Referee #2

R2: General comments. This paper is an interesting summary of an emerging research area, that of cryoconite as a record of fallout radionuclides and a potential concentrator of impurities. I cannot comment on the nuclide analysis methods, but they seem sound and reference other publications, so I have confidence in the research team to conduct these analyses appropriately. The paper is generally well written and presents some interesting results. I particularly liked the dating hypothesis discussion, and was gratified that the authors acknowledge that this is an area that needs more work, rather than trying to sew up everything in this one paper. I found the carbon discussion a little distracting and would recommend removing this section since it didn't really contribute to the main story.

Reply: Thank you very much for the positive comment. Regarding the section dedicated to carbonaceous matter, we would prefer to maintain it. We agree with the reviewer that in the first draft the section have sounded a little bit disconnected from the other sections of the manuscript. Reviewer 1 has asked for a deeper discussion about the relationships between cryoconite radioactivity and organic matter, also in relation to the differences observed between the two glaciers. We have expanded these sections and for this reason we would like to maintain it.

R2: The figures were sometimes a little confusing, with too much colour and too much information presented simultaneously. I make some suggestions for improvement below, but would certainly recommend testing for colour-blind readers as a minimum, and improving/simplifying the labelling and shortening the captions.

Reply: we have modified many of the figures, enhancing the color contrast, using markers in place of colors and trying to simplify them.

R2: I would also suggest that the abstract is rewritten to better reflect the key findings of the paper (which I understand as): that cryoconite is an important concentrator of FRNs; that FRNs in different Alpine Glaciers are similar to each other; that Alpine glaciers are similar to other glaciers but show important differences with respect to proximity to some sources; and that FRNs could be a way of dating cryoconite, since they accumulate over time (in contrast to previous suggestions). As written now, I didn't think it represented the key findings of the paper. The distinction between local and global sources is also confusing, since most cryoconite research considers 'local' to be within catchment (when defining, for example, debris sources or microbial seeding grounds). Instead, perhaps be specific that Chernobyl impacted the Alpine Glaciers but not so much the Svalbard one. The processes description in the abstract is particularly weak and I didn't think very relevant. Use the words for your dating hypothesis instead.

Reply: We have modified the abstract considering the suggestions from the reviewer. Here the new version:

"Cryoconite is rich in natural and artificial radioactivity, but a discussion about its ability to accumulate radionuclides is lacking. A characterization of cryoconite from two Alpine glaciers is here presented. Results confirm that cryoconite is significantly more radioactive than the matrices usually adopted for the environmental monitoring of radioactivity, as lichens and mosses, with activity concentrations exceeding 10,000 Bq kg^{-1} for single radionuclides. This makes cryoconite an ideal matrix to investigate the deposition and occurrence of radioactive species in glacial environments. In addition, cryoconite can be used to track environmental radioactivity sources. We have exploited atomic and activity ratios of artificial radionuclides to identify the sources of the anthropogenic radioactivity accumulated in our samples. The signature of cryoconite from different Alpine glaciers is similar and compatible with the stratospheric global fallout and Chernobyl accident products. Differences are found when considering other geographic contexts. A comparison with data from literature shows that Alpine cryoconite is strongly influenced by the Chernobyl fallout, while cryoconite from other regions is more impacted by events such as nuclear test explosions and satellite re-entries. To explain the

accumulation of radionuclides in cryoconite, the glacial environment as a whole must be considered, and particularly the interaction between ice, meltwater, cryoconite and atmospheric deposition. We hypothesize that the impurities originally preserved into ice and mobilized with meltwater during summer, including radionuclides, are accumulated in cryoconite because of their affinity for organic matter, which is abundant in cryoconite. In relation to these processes, we have explored the possibility to exploit radioactivity to date cryoconite."

R2: L20: 'extremely rich' is too subjective

Reply: we have now removed "extremely".

R2: L23: 'among the most radioactive environmental matrices' is rather vague – can you be specific?

