We thank Anonymous Referee #1 for their considered, detailed, and helpful review of our manuscript. Below we outline our response to each comment in turn (shown in blue text), with the reviewer's original comment in italics.

GENERAL COMMENTS

This manuscript provides an original dataset including gamma-emitting fallout radionuclides activities, geochemical oxide composition/trace element concentrations, and C/N compositions obtained on cryoconite samples (n=14), sediment sources (n=23) and a 38-cm long lacustrine sediment core collected in a 1-km² glaciated catchment of Sweden. The authors discuss the spatial and temporal distributions of these properties in response to glacial processes and sediment transport. They also underline the potential environmental and health deleterious impacts of the redistribution of these potentially harmful substances (e.g. in response to the ongoing global warming and snowmelt processes). Overall, the manuscript is well written, the data is well described and the results are well discussed at the light of those previously published in the literature. However, in my opinion, the study site should be further contextualized. Furthermore, the maps could be improved and the interpretation of some of the results could be taken further (e.g. relying on part of the data analysed although maybe under-exploited, regarding the particle size and the organic matter composition, in particular for cryoconite samples). The interest of this research for tracing the impacts of snowmelt-induced sediment redistribution could also be underlined (instead of focusing only on the potential harmful impacts of the release of radionuclide substances stored in this glaciated area in response to snowmelt, although I acknowledge this interest of doing so, in particular in Sweden, for the reasons well explained by the authors in the manuscript including the substantial post-Chernobyl fallout in this region and the specificities of the regional food diet). Accordingly, I recommend reconsidering acceptance of the manuscript after major revisions have been performed. Nevertheless, I would like to underline the fact that I enjoyed reading the manuscript, and improving the data presentation /interpretation will definitely further increase the impact of this nicely conducted study. I acknowledge the large quantity of fieldwork (in very likely adverse conditions!) and labwork conducted, and I am thankful of having been given the opportunity to discuss these results with the community!

- We respond to the general comments made here within our response to individual comments below.

DETAILED COMMENTS

Abstract

LL18-20 maybe adding values would be useful for the readers here; not sure whether you only refer to the Chernobyl and Fukushima exclusion zones here?

- We don't feel that adding specific values is necessary within the abstract, rather focussing on the key take-home messages of the work.

LL.25-26: as in the general comment above, I would also insist on the interest of analysing these substances as unique tracers to understand the impact of ongoing snowmelt processes on the dispersion of sediment and associated contaminants across catchments in this part of the world.

- We feel that discussion of FRNs as tracers of snowmelt processes and sediment distribution is outside the scope of this manuscript, despite being a very interesting topic. Furthermore, samples

were collected at the end of the ablation period when much snow cover had already disappeared, and no samples were collected within the glacier accumulation zone, thus we don't feel we can assess this aspect further. We acknowledge that radionuclides and FRNs specifically can be used in a wide range of environmental applications, but here we focus on FRNs as contaminants. We direct the referee and other interested readers to a recent paper that specifically focusses on application of ^{210}Pb to different areas of glaciological research: https://doi.org/10.1017/jog.2020.19

Introduction

L.42 not sure I would start this sentence with a 'but'?

- Changed to "However".

L.50 could you provide examples of what would be 'anthropogenic organics'?

- The term "anthropogenic organics" has been replaced with "persistent organic chemicals" and the specific compounds, well known to the scientific community, can be found in the references, as is also the case with the metals.

59 please remove the . before the (

- Removed.

L.60 (and elsewhere in the text): Is 'prevalence' the right term to use here? Wouldn't the terms 'persistence' or simply 'occurrence' be more appropriate?

- Text updated, apart form in the conclusion where we feel that 'prevalence' is the correct term.

L.62 'environmental fate of the radioactivity' >> fate of the radionuclides?

- Personal preference; we prefer the text as currently written.

63 unclear what you mean with 'in addition to any potential socio-economic impacts' here? Could you be more specific?

- Our paper does not try to explore what the socio-economics impacts would be, only to highlight that these types of impacts are possible. Thus, we don't feel it would be appropriate to suggest specific impacts at this stage, however we have added general comments to this matter within the text.

