, Barbara Delmonte^{1,2}, Valter Maggi^{1,2}

1. Environmental and Earth Sciences Department, University of Milano-Bicocca, P.za della Scienza n.1, Milano, 20126, Italy
2. INFN section of Milano-Bicocca, P.za della Scienza n.3, Milano, 20126, Italy
3. Physics Department, University of Milano-Bicocca, P.za della Scienza n.3, Milano, 20126, Italy
4. Physics Department, University of Genova, Genova, 16146, Italy
5. INFN section of Genova, Genova, 16146, Italy
6. School of Geography, Earth and Environmental Sciences, University of Plymouth, Plymouth, PL48AA, UK
7. Department of Physical, Earth and Environmental Sciences, University of Siena, Siena, 53100, Italy
8. Department of Mass Spectrometry, Institute of Nuclear Physics Polish Academy of Sciences, Kraków, 31-342, Poland
9. Department of Earth Sciences, Graduate School of Science, Chiba University, Chiba, Japan

* corresponding author: giovanni.baccolo@unimib.it

Abstract

Cryoconite, a sediment found on the surface of glaciers, is known for its ability to accumulate radionuclides. New data on cryoconite from the Morteratsch glacier (Switzerland) are presented with the aim to shed light on the mechanisms that control the distribution of radioactivity in cryoconite. Among the many radionuclides detected in our samples, we have identified ^{108m}Ag, an artificial species which has never been observed in terrestrial environments before. This finding supports that cryoconite has an extraordinary ability to accumulate radioactivity. Our results also show that the radioactivity of cryoconite is far from uniform. Both the absolute amount of radioactivity and the relative contribution of single radionuclides is highly variable in samples from the Morteratsch glacier. To investigate the processes responsible for such variability, we have explored the correlation between radionuclides, organic and inorganic carbon fractions and the morphological features of cryoconite deposits. We have found that the degree to cryoconite is connected with supraglacial hydrology is particularly important, since it strongly influences the accumulation of radionuclides in cryoconite. Cryoconite holes connected with supraglacial channels is rich in cosmogenic ⁷Be; in contrast, poorly connected deposits are rich in artificial fallout radionuclides and elemental carbon. The very different half-lives of ⁷Be and artificial radionuclides allowed us to discuss our findings in relation to the age and maturity of cryoconite deposits, highlighting the potential use of radionuclides to investigate hydrological supraglacial processes and material cycling at the surface of glaciers.

Keywords

40 Cryoconite; Environmental Radioactivity; ^7Be ; Supraglacial Processes; Glacial Hydrology; Artificial Radionuclides

1. Introduction

45 Radioactivity is a versatile tool in Earth Sciences, with many diverse applications. When focusing on the Earth's surface, common uses of radioactivity concern the study of fallout radionuclides (FRN) to develop chronologies for recent sediments (Appleby, 2008) and to investigate erosion and sedimentation processes (Mabit et al., 2008). One research field that owes much to radioactivity is glaciology. The first glaciological applications of radioactivity date back to the 1960s, when pioneering studies on the development of age scales for snow and ice through the detection of FRN appeared (Picciotto and Wilgain, 1963; Crozaz et al., 1964). Since then, the analysis of the decay rate of natural fallout ^{210}Pb and the identification of radioactive spikes corresponding to nuclear accidents and test explosions have become a routine procedure to date the upper layers of ice cores (Eichler et al., 2000; Clemenza et al., 2012). This has been possible because snow and ice, being atmospheric in origin, record the atmospheric history of a given region (Preunkert & Legrand, 2013), radioactive fallout included (Pinglot et al., 1994). But glaciers are not isolated systems, they are not exclusively a sink for fallout material, since through ablation and melt part of the airborne matter accumulated into glaciers is released in the downstream environment. This is now widely recognized, and glaciers are no longer considered simply as repositories, but also as secondary sources for many impurities of ecological importance (Gerringa et al., 2012), including pollutants (Bogdal et al., 2009; Rizzi et al., 2019) and radionuclides (Łokas et al., 2017; Owens et al., 2019).

Within this context radioactivity plays an important role. In recent years it has been observed that before being released in pro-glacial areas, FRN stored in glaciers are accumulated at their surface in association with cryoconite (Tieber et al., 2009; Łokas et al., 2016; Baccolo et al., 2019). Cryoconite is the dark sediment that forms at the surface of many glaciers across the global cryosphere during the melt season. It is mostly composed of fine mineral sediments, but it is also rich in organic matter, owing to notable microbial activity (Hodson et al., 2010). Its dark color, related to high organic content (Takeuchi, 2002), impacts the radiative properties of ice, enhancing its melt (Di Mauro et al., 2017; Li et al., 2019). The abundance of organic matter and the interaction with meltwater makes cryoconite a natural filter, capable of accumulating diverse impurities originally present in ice and mobilized through meltwater (Baccolo et al., 2017). In particular, it has been observed that pollutants are efficiently accumulated in cryoconite, including heavy metals (Nagatsuka et al., 2010; Łokas et al., 2016; Baccolo et al., 2017; Huang et al., 2019), organic compounds (Li et al., 2017; Weiland-Bräuer et al., 2017), pesticides (Ferrario et al., 2017) and radionuclides (Tieber et al., 2009; Łokas et al., 2016; Baccolo et al., 2019).

75 Despite early evidence about the ability of cryoconite to accumulate radionuclides, Tieber and coauthors (2009) were the first to present a quantitative radioecological characterization of cryoconite. They showed that the activity concentration of single radionuclides in cryoconite from an Austrian glacier exceeded $100,000 \text{ Bq Kg}^{-1}$ (Tieber et al., 2009). Similar evidence was reported in

80 following years for other geographic contexts (Łokas et al., 2016; Baccolo et al., 2019; Owens et al.,
2019), confirming that cryoconite accumulates FRN with unprecedented efficiency. The typical
activities observed in cryoconite are orders of magnitude higher than those characterizing the
matrices usually adopted for environmental radioactivity monitoring, such as lichens and mosses.
In terms of radioactive contamination, only samples from nuclear accident and test explosion sites
are comparable to or exceed cryoconite (Baccolo et al., 2019). Recent works have suggested that
85 the ability of cryoconite to retain and accumulate radionuclides depends on its composition, which
is rich in organic matter, and on its interaction with the meltwater that flows at the surface of
glaciers during summer (Łokas et al., 2016; Baccolo et al., 2019). Meltwater is in fact the means by
which the impurities deposited in the past on the glacier, and subsequently preserved in snow and
ice layers, come into contact with cryoconite (Baccolo et al., 2019). Studies have shown that
90 cryoconite accumulates both natural FRN, such as ^{210}Pb and ^7Be , and anthropogenic FRN, such as
 ^{137}Cs , Pu and Am isotopes, which were released in the environment as a consequence of nuclear
accidents, test explosions and atmospheric re-entries of nuclear powered satellites (Tieber et al.,
2009; Baccolo et al., 2019; Łokas et al., 2019). After being accumulated at the surface of glaciers,
cryoconite and its radioactive content are released by glaciers in association with meltwater.
95 Preliminary results have shown that a fraction of cryoconite radioactivity is accumulated in pro-
glacial areas, and pose a risk of potential ecological impacts (Łokas et al., 2014,2017,2019; Owens
et al., 2019).

In this study we present data concerning cryoconite collected at the Morteratsch glacier, in the Swiss
Alps. We aim to understand whether cryoconite radioactivity is uniform across the surface of
100 glaciers, or if it is influenced by local factors, such as the position of cryoconite deposits on the ice
surface, the connection with the supraglacial hydrological system, and its aggregation state. It is
well known that the supraglacial environment is highly dynamic, in particular when considering the
lower sectors of glaciers, where ice melt and motion, liquid precipitation, and meltwater runoff
continuously shape the glacial surface (Rippin et al., 2015; Rossini et al., 2018). The effects of these
105 processes on cryoconite are manifold, affecting the morphology and distribution of its deposits
(Cook et al., 2016; Takeuchi et al., 2018), its biological communities (Pittino et al., 2018; Zawierucha
et al., 2019) and its biogeochemical composition (Bagshaw et al., 2007). Improved knowledge on
these processes is necessary to better understand how the transience of the supraglacial
environment influences the radioactive content of cryoconite, and if the latter can be used as a
110 tracer to provide insight into the dynamics of supraglacial hydrology.