Reply: we have now changed to *"...cryoconite is significantly more radioactive than the matrices usually adopted for the environmental monitoring of radioactivity, as lichens and mosses…"*

R2: L27: can you elaborate here? What specific aspects of their interaction?

Reply: we have now added the following passage: "*We hypothesize that the impurities originally preserved into ice and mobilized with meltwater during summer, including radionuclides, are accumulated in cryoconite because of their affinity for organic matter, which is abundant in cryoconite, and in particular for extra-cellular polymeric substances."*

R2: P2 L33: 'the latter of these' instead 'of which'

Reply: we have replaced with "cryoconite".

R2: Suggest combining the first two paragraphs, they are very short.

Reply: we agree with the reviewer, done.

R2: L45: 'incoherent' is awkward, suggest replacing with 'unconsolidated'

Reply: changed accordingly.

R2: L46: I would dispute that cryoconite requires abundant meltwater to form - it is found on ice surfaces in Antarctica with extremely limited quantities of meltwater

Reply: we agree, we have now removed "abundant"

R2: L50: please include a reference on the role of cyanobacteria

Reply: we have now included Langford et al., 2010.

R2: L53: I think this is specific to cryoconite granules – cryoconite may be present without forming granules (eg. Antarctica). I would suggest adding 'granules' to the end of this sentence.

Reply: done.

R2: L59: could you include some example references or a review paper here?

Reply: not to increase too much the number of references, we have added a quote to suited papers that had already been cited in the paper.

R2: Figure 1: please indicate the scale on A and B, or state the approx. hole diameter in the text

Reply: we have now included the scale in the two panels.

R2: P4 L104: could you include some example references or a review paper here?

Reply: not to increase too much the number of references, we have added a quote to suited papers that had already been cited in the paper.

R2: L106: can you tell us when it detached, rather than 'few years'?

Reply: of course, it was 2015.

R2: L114: tell us why this is favourable for the formation of cryoconite (simply put: because there is more source material)

Reply: we have added the passage.

R2: L118: define 'clean' – how were they cleaned? Deionised water? Ethanol? Between samples? In what vessels were the samples stored, and how were they treated?

Reply: we have added the details. We used sterile disposable pipettes, sterile plastic tubes and spoons cleaned with ethanol between samples.

R2: L128: How were the sampling sites chosen, and how widespread were they?

Reply: they were extremely common, on average the distance between two adjacent deposits or holes was of few meters. We have chosen the ones where cryoconite was more abundant, we have now added this detail to the text: *"We selected the most abundant cryoconite deposits, so as to have material available for other analyses also: twelve samples have been gathered on the Morteratsch Glacier (between 2100 and 2300 m a.s.l.) and ten on the Forni Glacier (between 2600 and 2800 m a.s.l.), each one consisting in 10-40 g of wet cryoconite."*

R2: L131: this is the assumption of all papers. Instead of saying that the material are not published, I would suggest rephrasing to say that accompanying gamma spectroscopy data can be found in the 2017 publication.

Reply: we agree, we have rephrased accordingly: *"Part of the data concerning gamma spectrometry applied to cryoconite from the Morteratsch samples has already been published (Baccolo et al., 2017)."*

R2: L172: Is the equation and description of Pearson Correlation necessary? I think the reference is sufficient, but leave this at the author's discretion.

Reply: we have shortened the passage where we described the link between r and d, however we would to prefer to maintain the equation since MDS is not usually applied to correlation but to distance.

R2: Figure 3: Can the lines be labelled on the plot rather than in the very long caption? For example, the yellow (continental crust), black (average (mean?!)) and dashed (st dev) could be labelled instead, reducing the overlong caption. I would also check the colours for use by colour-blind readers – perhaps patterns could be used instead?