LL.64-65 same remark with 'subsequent dilution in the hydrological system', could you specify what you mean?

- We have now made it clear in the text that this dilution relates to when cryoconite enters proglacial waters.

LL.67-76; I had the impression that this paragraph could be rewritten to better outline scientific questions instead of listing the analyses made and 'arguing' that FRN activities are higher than those found in other environments?
- The statement "we argue" has been removed in line with a suggestion from referee 2, and the text changed to reflect that this finding is a combination of work from different studies.

Study site

In my opinion, this section was pretty short and focused on glacial geomorphology. What about rainfall/snowfall in this catchment? What about the bedrock lithology, the soil characteristics (if this is relevant in this catchment?), etc.

- We agree that the addition of information on climate and geology of the catchment would be helpful here, and have added this to the manuscript.

Methods

Overall, in this section, the sampling period should be better defined (in the introduction, the period of 'August 2017' is mentioned, then nothing else is added… How high was the precipitation during the months before sampling? How was it distributed with time? I guess that this is a crucial aspect for supporting the interpretation of the results (e.g. mainly for Be-7 and Pb-210 data).

- More information on the sampling period has been added to the opening paragraph of the methods section. We do not have access to weather station data local to the site to further analyse the influence of precipitation on ^{7}Be and ^{210}Pb activities, and would only be speculating on the content of ^{7}Be and ^{210}Pb in precipitation.

L.98 the authors refer to 'sources' here, although the reference to sources is no longer used in the interpretation… Were all the potential sources covered by this sampling (or maybe this wasn't the purpose…)?

- We list the sources considered here in the sentence immediately following this statement. A potential source not included is rockfall from surrounding slopes, however this cannot be sampled safely at this location.

L.102 could you be more specific on what you mean when referring to the 'proglacial outlets'?

- Replaced with "proglacial outlet streams".

L.106 At 100°C? Isn't it too high for the organic matter composition analyses?

Had the particulate samples remained moist their composition may have been affected during their transport to the UK. Thus, the samples were carefully dried, immediately after collection, on site, to remove the interstitial water. Water removal is essential prior stable isotope analysis and the samples were sent for analysis directly on return to the lab

L.107 'due to the limited amount of cryoconite available' >> what did it represent in grams of material (to have an idea of the difficulties encountered)?

- We can't put a number on this; it more reflects the difficulty of sampling cryoconite when it is smeared on a rough ice surface, or easily displaced when in water etc. Cryoconite was typically not found in large individual accumulations at this site. However, cryoconite is typically found on glaciers as an aggregate consisting of a few grams each. Sometimes they are easy to sample because they are concentrated (cryoconite holes), but sometimes (as in this case) cryoconite is spread on the surface of the glacier requiring careful consideration during collection.

L.107 why using a < 75µm sieving threshold (compared to the classical 63 µm threshold for instance?)

- It would have possibly been better to apply a standard 63 µm sieve, but only a 75 µm mesh sized sieve was available on site.

L.109 was there a specific preparation protocol implemented before conducting the particle size analyses?

- Samples were prepared as previously outlined in the methods in terms of drying and sieving, and analysed in triplicate as stated within the manuscript.

L.115 the article cited here (Wynants et al., 2020) refers to a study conducted in the African Rift Region with limited additional details on the gamma spectrometry analyses: I am not sure this has an added value here, or did I miss something?

- This reference is included with respect to analytical method only (as it was also conducted using the facilities at the University of Plymouth), and not because this paper is of relevance to glacier systems.

L.130 I guess that you used two certified material samples to account for the matrix composition differences between cryoconites (more similar to moss soil?) and other sediment materials (more similar to the 'soil' TEL-2012-03); is it so?

- The CORiF laboratory regularly participates in IAEA international proficiency tests using their certified materials. The purpose of the repeated analyses of two IAEA soils in this work was to confirm our analytical quality assurance, thereby giving confidence that the gamma counters used in the analysis of key radionuclides were performing at the required standard throughout the analytical phase of the study.