2. Study site and sampling

Cryoconite was sampled at the Vadret da Morteratsch glacier on July 18th 2018. The glacier is the
largest in the Bernina range (Rhaetian Alps, Switzerland, see Fig. 1). It is a north-facing valley glacier
whose basin extends from 2100 to 4049 m a.s.l.. The glacier is experiencing a rapid retreat as a
115 consequence of a strong imbalance between its geometry and contemporary climate. Retreat and
thinning are contributing to a significant darkening of the ice surface in the lower sectors of the
glacier, with the emergence of sub glacial debris (Rossini et al., 2018) and the deposition of mineral
dust from the surrounding moraines (Oerlemans et al., 2009). This environment is favorable for the
formation of cryoconite, which has previously been investigated at this site in relation to the effects
120 on the optical properties of ice (Di Mauro et al., 2017) and to its ability to accumulate radioactivity

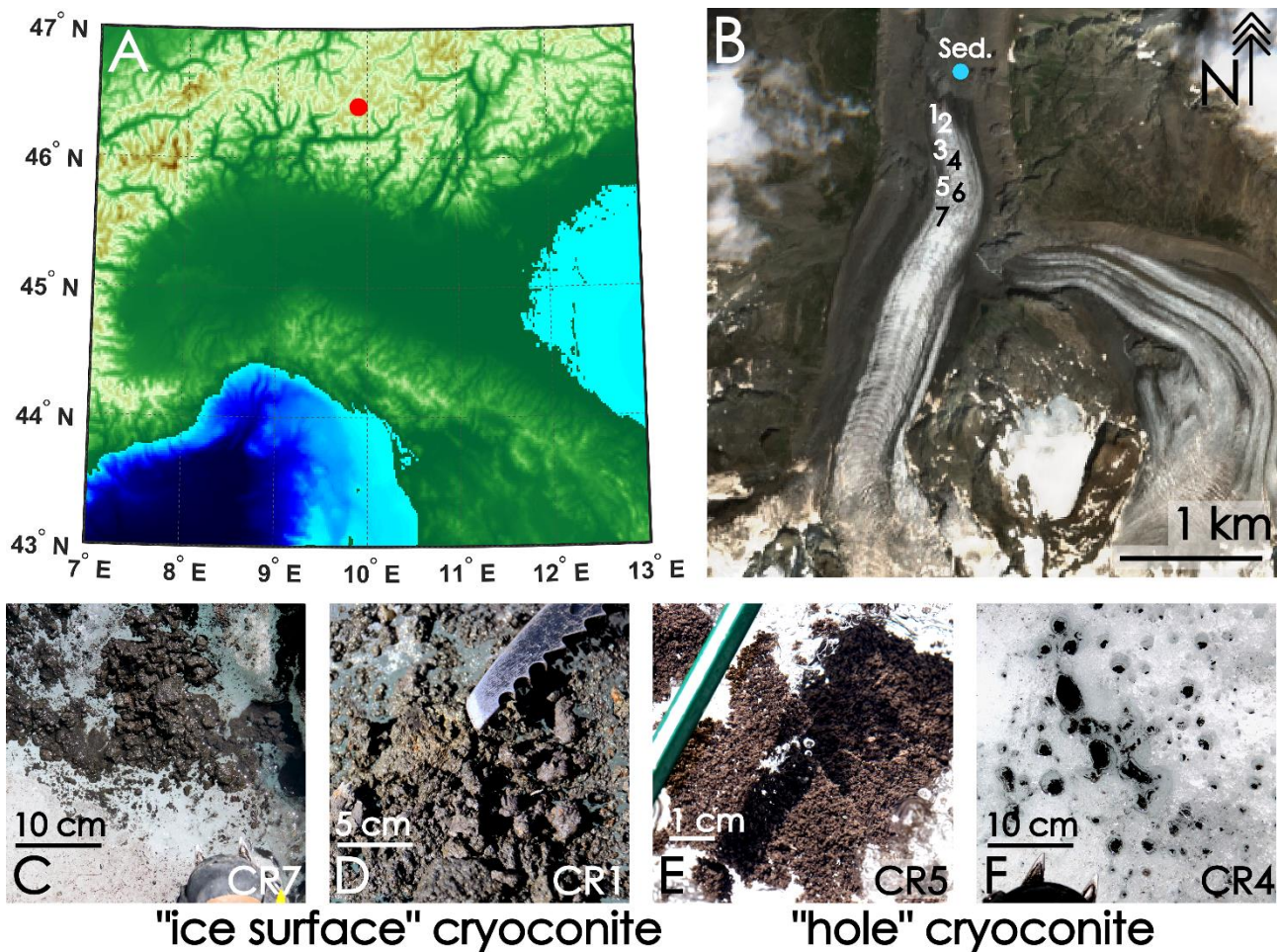


Fig. 1 The geographic setting of the present study. The relief map (panel A) shows the position of Vadret da Morteratsch within the Alps (red dot). In panel B a satellite picture of the glacier is presented (Sentinel 2, ESA), retrieved on July 16th, 2018. Sampling sites are highlighted by numbers corresponding to cryoconite sample codes, and by a blue dot (riverine proglacial sediments). In the lower row, examples of cryoconite samples belonging to the “ice surface” (C,D) and “hole” (E,F) types are shown.

125

130

135

140

and heavy metals (Baccolo et al., 2017). The sampling campaign of July 2018 was carried out to continue monitoring activities established in 2015. Attention was paid to the morphological features of cryoconite and to its proximity to supraglacial hydrology. It was noted during previous campaigns that cryoconite deposits at Vadret da Morteratsch can be distinguished in two types, namely “hole” and “ice surface” deposits. Cryoconite accumulated in deposits defined as “hole”, is found at the bottom of holes melted into ice which are filled with meltwater. The diameter of ice holes is variable, ranging from less than 1 cm to tens of cms. “Ice surface” cryoconite is accumulated in thick and massive deposits only poorly connected with meltwater. During our expeditions, we have always found both cryoconite types, but “hole” deposits are more common at the beginning of the melt season and at high elevation, while “ice surface” deposits are more commonly found in the lower sectors of the glacier and at the end of the melt season.

Since the main objective of our work is to understand if the radioactivity content of cryoconite is somehow influenced by cryoconite morphology and supraglacial processes, we planned the campaign for late July, in the middle of the melt season, so as to optimize the chances of finding both “hole” and “ice surface” deposits. We collected three cryoconite samples from deposits

145 corresponding to “hole” features, three from “ice surface” deposits and one from a deposit showing
“intermediate” characteristics. The elevation of cryoconite sampling sites ranged from 2100 to 2300
m a.s.l.. In addition, sediments have been collected near the terminus at a small proglacial river. All
the collected samples have been considered to the aims of this study and no selection has been
carried out. Example pictures of cryoconite types are shown in Fig. 1, while extensive descriptions
and photographs of our sampling sites/types are found in the Supplementary Material.

150 Sample collection was conducted with clean plastic spatulas and disposable pipettes, and samples
were stored in sterile plastic tubes. Cryoconite deposits with abundant material were chosen during
the field campaign, so as to obtain large samples and facilitate subsequent analyses. After the
collection, samples were kept at 0°C in a thermal bag during the campaign and subsequently stored
at -20°C in the Eurocold Laboratory of the Milano-Bicocca University, until the preparation for the
analyses.

155 3. Materials and methods

The radioactivity of cryoconite in our samples has been investigated through γ -spectrometry.
Samples were dried until a constant weight at about 60°C. Coarser fragments were manually
removed. Cryoconite was then stored in clean polyethylene Petri dishes (diameter 9.0 cm, height
1.5 cm) and sealed in plastic bags to prevent radon loss. Sample mass varied between 4 and 77 g
160 depending on availability. Dishes were chosen to increase the surface area to volume ratio of
samples, so as to limit radiation self-absorption and improve the analytical efficiency. Counting took
place some months after sealing the samples, when the secular equilibrium between ^{222}Rn and its
progenies was reached. A Broad Energy High Purity Germanium detector was used for the
acquisition of the spectra. The detector registers an energy spectrum from 3 to 3,000 keV, with the
165 following energy resolution: 0.5 keV at 59.5 keV, 1.2 keV at 661 keV and 1.6 keV at 1,332 keV
(intended as full width half maximum resolution). The detector is provided with a germanium crystal
(relative efficiency 50 %) whose active volume is 150 cm³. Each sample was counted for a variable
time ranging from 7 to 14 days in relation to its mass. After the acquisition, spectra were processed
for peak identification and fitting and the absolute detection efficiency was calculated via Monte
170 Carlo simulations (Baccolo et al., 2017). If possible, for each nuclide two γ -energies were considered
for the analysis. No corrections related to spectral interferences were required with the exception
of ^{226}Ra , as its main γ -ray (186.2 keV) overlapped that of ^{235}U at 185.7 keV. To disentangle the two
contributions an additional γ -line (163.4 keV) from ^{235}U was considered. A blank spectrum was
acquired by counting an empty Petri dish for 10 days. Traces of ^{214}Pb (22 ± 3 mBq), ^{214}Bi (20 ± 5 mBq)
175 and ^{40}K (80 ± 3 mBq) were detected and subtracted from the sample signals. The blank contribution
of ^{214}Pb and ^{214}Bi represents 1 % of the signal detected in the samples, while for ^{40}K the value is 0.3
%. Minimum detectable activities (MDA) have been calculated considering the background integral
counts below the peaks associated to the γ -energies of interest. They range from 0.1 Bq kg⁻¹ for ^{137}Cs
to 22 Bq kg⁻¹ for ^{230}Th . Full details concerning analytical performance are reported in the
180 Supplementary Material.