Reply: thanks for the suggestion, we have modified the couple figure-caption as it follows, adding a legend and enhancing color contrast:

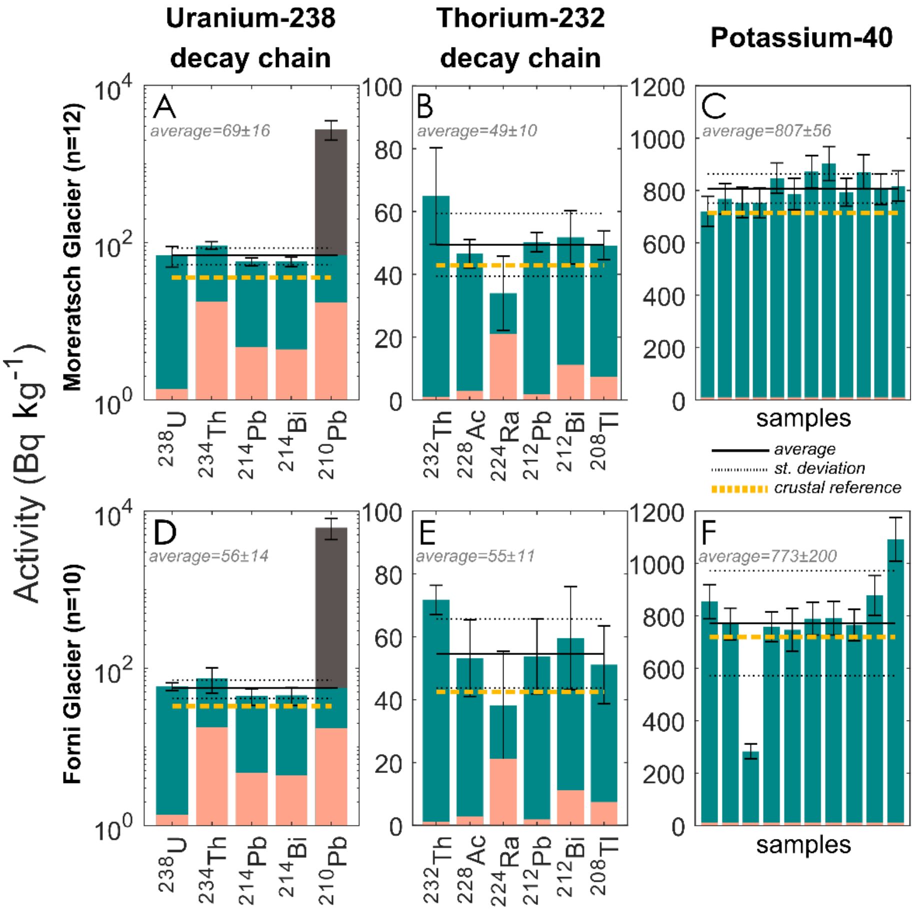


Fig. 3 Activity of the radionuclides belonging to the decay chains of ^{238}U and ^{232}Th and of ^{40}K. The upper row (panels A-C) refers to the cryoconite samples from the Morteratsch glacier, the lower ones (panels D-F) to the samples collected on the Forni glacier. Red bars represent detection limits, green bars measured activities. The activity of ^{210}Pb is divided into supported (green bar) and unsupported fractions (grey bar), considering the upper ^{238}U decay chain as reference for the supported fraction. Crustal references are inferred from Rudnick & Gao (2003).