LL.134-147: the section on WD-XRF analyses is well described, except maybe the calibration/validation issue: was it exclusively based on comparing the results obtained with WD-XRF and ICP-OES on a selection of samples, or were certified materials also used for this crucial step?

- Empirical calibration for XRF spectrometry requires the use of matrix-matched standards of known composition and concentration. Given the heterogeneous nature of cryoconite it is not practical to obtain matrix-matched standards for instrument calibration in this instance. Therefore, we used the analysis package, Omnian, developed by PANalytical, which is designed to handle a range of different matrices. Measurement validation can either be obtained by using reference materials or by comparison to results obtained from a different technique. Given the importance of sample composition in XRF analysis, reference materials would need to be matrix matched and there are no suitable reference materials available for this. We, therefore, validated our measurements by comparing them to results obtained from ICP-OES analysis. ICP-OES determines element concentrations in the dissolved phase following digestion of cryoconite, overcoming any matrix effects associated with analysis of the solid material. The ICP-OES procedure was itself a validated approach undertaken in an ISO9001-2015 certified laboratory. Close agreement between results obtained from the two approaches provides confidence in the XRF data.

L.149 wasn't the drying of cryoconites at 100°C a problem for the subsequent stable isotope analyses? Where are the analysed d13C /d15N values of the samples provided? Or did I misunderstand this paragraph and only the TOC/TN were analysed?

- Stable isotope data are publicly available on the Pangaea data repository alongside all other data from this study, and both %C/%N and d13C/d15N were included in the PCA analysis. Please see our previous response regarding drying of the samples.

L.164: the attribution of the year 1952 or 1954 (or even 1955) may be debated for the lower level of lacustrine sediment in which Cs-137 is detected. This gives at least an idea of the uncertainties associated with core dating (+/- 4 years?). What about the level to which the year 1959 (L.169) is attributed? To which event is it related (e.g. the Tsar bomb fallout took place in 1961)? Or did I miss something in the rationale here?

- These are important points of detail that raise issues of clarity in our message. The initial mention of 1959 in the text refers to the known geomorphological event of stream capture by the new lake and hence the rationale for the split approach to dating. The second reference in this paragraph to 1959 was more of a 'wiggle matching' exercise to fallout records in cited literature but not presented. While the reviewer offers interesting detail on the potential cause of this small peak, on reflection of the above comments we think this is a distraction from the main geochronological messages so we have modified the text accordingly to focus on the known date of onset (notwithstanding potential uncertainty).

Results and discussion

L.177 is the title inclusive enough here ('geochemical composition of cryoconite') given the section contents?

- Changed to "Cryoconite composition".

LL.183-184/L.185/etc. at some places, some generalities are given on radionuclides, although I wonder whether they are really relevant (e.g. on the solubility of Cs-137 and the long range transport of Chernobyl fallout…?)

- We feel that this information is relevant for context as the general readership of the Cryosphere may not have prior knowledge of FRN sources and impacts.

LL.193-195 Providing additional information on the lithologies found in the catchment would be very useful to support the interpretations made on the K-40 levels measured in the samples

- This information has been added to the site description in line with the previous comment.

LL.199-200 maybe I missed it here, but I don't see how Fig. 2C illustrates the influence of organic content on the accumulation of radionuclides; additional elements should be provided here to better support this statement…

- We agree that this statement is not helpful here and have edited the text.

L.203 not sure it is relevant to use 2 decimal digits here for the %C and %N?

- We have changed this to one decimal digit.

L.212-230: to better support the interpretation of Be-7 results here, additional information should be provided on precipitation (snow/rainfall + snowmelt) during the months before the sampling campaign was conducted here. Furthermore, given the short half-life of Be-7 (~53 days), information on the analysis period after sampling is also crucial (as Be-7 could be <MDA in some samples just

because some samples were analysed too late, and Be-7 initially present had just decayed to undetectable levels?)