Most of the detected radionuclides belong to the ^{238}U (^{234}Th , $^{234\text{m}}\text{Pa}$, ^{230}Th , ^{226}Ra , ^{214}Pb , ^{214}Bi , ^{210}Pb),
 ^{235}U (^{235}U , ^{227}Th) and ^{232}Th (^{228}Ac , ^{224}Ra , ^{212}Pb , ^{212}Bi , ^{208}Tl) decay chains. Other observed nuclides are
cosmogenic (^7Be), primordial (^{40}K) or artificial ($^{108\text{m}}\text{Ag}$, ^{137}Cs , ^{207}Bi , ^{241}Am). Activity concentrations of
 ^7Be , $^{108\text{m}}\text{Ag}$, ^{137}Cs , ^{207}Bi , $^{210}\text{Pb}_{\text{exc}}$ and ^{241}Am have been corrected for decay to the sampling date.

185 Mean activities of ^{214}Pb and ^{214}Bi have been used to distinguish the atmospheric (excess or unsupported) and lithogenic (supported) fractions of ^{210}Pb .

The carbonaceous content of samples was also analyzed. Elemental and organic carbon (EC and OC respectively) were quantified through a thermo-optical method. A Sunset EC/OC analyzer (Sunset Lab Inc.) was used, following the NIOSH 5040 protocol (Birch and Cary, 1996). Samples were
190 suspended on quartz fiber filters (Pall, 2500QAO-UP, 47 mm diameter), after pre-firing at 700°C for 1 hour to remove contaminations. Filters were weighed before and after deposition in a conditioned room ($T = 20 \pm 1^\circ\text{C}$, rel. um. = $50 \pm 5\%$). The amount of cryoconite deposited on each filter was determined with an analytical microbalance (precision = $1\mu\text{g}$) operated in the conditioned room. Electrostatic interferences were prevented through a deionizing gun. EC and OC mass concentration
195 were inferred from the mass of cryoconite deposited on filters and the EC-OC surface concentration. OC was converted into organic matter (OM), following the convention by Pribyl (2010).

4. Results

4.1. Analytical performance

With respect to previous radiological measurements in cryoconite, this work represents a step
200 toward increased analytical performances. The use of a customized Broad Energy HP-Ge detector in place of a commercial well HP-Ge detector and the analysis of massive cryoconite samples contributed to the improved results presented here. Considering our previous work as a benchmark (Baccolo et al., 2017), analytical sensitivity has been increased on average by a factor of 8.4, with several MDA less than 1 Bq kg^{-1} . With respect to other studies concerning the characterization of
205 sediments and soils, MDAs presented here (Supplementary Material) are one order of magnitude lower (Malain et al., 2012). This improvement can be attributed to the better shielding from background radioactivity, higher mass of the samples, increased energy resolution and detector efficiency. Thanks to the increased sensitivity, we have expanded the number of detected radionuclides, now including nuclides of the ^{235}U decay chain, ^7Be (cosmogenic), others belonging
210 to the ^{238}U chain and $^{108\text{m}}\text{Ag}$ (artificial). The identification of $^{108\text{m}}\text{Ag}$ is an important finding, since this artificial radioactive nuclide ($t_{1/2}$ 418 yr) has never been reported in terrestrial environments. It has only been occasionally observed in tissues of marine organisms where silver is bio-accumulated (Morita et al., 2010), in marine sediments collected at nuclear testing sites in the Pacific (Beasley and Held, 1971), and during the evaluation of material radiopurity for rare event physics
215 experiments (Laubenstein, 2017).

4.2. Radioactive content of cryoconite samples

4.2.1. Natural Radionuclides

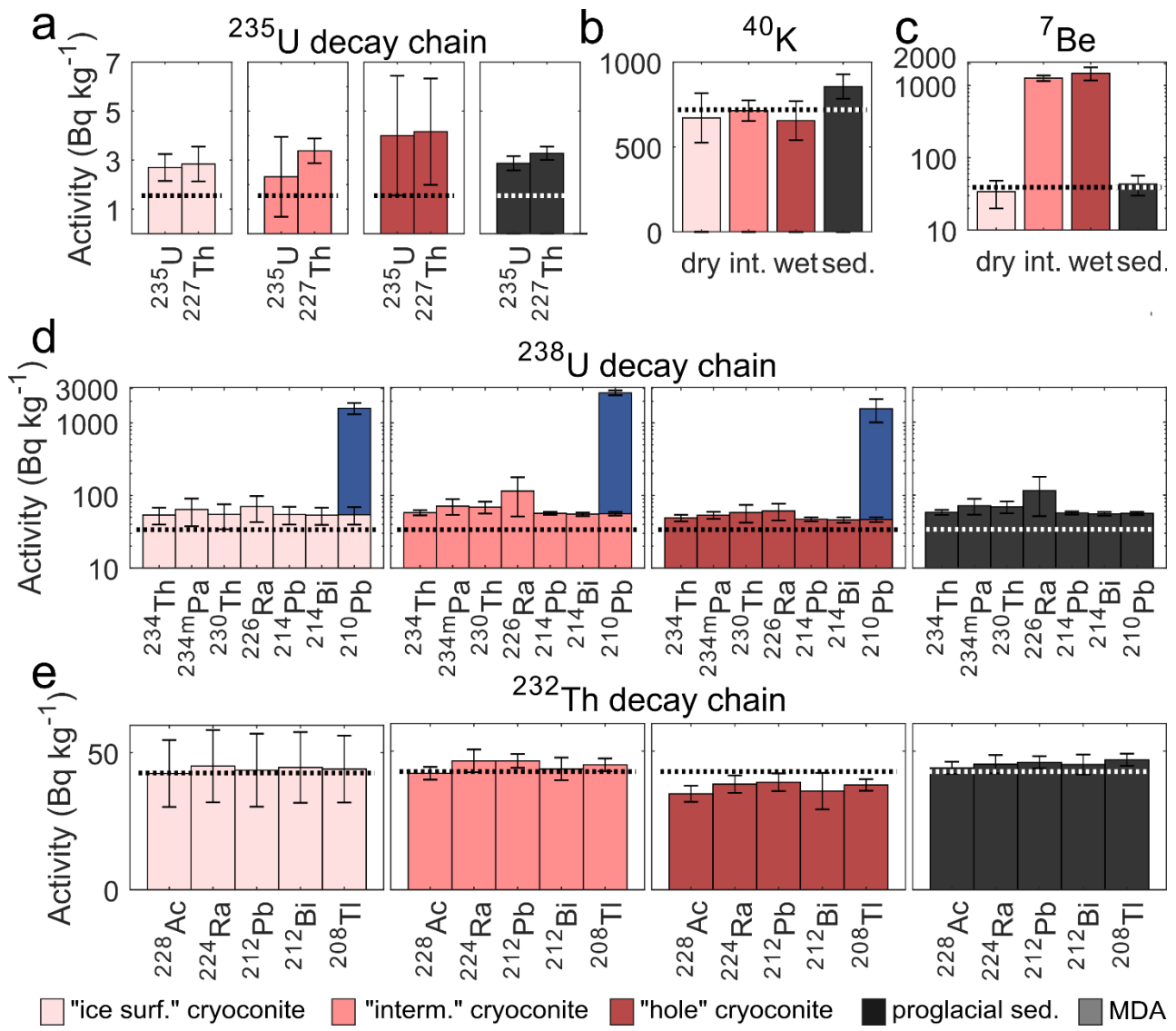
The activity of natural radionuclides detected in cryoconite samples is shown in Fig. 2. It can be seen that in dealing with lithogenic nuclides, the radioactivity is comparable to the values characterizing
220 the upper continental crust (UCC) reference, particularly for ^{40}K and ^{232}Th -related nuclides. For ^{238}U -progenies, an activity slightly higher than UCC is noted. The UCC reference for ^{235}U and ^{238}U is 1.55 and 33.8 Bq kg^{-1} respectively, while on average cryoconite samples present an activity of $55(13)\text{ Bq kg}^{-1}$ (standard deviation) for ^{238}U -chain nuclides (without considering ^{210}Pb) and of $3.3(1.5)\text{ Bq kg}^{-1}$ for ^{235}U . The enrichment is uniform among cryoconite samples from the Morteratsch glacier, and
225 no significant differences are noted when comparing cryoconite types considered here. By

comparing the activity of ^{235}U and $^{234\text{m}}\text{Pa}$, a reference for ^{238}U activity, it is possible to understand if the uranium radioactivity excess depends on a non-lithogenic uranium fraction. Natural uranium has a regular $^{238}\text{U}/^{235}\text{U}$ activity ratio of 21.7 (Salama et al., 2019); our samples display an average ratio of 21.8, in agreement with the natural value. The result rules out the presence in cryoconite of a uranium fraction whose isotopic composition has been artificially altered (depletion or enrichment for military and commercial purposes). In addition, proglacial sediments collected near the glacier show an enrichment of U- related nuclides, with an activity for the 235- and 238-chains of 3.1(0.3) and 62(9) Bq kg⁻¹ respectively, comparable to what is observed in cryoconite. This confirms that the enrichment is specific to the Morteratsch glacial basin. This observation is probably related to uraniferous minerals in the rocks surrounding the glacier, as has also suggested by studies about mineral waters (Stalder et al., 2012).