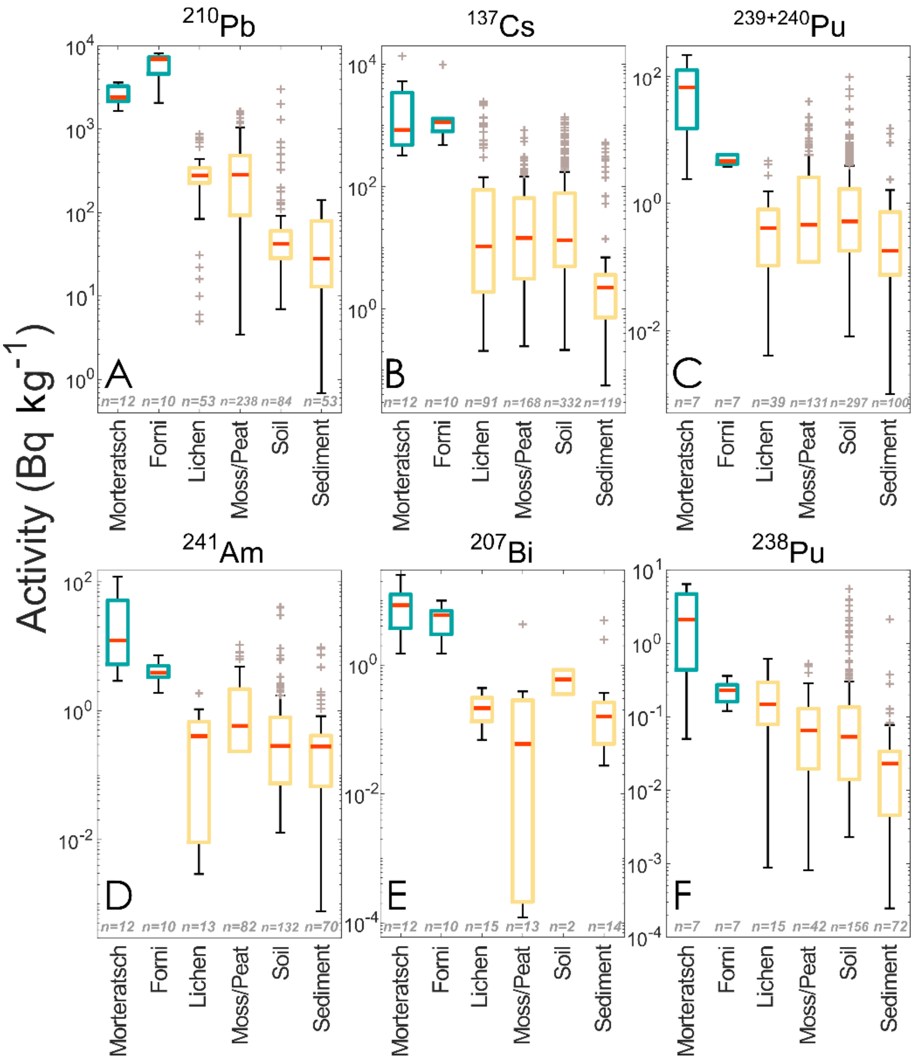
R2: L190: I don't understand why the difference between K40 and the UCC is not significant, but the difference between U and Th is significant, considering the scales on the activity plots. This is because this is beyond my subject area, but may be the case for other readers, so I suggest clearer explanation on the differing scales and assignation of significant differences.

Reply: potassium is one of the most abundant elements in Earth crust and is also the largest responsible for Earth crust radioactivity. It is a common element found in heaps of different minerals. On the contrary both Th and U are orders of magnitude less abundant and are considered trace elements. In addition, they share peculiar chemical properties, with large ionic radii and high ionic charges. Such features make the two elements incompatible with many common minerals and enhance their fractionation in rare and ancillary minerals which are found in specific lithologic contexts. For this reason, it is not surprising that dealing with K we have found concentrations similar to the Earth crust, while this is not the case for Th and U which are enriched in our samples. In Baccolo et al., 2017 we have discussed the fractionation of some elements in cryoconite. We found that elements usually associated to heavy minerals (as Th and U) are enriched in cryoconite because meltwater flow preferentially removes light minerals and enriches the heavy ones. We have added a passage to explain this point:

"These values, as seen in Fig. 3, are slightly higher than the average ^{238}U and ^{232}Th radioactivity of upper continental crust (UCC) reference (Rudnick and Gao, 2003), which is 34 and 43 Bq kg^{-1} for ^{238}U and ^{232}Th respectively. The difference is probably related to the accumulation in cryoconite of heavy minerals, where both U and Th are typically enriched, because of hydraulic sorting related to meltwater flow (Baccolo et al. 2017)."