With respect to the lithogenic radionuclides, $^{210}\text{Pb}_{\text{exc.}}$ ($t_{1/2}$ 22.3 yr) and ^7Be ($t_{1/2}$ 53.1 d) have notable differences. Firstly, they are not directly derived from the radioactive decay of crustal material, since they are natural FRN, as $^{210}\text{Pb}_{\text{exc.}}$ (excess ^{210}Pb) is the decay product of ^{222}Rn escaped into the atmosphere from the Earth crust, and ^7Be is a cosmogenic nuclide produced in the high atmosphere. Their activity in cryoconite is above typical environmental values. In soils and sediments $^{210}\text{Pb}_{\text{exc.}}$ usually doesn't exceed tens or a few hundred Bq kg⁻¹ (Persson and Holm, 2011), while for ^7Be typical values in surficial samples are in the range of 10-50 Bq kg⁻¹ (Blake et al., 1999; Mabit et al., 2008). The mean activity in cryoconite from the Morteratsch glacier is 1,700(500) and 820(760) Bq kg⁻¹ respectively. Unlike uranium, in this case the anomaly concerns the cryoconite only, rather than being characteristic of the catchment since proglacial sediments have an activity compatible with ordinary environmental values: 43(13) Bq kg⁻¹ for ^7Be , while the activity of $^{210}\text{Pb}_{\text{exc.}}$ is below MDA (2 Bq kg⁻¹). In addition, significant differences are found between cryoconite types, in particular considering ^7Be . For this nuclide the average activity in "ice surface" cryoconite is 34(14) Bq kg⁻¹, in "intermediate" cryoconite it 1,261(111) Bq kg⁻¹ and in "hole" cryoconite is 1,473(301) Bq kg⁻¹. For $^{210}\text{Pb}_{\text{exc.}}$ the higher activity is observed in "intermediate" samples, with a value of 2,550(202) Bq kg⁻¹, while "ice surface" and "hole" samples show similar activities: 1,550 and 1,530 Bq kg⁻¹.

4.2.2. Anthropogenic Radionuclides

The results of this study confirm those of a previous work (Baccolo et al., 2017), demonstrating that cryoconite from the Morteratsch glacier is contaminated with artificial FRN. Results concerning analysis of artificial FRN are presented in Fig. 3 and Tab. 1, and the following radionuclides have been identified in our samples: $^{108\text{m}}\text{Ag}$, ^{137}Cs , ^{207}Bi and ^{241}Am . The identification of $^{108\text{m}}\text{Ag}$ ($t_{1/2}$ 418 yr) is particularly relevant, since this is the first finding of $^{108\text{m}}\text{Ag}$ in terrestrial environments. It was produced from nuclear reactions involving silver components during test explosions in the 1960s (Grismore et al., 1972). This radionuclide has been detected in three samples, one per cryoconite type. Activity concentrations of $^{108\text{m}}\text{Ag}$ in cryoconite are low, ranging from 0.26±0.10 to 0.77±0.37 Bq kg⁻¹ (MDA: 0.24 Bq kg⁻¹). It has not been observed in proglacial sediments. The identification of (Grismore et al., 1972). This radionuclide has been detected in three samples, one per cryoconite type. Activity concentrations of $^{108\text{m}}\text{Ag}$ in cryoconite are low, ranging from 0.26±0.10 to 0.77±0.37 Bq kg⁻¹ (MDA: 0.24 Bq kg⁻¹). It has not been observed in proglacial sediments. The identification of ^{207}Bi ($t_{1/2}$ 31.5 yr) in the environment is also rather rare, although it has been previously detected in some terrestrial environments (Bossey et al., 2006) and in cryoconite (Tieber et al., 2009; Baccolo et al., 2017, 2019). Its production in the Northern Hemisphere is associated



270 **Fig. 2** Natural radionuclides in cryoconite samples from the Morteratsch glacier. Data refer to “ice surface”, “intermediate” and “hole” cryoconite types, and to proglacial sediments (sed.) collected near the glacier terminus. The dashed line corresponds to average UCC activities, calculated from the reference by Rudnick and Gao (2003). The reference for ${}^7\text{Be}$ is taken from Blake et al. (1999) and Mabit et al. (2008). ${}^{210}\text{Pb}$ activity is represented for both supported (red shades) and excess fractions (blue).

275

with the explosion of the Tzar thermonuclear device in 1961 in Novaja Zemlja (Bossew et al., 2006). ${}^{207}\text{Bi}$ was found in six of the seven cryoconite samples presented in this study. On average its activity in “ice surface” cryoconite is 2.8(3.1) Bq kg^{-1} , in “intermediate” cryoconite is 1.4(0.3) Bq kg^{-1} and in “hole” cryoconite is 0.5(0.2) Bq kg^{-1} . In proglacial sediments its activity is lower than MDA, (0.3 Bq kg^{-1}). We found similar results for ${}^{241}\text{Am}$ ($t_{1/2}$ 432.2 yr), where the highest concentrations are found in “ice surface” cryoconite with an average activity of 17(20) Bq kg^{-1} . Lower activities characterize “intermediate” and “hole” samples for ${}^{241}\text{Am}$, with mean values of 8.7(0.6) and 1.5(1.4) Bq kg^{-1} respectively. In proglacial sediments the activity is lower than MDA (0.2 Bq kg^{-1}). Despite being considered a relatively rare artificial radionuclide, ${}^{241}\text{Am}$ has been reported several times in the environment though at lower concentrations in comparison to the results presented here (Shabana and Al-Shammari, 2001). Its occurrence is mostly related to nuclear test explosions and its

280

285

Tab. 1 Average activity data for the four sample types considered here: “ice surface”, “hole” and “intermediate” cryoconite, and proglacial sediments.

	analyte	“ice surf.” cryoconite (n=3)	“intermediate” cryoconite (n=1)	“hole” cryoconite (n=3)	proglacial sediments (n=1)
²³⁵U chain (Bq kg ⁻¹)	²³⁵ U	2.7±0.5	2±2	4±2	2.9±0.3
	²²⁷ Th	2.8±0.7	3.4±0.5	4±2	3.3±0.3
²³⁸U chain (Bq kg ⁻¹)	²³⁴ Th	54±14	58±5	49±5	54±4
	^{234m} Pa	64±27	54±6	71±18	70±16
	²³⁰ Th	55±21	69±13	58±16	58±11
	²²⁶ Ra	71±28	115±64	61±16	78±15
	²¹⁴ Pb	55±15	57±3	47±3	57±3
	²¹⁴ Bi	54±14	55±3	46±4	57±3
	²¹⁰ Pb _{tot}	1,604±285	2606±199	1577±557	1,684±132
	²¹⁰ Pb _{supp}	54±15	56±3	47±4	57±3
	²¹⁰ Pb _{exc}	1,550±271	2,550±202	1,530±557	< 2
²³²Th chain (Bq kg ⁻¹)	²²⁸ Ac	42±12	42±2	35±3	44±2
	²²⁴ Ra	45±13	47±4	38±3	45±3
	²¹² Pb	44±13	47±3	39±3	46±2
	²¹² Bi	45±13	44±4	36±7	45±4
	²⁰⁸ Tl	44±12	45±2	38±2	47±2
single nuclides (Bq kg ⁻¹)	⁷ Be	34±14	1,261±111	1,473±301	43±13
	⁴⁰ K	671±14	714±61	655±115	856±72
	^{108m} Ag	0.3±0.1	0.3±0.1	0.4±0.3	< 0.2
	¹³⁷ Cs	1,369±1,455	628±40	150±97	0.89±0.09
	²⁰⁷ Bi	3±3	0.5±0.2	1.4±0.3	< 0.3
	²⁴¹ Am	17±20	9±1	1±1	< 0.2

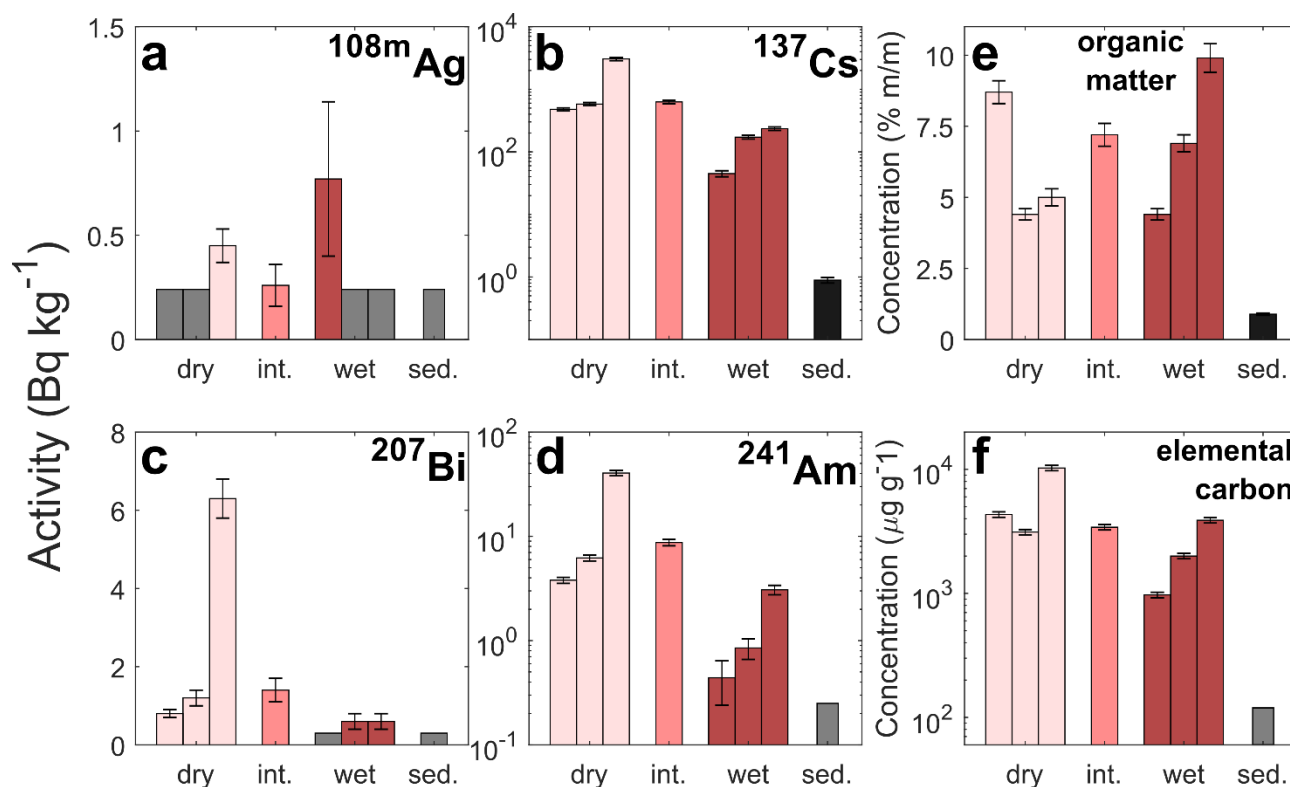
(continued)

	analyte	“ice surf.” cryoconite (n=3)	“intermediate” cryoconite (n=1)	“hole” cryoconite (n=3)	proglacial sediments (n=1)
carbonaceous content (% m/m)	organic carbon	3±1	3.6±0.2	4±1	0.45±0.02
	elemental carbon	0.6±0.4	0.34±0.02	0.2±0.1	< 0.01
	organic matter	6±2	7.2±0.4	8±2	0.90±0.04

290 environmental activity is increasing, owing the decay of its parent nuclide ^{241}Pu (Thakur and Ward, 2018). The distribution of ^{137}Cs in cryoconite resembles those of ^{207}Bi and ^{241}Am , with the highest activity being found in “ice surface” cryoconite, with a mean value exceeding $1,000 \text{ Bq kg}^{-1}$. In “intermediate” and “hole” samples the activity is lower: $628(40)$ and $150(97) \text{ Bq kg}^{-1}$ respectively.

4.3. Carbonaceous content

295 The variability of OM content in cryoconite samples is high, as is depicted in Fig. 3e, where it is evident that the concentration of OM is more variable within single cryoconite types than when comparing the different types with each other. The average values for each cryoconite type is similar, with a mean concentration (% mass fraction) in “ice surface” cryoconite of $3(1) \%$, in



300

Fig. 3 Concentration activity of artificial radionuclides detected in cryoconite from the Morteratsch glacier (a-d) and carbonaceous fractions (e-f). Data are grouped based on the three cryoconite types; red bars represent single samples, and grey bars MDA values.

305 “intermediate” and “hole” cryoconite of 3.6(0.2) and 4(1) % respectively. The OM fraction in proglacial sediments is limited, representing only 0.45(0.02) % of total mass. Results of analysis for EC (Fig. 3f) are similar to those concerning ^{137}Cs , ^{207}Bi and ^{241}Am , that is, the highest concentrations are found in “ice surface” cryoconite, with a mean mass fraction of 0.59(0.38) % and the lowest in “hole” cryoconite, with a mean value of 0.23(0.15) %. “Intermediate” cryoconite has a concentration of 0.34(0.02) %. In proglacial sediments the concentration of EC is lower than MDA (0.01 %).

310 5. Discussion

5.1. Correlation analysis

To explore the relationships between radionuclides and impurities analyzed in cryoconite, a combination of Pearson’s correlation coefficients and cluster analysis was applied (Fig. 4a). To obtain robust results, ^{235}U , $^{234\text{m}}\text{Pa}$, ^{226}Ra and $^{108\text{m}}\text{Ag}$ were not considered, owing the high relative errors associated with these variables (larger than 30%). Three clusters in the data are identified. The largest cluster includes the natural lithogenic radionuclides, highlighting that these nuclides correlate with each other, as they are associated with the mineral fraction of cryoconite. An example of the high correlation within this cluster is depicted in Fig. 4b, where the activity of ^{214}Pb (belonging to the ^{238}U -chain) is compared to ^{208}Tl (belonging to the ^{232}Th -chain). A second cluster is defined by artificial FRN and EC, i.e. the anthropogenic species detected in cryoconite. The correlation between the variables of this cluster is notable, with coefficients exceeding 0.9 (see also Fig. 4c). This suggests that both FRN and EC display an affinity for cryoconite and likely for its OM fraction. It is well-known that many radionuclides have an affinity for OM (Livens and Baxter, 1988; Chuang et al., 2015), which is abundant in cryoconite, while the relationship between OM and EC remains poorly investigated. In addition, the positive correlation coefficients characterizing this cluster are indicative of the common source of the anthropogenic species found in cryoconite, the atmosphere. In fact, the only possible source for both EC and anthropogenic radionuclides in glacial environments is the atmosphere, and specifically atmospheric deposition. The third cluster is defined by ^7Be , $^{210}\text{Pb}_{\text{exc}}$ and OM. The correlation coefficients within this cluster are lower and range from 0.5 to 0.7 (see also Fig. 4d), however the correlation between these variables confirms that OM in cryoconite plays an important role in the accumulation of atmospheric fallout and pollutants, as previously described for other contexts (Gadd, 1996; Accardi-Dey and Gschwend, 2002; Chuang et al., 2015).

Also visible in the correlation matrix is the negative correlation between ^{40}K and OM. This is possibly related to the fact that as cryoconite is composed of both mineral and organic fractions of different proportions, the higher is one of the two and the lower is the other one. In fact, organic matter is not only negatively correlated with ^{40}K , but with all the lithogenic radionuclides. Another observation from the matrix concerns ^7Be . This is negatively correlated with artificial species and with many of the lithogenic nuclides. The negative correlation with lithogenic nuclides is likely related to its positive correlation with OM, which is in turn negatively correlated with lithogenic nuclides. The negative correlation between ^7Be and the anthropogenic species is more difficult to interpret, since both share an atmospheric origin and are deposited on the glacier in association with precipitation. What sets them apart, however, is their residence time on glaciers. Artificial FRN considered here have half-lives of decades or more, and EC is a stable compound, particularly in the cold environments associated with glacial settings (Cheng et al., 2008). Conversely, ^7Be has a short half-life (53 d) and its presence within cryoconite is likely controlled by different processes.