R2: Figure 4: Nice clear plot, although check the colours again.

Reply: thanks, we increased the color contrast between cryoconite and not-cryoconite samples, now the graph is:



R2: L246: This is really interesting!

Reply: thank you!

R2: L260: Include a reference

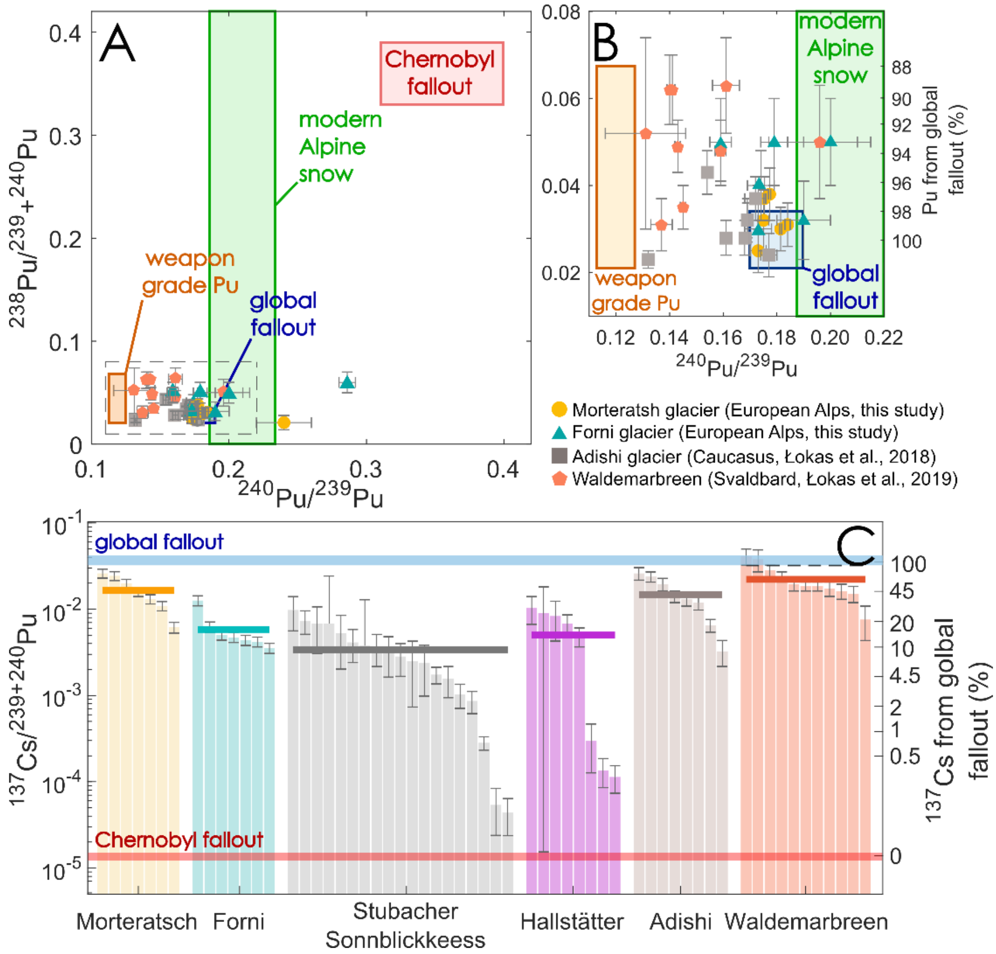
Reply: we have now added two references, Bossew et al., 2006 and Shabana & Al-Shammari, 2001.

R2: L362: Include a ref on plutonium deposition in snow here

Reply: we couldn't find the right position, maybe there's a typo with the line number.