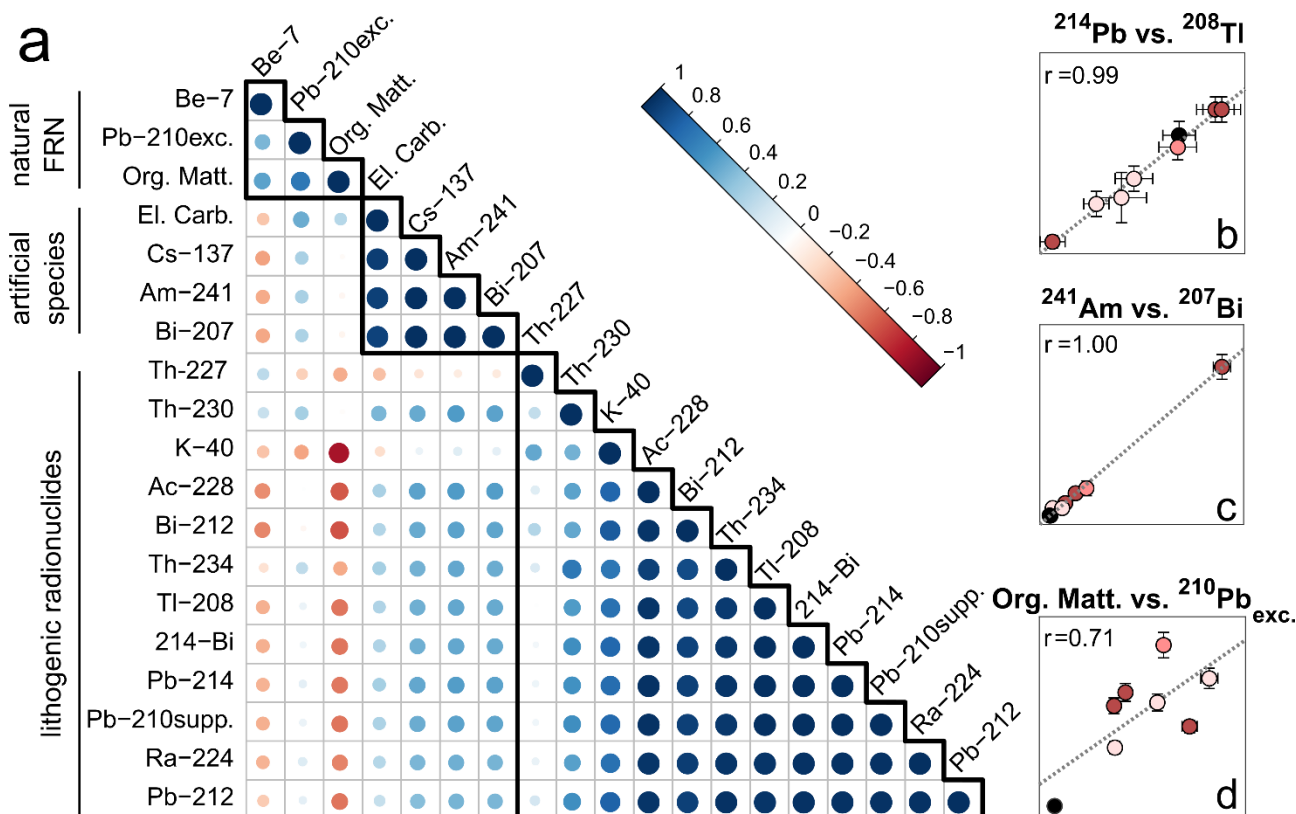


Fig. 4 Correlation among the variables considered in this study. In panel a the correlation is illustrated using a Pearson's correlation matrix where correlation coefficients are represented by circles whose color and size depend on their value. Variables have been ordered and clustered into three groups, following a hierarchical clustering approach. Clusters are defined by black solid lines. Panel b-d: linear correlation between selected variables belonging to the three clusters; the same color palette of Fig. 2 has been used.

350

5.2. Radioactivity content in different cryoconite types

355

The cryoconite types explored in this study show differences in terms of radioactivity and carbonaceous content (Fig. 2, Fig. 4 and Tab. 1). In "ice surface" cryoconite artificial FRN are abundant, while "hole" samples are rich in ^7Be . To investigate if such differences are relevant, a Student's t test (significant at 5%) was applied. Considering the "ice surface-intermediate", "ice surface-hole" and "intermediate-hole" pairs, we have selected only the variables showing a statistically significant difference with respect to two or three pairs. $^{210}\text{Pb}_{\text{exc.}}$, ^{137}Cs , ^7Be and OM satisfy this condition.

360

To understand which variables contribute most to variance between different cryoconite types, multiple linear regression has been applied to model the relationship between the variables and the cryoconite type ("ice surface", "intermediate" or "hole"). The complete model taking into account all four variables, has relatively good performances (R-squared = 0.73, Supplementary Material). To determine which of the variables contributes most, four additional models have been calculated in which single variables have been excluded one at a time (Supplementary Material).

365

The variable whose absence determines the largest decrease in classification performance is ^7Be , which is responsible for more than 85% of the variance explained in the complete model. In fact, the model calculated excluding ^7Be has an R-squared value lower than 0.1. ^7Be is the most discriminating variable being abundant in "hole" cryoconite, scarce in "ice surface" samples and showing intermediate activity concentrations in "intermediate" samples (Fig.

370

2c). The only other variable which contributes substantially to the model is OM, which accounts for 8 % of the variance explained in the complete model. The variability of OM in cryoconite resembles that of ^7Be : higher concentrations in “hole” samples, lower ones in “ice surface” samples.

375 5.3. Understanding cryoconite radioactivity in relation to the supraglacial environment

It has been demonstrated that the composition of cryoconite from the Morteratsch glacier is far from uniform. To begin to understand the processes involved in creating these differences, the features characterizing cryoconite types must be considered. “Hole” cryoconite deposits display
380 small defined granules (1-2 mm) and are always in contact with meltwater (see the Supplementary Material). In fact, cryoconite sediment characterizing “hole” deposits is found at the bottom of small cavities deepened into ice and filled with meltwater which is continuously supplied by active meltwater channels. On the contrary, “ice surface” cryoconite is accumulated in thick and dry deposits not in contact with meltwater, being usually found in areas of the glacier rich in ice cracks and crevasses which locally drain surficial channels. In addition, “ice surface” cryoconite doesn’t
385 present granules, being the sediment more massive and not showing aggregation patterns.

The concentration of ^7Be in cryoconite, which ranges from 34 Bq kg^{-1} in “ice surface” cryoconite to $1,473 \text{ Bq kg}^{-1}$ in “hole” cryoconite, can be explained considering the different degree of connection with supraglacial hydrology. On glaciers, the sources of ^7Be are snow and rain, where it accumulates through scavenging in association with aerosol (Ioannidou et al., 2005). Due to its relatively short
390 half-life (53 d), this radionuclide is typically found in recently deposited and shallow snow, and is less likely to be present in older, deeper snow and ice. The process most likely responsible for rapid mobilization of ^7Be on the surface of glaciers is supraglacial runoff induced by the melt of recent snow (Smith et al., 2000). “Hole” cryoconite is in contact with active supraglacial channels, it therefore interacts with meltwater and accumulates ^7Be . The presence of OM and extra-cellular polymeric substances, which are both affine for radionuclides (Gadd, 1996; Chuang et al., 2015),
395 likely favors the accumulation.

The activity of ^7Be in “ice surface” samples is similar to that which was measured in proglacial sediments (34 vs. 43 Bq kg^{-1}). These values are comparable with those observed in soils and
400 sediments where ^7Be is present because of direct atmospheric deposition, without the involvement of accumulation processes (Blake et al., 1999; Mabit et al., 2008). The scarcity of ^7Be in “ice surface” cryoconite suggests a limited interaction between these deposits and surficial meltwater in the weeks preceding sampling. This agrees with the poor connection with supraglacial hydrology characterizing these samples. But while ^7Be is scarce in “ice surface” cryoconite, the opposite is true
405 for artificial FRN and EC, which are more abundant in “ice surface” samples than in “hole” ones (Fig. 3). Considering that the only source of artificial FRN and EC on the surface of glaciers is meltwater (Baccolo et al., 2019), their abundance in “ice surface” cryoconite points to a prolonged interaction time with meltwater, but not in the season when our samples were collected, as revealed by the scarcity of ^7Be , rather in the past ones. Contrastingly, for “hole” samples the total interaction time with meltwater has likely been shorter (relatively low artificial FRN content), but more recent (high
410 ^7Be activity), suggesting that they have been in contact with meltwater for a longer time in the present melt season compared with “ice surface” samples. “Ice surface” samples described in this study are thus interpreted as samples rich in mature cryoconite which has been accumulated at the

415 surface of the Morteratsch glacier in previous ablation seasons. Conversely, relatively recent cryoconite, which was still forming when samples were collected, prevails in “hole” samples.

The aggregation state of cryoconite and its OM content agree with the interpretation. “Hole” samples display millimetric aggregates, while “ice surface” samples show no clear aggregation patterns (see Fig. 1 and Supplementary Material). Formation of cryoconite granules has been previously attributed to the interaction between mineral sediments and microbes. Clear aggregation in cryoconite relates to high microbiological activity, while poor or absent aggregation is indicative of limited microbial activity (Takeuchi et al., 2010; Langford et al., 2010). The lack of aggregation in “ice surface” samples and their low OM content, suggest a limited microbiological activity, in accordance with the poor hydrological connection and paucity of liquid water, essential for enabling microbes to flourish on glaciers (Hodson et al., 2010; Langford et al., 2010). “Hole” samples are well connected with meltwater channels and represent an ideal environment for microorganisms, as confirmed by the presence of aggregates and higher OM concentrations. Another factor supporting the interpretation is the variable mass of the cryoconite deposits sampled in this study. “Ice surface” samples are characterized by a notable accumulation of sediments, “hole” ones are more scarce (see photographs in the Supplementary Material). This difference can be related to the degree of maturity of the deposits as well. “Ice surface” deposits, partially consisting in cryoconite formed in previous melt seasons, have had a longer time to develop and accumulate mass. “Hole” deposits are more recent and were still actively forming during field campaign for this study. The sample identified as “intermediate” displays intermediate features between “ice surface” and “hole” cryoconite types. It has been sampled from a deposit that, despite presenting the typical “hole” features (aggregation and connection with meltwater), was particularly abundant, which is more characteristic of “ice surface” deposits. From a radiological point of view, “intermediate” cryoconite is rich in ^7Be , artificial FRN and EC, suggesting both recent and past interactions with meltwater. These features could be explained as follows: the “intermediate” sample consists in a deposit where cryoconite from past seasons has been reactivated in response to a renewed connection with meltwater during the 2018 ablation season.

6. Conclusions

Analysis of cryoconite from the Morteratsch glacier further supports previous results suggesting that cryoconite has an extraordinary ability to accumulate radionuclides. $^{108\text{m}}\text{Ag}$ has been detected in three cryoconite samples within this study, representing the first time this artificial radionuclide has been described within terrestrial environments. This result suggests that cryoconite should be considered for future studies concerning the occurrence of rare and poorly investigated radionuclides. But despite the notable radioactive content, the amount and type of radioactivity present in cryoconite collected on Morteratsch glacier are not uniform. Cryoconite from deposits that are in contact with meltwater channels and are characterized by well-defined granules, are extremely rich in short-lived ^7Be ($t_{1/2}$ 53.1 d). Deposits that are poorly connected with the supraglacial hydrological system, and that are not characterized by aggregation, are rich in long-lived artificial fallout radionuclides ($t_{1/2} > 30$ yr) and elemental carbon. The different half-lives of radionuclides have allowed to discuss these differences in relation to the dynamics characterizing the supraglacial hydrological system and to the degree of maturity of cryoconite. A high concentration of ^7Be in cryoconite is indicative of a recent interaction with meltwater, while the progressive accumulation of artificial radionuclides can be referred to a prolonged interaction with

meltwater. Focusing on these radiological features, it has been possible to distinguish deposits rich in cryoconite that formed in the previous ablation seasons, from deposits that were likely undergoing active formation while sampling was conducted.

460 Despite being preliminary and involving a limited number of samples, this study shows that radionuclides, in particular ^7Be , can be used as tracers to gather information on the maturity degree of cryoconite and on its age and explore supraglacial dynamics. Further studies are needed to investigate the relationships between the accumulation of radioactivity, organic matter and biological activity of cryoconite. To this aim, it would be desirable to consider different
465 granulometric and compositional fractions of cryoconite and also other environmental samples from the surface of glaciers, such as ice and meltwater.

Acknowledgments

This study has been supported by the Project of Strategic Interest NextData, funded by the Italian National Research Program PNR 2011-2013, and by the project “Impatto degli aerosol e delle polveri
470 fini provenienti dall’area Mediterranea e dal Nord Africa sulla criosfera Alpina”, funded by “Dipartimento per gli affari regionali e le autonomie della Presidenza del Consiglio dei Ministri”.

References

- Accardi-Dey, A.M., Gschwend, P.M. 2002. Assessing the combined roles of natural organic matter and black carbon as sorbents in sediments. *Environ. Sci. Technol* 36, 21-29.
- 475 Appleby, P.G. 2008. Three decades of dating recent sediments by fallout radionuclides: a review. *Holocene* 18, DOI: 10.1177/0959683607085598.
- Baccolo, G., Di Mauro, B., Massabò, D., Clemenza, M., Nastasi, M., Delmonte, B., Prata, M., Prati, P., Previtali, E., Maggi, V. 2017. Cryoconite as a temporary sink for anthropogenic species stored in glaciers. *Sci. Rep.* 7, DOI: 10.1038/s41598-017-10220-5.
- 480 Baccolo, G., Łokas, E., Gaca, P., Massabò, D., Ambrosini, R., Azzoni, R.S., Clason, C., Di Mauro, B., Franzetti, A., Nastasi, M., Prata, M., Prati, P., Previtali, E., Delmonte, B., Maggi, V. 2019. Cryoconite: an efficient accumulator of radioactive fallout in glacial environments. *Cryosphere Discussion*, in discussion.
- Bagshaw, E.A., Tranter, M., Fountain, A.G., Welch, K.A., Basagic, H., Berry Lyons, B. 2007. Biogeochemical evolution of cryoconite holes on Canada Glacier, Taylor Valley, Antarctica. *Biogeosciences* 112, DOI: 10.1029/2007JG000442.
- 485 Beasley, T.M., Held, E.E. 1971. Silver-108m in biota and sediments at Bikini and Eniwetok atolls. *Nature* 230, 450-451.
- Birch, M.E., Cary, R.A. 1996. Elemental carbon-based method for monitoring occupational exposures to particulate diesel exhaust. *Aerosol Sci. Tech.* 25, 221-241.
- Blake, W.H., Walling, D.E., He, Q. 1999. Fallout beryllium-7 as a tracer in soil erosion investigation. *Appl. Rad. Isotopes* 51, 599-605.
- 490 Bogdal, C., Schmid, P., Zennegg, M., Anslometti, F.S., Scheringer, M., Hungerbühler, K. 2009. Blast from the Past: Melting Glaciers as a Relevant Source for Persistent Organic Pollutants. *Environ. Sci. Technol.* 43, 8173-8177.
- Bossew, P., Lettner, H., Hubmer, A. 2006. A note on ^{207}Bi in environmental samples. *J. Environ. Radioactiv.* 91, 160-166.
- Cheng, C.H., Lehmann, J., Thies, J.E., Burton, S.D. 2008. Stability of black carbon in soils across a climatic gradient. *J. Geophys. Res. Biogeo.*, 113, DOI: 10.1029/2007JG000642.

- 495 Chuang, C.Y., Santschi, P.H., Wen, L.S., Guo, L., Xu, C., Zhang, S., Jiang, Y., Ho, Y.F., Schwehr, K.A., Quigg, A., Hung, C.C., Ayrarov, M., Schumann, D. 2015. Binding of Th, Pa, Pb, Po and Be radionuclides to marine colloidal macromolecular organic matter. *Mar. Chem.* 173, 320-329.
- Clemenza, M., Cucciati, G., Maggi, V., Pattavina, L., Previtali, E. 2012. Radioactive fallouts as temporal markers for glaciers ice cores dating. *Eur. Phys. J. Plus* 127, 1-8.
- 500 Cook, J.M., Hodson, A.J., Irvine-Fynn, T.D.L. 2016. Supraglacial weathering crust dynamics inferred from cryoconite hole hydrology. *Hydrol. Process.* 30, DOI: 10.1002/hyp.10602.
- Crozaz, G., Picciotto, E., De Breuk, W. 1964. Antarctic snow chronology with Pb210. *J. Geophys. Res.* 69, 2597-2604.
- Di Mauro, B., Baccolo, G., Garzonio, R., Giardino, C., Massabò, D., Piazzalunga, A., Rossini, M., Colombo, R. 2017. Impact of impurities and cryoconite on the optical properties of the Morteratsch glacier (Swiss Alps). *Cryosphere* 11, DOI: 10.5194/tc-11-2393-2017.
- 505 Eichler, A., Schwikowski, M., Gäggeler, H.W., Furrer, V., Synal, H.A., Beer, J., Saurer, M., Funk, M. 2000. Glaciochemical dating of an ice core from upper Grenzgletscher (4200 m a.s.l.). *J. Glaciol.* 46, 507-515.
- Ferrario, C., Pittino, F., Tagliaferri, I., Gandolfi, I., Bestetti, G., Azzoni, R.S., Diolaiuti, G., Franzetti, A., Ambrosini, A., Villa, S. 2017. Bacteria contribute to pesticide degradation in cryoconite holes in an Alpine glacier. *Environ. Pollut.* 230, 919-926.
- 510 Gadd, G.M., 1996. Influence of microorganisms on the environmental fate of radionuclides. *Endeavour* 20, 150-156.
- Gerringa, L.J.A., Alderkamp, A.C., Laan, P., Thuróczy, C.E., De Baar, H.J.W., Mills, M.M., van Dijken, G.L., van Haren, H., Arrigo, K.R. 2012. Iron from melting glaciers fuels the phytoplankton blooms in Amundsen Sea (Southern Ocean): Iron biogeochemistry. *Deep-Sea Res. Pt. II* 71-76, 16-31.
- 515 Grismore, R., Folsom, T.R., Hodge, V.F., Young, D.R. 1972. A study of the radiosilver signature of the 1961-1962 nuclear weapons testing period. *T. New York Acad. Sci.* 34, 392-415.
- Hodson, A., Cameron, K., Bøggild, C., Irvine-Fynn, T., Langford, H., Pearce D., Banwart, S. 2010. The structure, biological activity and biogeochemistry of cryoconite aggregates upon an Arctic valley glacier: Longyearbreen, Svalbard. *J. Glaciol.* 56, 349-362.
- 520 Huang, J., Kang, S., Ma, M., Guo, J., Cong, Z., Dong, Z., Yin, R., Xu, J., Tripathee, L., Ram, K., Wang, F. 2019. Accumulation of Atmospheric Mercury in Glacier Cryoconite over Western China. *Environ. Sci. Technol.* 53, 6632-6639.
- Ioannidou, A., Manolopoulou, M., Papastefanou, C. 2005. Temporal changes of ⁷Be and ²¹⁰Pb concentrations in surface air at temperate latitudes (40°N). *Appl. Rad. Isotopes* 63, 277-284.
- Langford, H., Hodson, A., Banwart, S., Bøggild, C. 2010. The microstructure and biogeochemistry of Arctic cryoconite granules. *Ann. Glaciol.* 51, 87-94.
- 525 Laubenstein, M. 2017. Screening of materials with high purity germanium detectors at the Laboratori Nazionali del Gran Sasso. *Int. J. Mod. Phys. A* 32, DOI: 0.1142/S0217751X17430023.
- Li, Q., Kang, S., Wang, N., Li, Y., Li, X., Dong, Z., Chen, P. 2017. Composition and sources of polycyclic aromatic hydrocarbons in cryoconites of the Tibetan Plateau glaciers. *Sci. Tot. Environ.* 574, 991-999.
- 530 Livens, F.R., Baxter, M.S. 1988. Chemical association of artificial radionuclides in Cumbrian soils. *J. Environ. Radioactiv.* 7, 75-86.
- Li, Y., Kang, S., Yan, F., Chen, J., Wang, K., Paudyal, R., Liu, J., Qin, X., Sillanpää, M. 2019. Cryoconite on a glacier on the north-eastern Tibetan plateau: light-absorbing impurities, albedo and enhanced melting. *J. Glaciol.* 65, 633-644.
- Łokas, E., Barmiński, P., Wachniew, P., Mietelski, J.W., Kawiak, T., Środoń, J. 2014. Sources and pathways of artificial radionuclides to soils at a High Arctic site. *Environ. Sci. Pollut. R.* 21, 12479-12493.
- 535