R2: Figure 5 is quite baffling. I like the labelled sections, but it's unclear whether the labels refer to a whole box or a specific point. The percentage lines on the lower plot are also quite confusing – would this be better presented in a table?

Reply: thank you for the comment, we have exaggerated with colors.... We have better highlighted the boxes, removing the colored bands which could be somewhat confusing. In addition, we changed sample markers to help distinguishing them regardless their color. In the lower graph we have removed the labels about the percentage for each glacier, but we have decided to keep the lines corresponding to average values calculated for each glacier. We agree with the reviewer, to increase readability we have now included a table presenting the average fractions of Pu and 137Cs related to global fallout at the different glaciers. This is the new version of the figure:



R2: L360-366: include more details on this in the methods section

Reply: we have moved the technical details in the methods section. A full technical description of this procedure can be found in the cited literature, for this reason we have decided no to add further information here.

R2: Section 4.4: is this relevant to the overall story of the paper? There are many studies exploring carbon and black carbon content of cryoconite, particularly in Greenland, and I wonder if these data would be more relevant in another comparative study.

Reply: we have decided to maintain the section. The first reviewer suggested to better discuss the relationships existing between radionuclides and organic matter content, in particular to explain the differences observed at the two glaciers (cryoconite from Morteratsch is richer in cryoconite than Forni).

R2: L387: typographical error

Reply: corrected.

R2: Figure 7 is slightly confusing, could only the most important be labelled in C?

Reply: we agree, there are a lot of labels, but we don't know hot to select only some of them. The only reasonable way would be to remove all the labels, but without labels the figure would be completely unreadable.

R2: L406: yes, this would be really cool! You could refer to the work of Tranter, Fountain or Bagshaw on using chloride to date hydrological age of cryoconite in Antarctica as an example if you wanted to include a comparison.

Reply: thanks for the advice. We have deepened the discussion about the influence of supra-glacial dynamics on the age of cryoconite. We have added the suggestion references few lines below the point suggested by the reviewer. The new extended passages now read:

"The distribution of cryoconite on glaciers is extremely dynamic and is influenced by meteorological processes, local ice morphology, and supraglacial melting and runoff. It has been observed that within only a few days, single cryoconite holes can form, deepen and collapse, scattering cryoconite granules downstream on the glacier (Takeuchi et al., 2018). In addition, it is known that cryoconite is far from being a static sediment: cryoconite granules are in fact subjected to uninterrupted changes, such as aggregation and break-up, and their lifetime on glaciers don't exceed a few years (Takeuchi et al., 2010). In Antarctica, where cryoconite holes are usually covered by a permanent ice lid and supra-glacial hydrology is poor, the isolation age (i.e. the time period during which a single cryoconite hole have remained isolated from glacial hydrology) of single cryoconite holes has been estimated through a biogeochemical method: it never exceeds a few years (Fountain et al., 2004; Bagshaw et al., 2007). The transience of surficial glacial environments is also confirmed by glacier moss balls, whose lifespan was observed not to exceed few years (Hotaling et al., 2019). Given these evidences, we find it unlikely that a fraction of the cryoconite sampled on the surface of a small and steep glacier as the Stubacher Sonnblickkees, could form at the end of the 19th century and persist there since then without being subjected to significant compositional changes."

R2: L 431-445: This hypothesis seems sound and defendable, except the supposition that cryoconite only forms when meltwater is available (L445). I would rephrase this.

Reply: we have removed that passage

R2: L466: give examples of the legislations, or remove this sentence (it's not particularly relevant)

Reply: we have removed the passage

R2: L484: I think that rather than 'absorbs', 'binds' would be a better description, since you seem to show that the EPS sticking the granules together binds up the impurities as well

Reply: we agree, we have now modified with *"binds and accumulates"*

R2: Final sentence is not strictly relevant and a bit literary.

Reply: we have removed the passage

R2: Data availability are not shown. This must be corrected.

Reply: we have now included the availability statement: *“Full data are available as supplementary material.“*