- 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.
- Łokas, E., Wachniew, P., Jodłowski, P. & Gasiorek, M. 2017. Airborne radionuclides in the proglacial environment as indicators of sources and transfers of soil material. *J. Environ. Radioactiv.* 178-179, 193-202.
- 540 Łokas, E., Zaborska, A., Sobota, I., Gaca, P., Milton, J.A., Kocurek, P., Cwanek, A. 2019. Airborne radionuclides and heavy metals in High Arctic terrestrial environment as the indicators of sources and transfers of contamination. *Cryosphere* 13, DOI: 10.5194/tc-13-2075-2019.
- Mabit, L., Benmansour, M., Walling, D.E., 2008. Comparative advantages and limitations of the fallout radionuclides ^{137}Cs , $^{210}\text{Pb}_{\text{ex}}$ and ^7Be for assessing soil erosion and sedimentation. *J. Environ. Radioactiv.* 99, 1799-1807.
- 545 Malain, D. Regan, P.H., Bradley, D.A., Matthews, M., Al-Sulaiti, H.A., Santawamaitre, T. 2012. An evaluation of the natural radioactivity in Andaman beach sand samples of Thailand after the 2004 tsunami. *Appl. Radiat. Isotopes* 70, 1467-1474.
- Morita, T., Ohtsuka, Y., Fujimoto, K., Minamisako, Y., Iida, R., Nakamura, M., Kayama, T. 2010. Concentrations of ^{137}Cs , ^{90}Sr , $^{108\text{m}}\text{Ag}$, $^{239+240}\text{Pu}$ and atom ratio of $^{240}\text{Pu}/^{239}\text{Pu}$ in tanner crabs, *Chionoecetes japonicus* and *Chionoecetes opilio* collected around Japan. *Mar. Pollut. Bull.* 60, 2311-2322.
- 550 Nagatsuka, N., Takeuchi, N., Nakano, T., Kokado, E., Li, Z. 2010. Sr, Nd and Pb stable isotopes of surface dust on Ürümqi glacier No. 1 in western China. *Ann. Glaciol.* 51, 95-105.
- Oerlemans, J., Gleisen, R.H., Van Der Broecke, M.R., 2009. Retreating alpine glaciers: increased melt rates due to accumulation of dust (Vadret da Morteratsch, Switzerland). *J. Glaciol.* 55, 729-736.
- 555 Owens, P.N., Blake, W.H., Millward, G.E., 2019. Extreme levels of fallout radionuclides and other contaminants in glacial sediment (cryoconite) and implications for downstream aquatic ecosystems. *Sci. Rep.* 9, DOI: 10.1038/s41598.
- Persson, B.R.R., Holm, E. 2011. Polonium-210 and lead-210 in the terrestrial environment: a historical review. *J. Environ. Radioactiv.* 102, 420-429.
- Picciotto, E., Wilgain, S. 1963. Fission products in Antarctic snow, A reference level for measuring accumulation. *J. Geophys. Res.* 68, 5965-5972.
- 560 Pinglot, J.F., Pourchet, M., Lefauconnier, B., Hagen, J.O., Vaikmäe, R., Punning, J.M., Watanabe, O., Takahashi, S., Kameda, T. 1994. Natural and artificial radioactivity in the Svalbard glaciers. *J. Environ. Radioactiv.* 25, 161-176.
- Pittino, F., Maglio, M., Gandolfi, I., Azzoni, R.S., Diolaiuti, G., Ambrosini, R., Franzetti, A. 2018. Bacterial communities of cryoconite holes of a temperate alpine glacier show both seasonal trends and year-to-year variability. *Ann. Glaciol.* 59 DOI: 10.1017/aog.2018.16.
- 565 Preunkert, S., Legrand, M. 2013. Towards a quasi-complete reconstruction of past atmospheric aerosol load and composition (organic and inorganic) over Europe since 1920 inferred from Alpine ice cores. *Clim. Past* 9, 1403-1416.
- Pribyl, D.W. 2010. A critical review of the conventional SOC to SOM conversion factor. *Geoderma* 156, 75-83.
- Rippin, D.M., Pomfret, A., King, N. 2015. High resolution mapping of supra-glacial drainage pathways reveals link between micro-channel drainage density, surface roughness and surface reflectance. *Earth Surf. Proc. Land* 40, DOI: 10.1002/esp.3719.
- 570 Rizzi, C., Finizio, A., Maggi, V., Villa, S. 2019. Spatial-temporal analysis and risk characterization of pesticides in Alpine glacial streams. *Environ. Poll.* 248, 659-666.
- Rossini, M., Di Mauro, B., Garzonio, R., Baccolo, G., Cavallini, G., Mattavelli, M., De Amicis, M., Colombo, R. 2018. Rapid melting dynamics of an alpine glacier with repeated UAV photogrammetry. *Geomorphology* 304, 159-172.
- 575 Salama, E., El-kameesy, S.U., Elrawi, R. 2019. Depleted uranium assessment and natural radioactivity monitoring in North West of Iraq over a decade since the last Gulf War. *J. Environ. Radioactiv.* 201, 25-31.

- Shabana, E.I., Al-Shammari, H.L. 2001. Assessment of the global fallout of plutonium isotopes and americium-241 in the soil of the central region of Saudi Arabia. *J. Environ. Radioactiv.* 57, 67-74.
- 580 Smith, A.M., Fink, D., Child, D., Levchenko, V.A., Morgan, V.I., Curran, M., Etheridge, D.M., Elliott, G. 2000. ^7Be and ^{10}Be concentrations in recent firn and ice at Law Dome, Antarctica. *Nucl. Instrum. Meth. B* 172, 847-855.
- Stalder, E., Blanc, A., Haldimann, M., Dudler, V. 2012. Occurrence of uranium in Swiss drinking water. *Chemosphere* 86, 672-679.
- 585 Takeuchi, N. 2002. Optical characteristics of cryoconite (surface dust) on glaciers: the relationship between light absorbency and the property of organic matter contained in the cryoconite. *Ann. Glaciol.* 34, 409-414.
- Takeuchi, N., Nishiyama, H., Li, Z. 2010. Structure and formation process of cryoconite granules on Urumqi glacier No. 1, Tien Shan, China. *Ann. Glaciol.* 51, 9-14.
- 590 Takeuchi, N., Sakaki, R., Uetake, J., Nagatsuka, N., Shimada, R., Niwano, M., Aoki, T. 2018. Temporal variations of cryoconite holes and cryoconite coverage on the ablation ice surface of Qaanaaq Glacier in northwest Greenland. *Ann. Glaciol.* 59, DOI: 10.1017/aog.2018.19.
- Thakur, P., Ward, A.L., 2018. ^{241}Pu in the environment: insight into the understudied isotope of plutonium. *J. Radioanal. Nucl. Chem.* 317, 757-778.
- Tieber, A., Lettner, H., Bossew, P., Hubmer, A., Sattler, B., Hofmann, W. 2009. Accumulation of anthropogenic radionuclides in cryoconites on Alpine glaciers. *J. Environ. Radioactiv.* 100, 590-598.
- 595 Weiland-Bräuer, N., Fischer, M.A., Schramm, K.W., Schmitz, R.A. 2017. Polychlorinated Biphenyl (PCB)-Degrading Potential of Microbes Present in a Cryoconite of Jamtalferner Glacier. *Front. Microbiol.* 8, DOI: 10.3389/fmicb.2017.01105.
- 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, 370-379.