- We agree that comparison with precipitation data could be useful here, however precipitation data are not available via the Tarfala Research Station database post-2013 currently, and we did not collect this data ourselves. We also believe that the ^{7}Be detected in cryoconite samples is likely sourced from recent snowmelt, which precipitation data alone would not help to explore. We are aware, due to considerable team experience of working with 7Be, that samples must be prepared for gamma counting shortly after collection. Thus, prior to the fieldwork (7 - 17/08/2017) it was arranged with the analysts that certain samples would be packed and analysed as soon as practically possible after return to Plymouth.

L.220 maybe the following ESSD manuscript (on Be-7 and Pb-210 levels across the globe) could be of interest to the authors here: https://essd.copernicus.org/preprints/essd-2021-35/, similar work must have been published on Cs-137 as well.

- We thank the reviewer for drawing this publication to our attention.

L.248 how were these metals selected?

- The narrow range of metals is reported in Table A3 arises because there is only a limited number of metals (Cr, Cu, Ni, Pb and Zn) that have appropriate sediment quality guidelines (CCME, 1995). We felt that it was important to set the results of the cryoconite analyses within the context of probable effect levels (PELs) thereby indicating whether a toxic effect would be exerted on local fauna.

L.253 is the normalization to the upper continental crust the most relevant option here to calculate the enrichment factors? Or did you use a more specific dataset for normalization?

- We used the Wedepohl (1995) values for the upper continental crust because this data has international relevance.

L.275 'it is perhaps unsurprising' > I would consider rephrasing the sentence here?

- Now reworded in the text.

L.281/283 problem with the notation of Na2O here (should be a subscript instead of a superscript?)

- Both instances now fixed.

Discussion in subsection 4.2

LL.293-… Overall, here, I think that the comparison of properties in cryoconite vs. other samples should be supported by the comparison of their respective particle size/organic matter compositions, as an enrichment in fine and organic material in cryoconites vs. other samples is expected to control the higher levels of contaminants measured in these samples and, according to the Materials and Methods section, you did analyse these properties…

- We have now added a correlation matrix for each sample type as an appendix to the manuscript (Figure B1). Organic matter composition is only available for the cryoconite samples, so no comparisons can be made here, however the correlation matrices demonstrate where relationships exist with particle size (D[3,2]). Text has also been added to section 4.2 to further discuss this.

LL.299-301 see my remark above on the precipitation before sampling to support the interpretation of the Be-7 results

- Please see our previous response with regards to ^{7}Be and precipitation data, however what we would say is that even if precipitation data had been collected, it would be speculation as to whether it might affect the ^{7}Be loadings without understanding the radionuclide composition of precipitation.

L.323 see my remark above about referring to the lithological characteristics of the study site to interpret K-40 (and I guess supported) Pb-210 here…?

- Lithological information has been added in the study site description.

LL.363-366: not sure if you can go that far in the interpretation regarding Am-241 activities here?

- We are comfortable with this interpretation since the protracted delivery of ^{241}Am to the lake sediment column is not the norm. It implies a continued release from the secondary source – certainly an inference and not fact but worthy of mention given the interest in delivery and transit of FRNs. We have modified the text to be clear this is an inference.

Section 4.4

LL.372-… importantly, here, I would add the implications of these results and the detection of such high FRN activities in material transiting glaciated environments to trace and understand the icemelt/snowmelt-induced redistribution processes in the future?

- We don't yet know what the implications of findings specifically in glacial environments are, so we can only discuss what is understood from other studies. Again, we feel that discussion of snowmelt processes are not within the scope of this study, especially since it was conducted during August which is well into the ablation season when much of the snowpack at lower altitudes has already melted (we did not sample anything within the accumulation zone of the glacier because cryoconite, if indeed it exists there, is hidden by remnant snow).

LL.384-386 'It has been suggested that areas with previous 137Cs contamination may augment 137Cs transfer following future contamination events due to fixation in soils' >> unclear what you mean here, could you please clarify this statement?

- The ^{137}Cs soil fixation process implies a strong adsorption of the radionuclide to the solid phase. Fallout of ^{137}Cs from future contamination events would add to the existing ^{137}Cs burden of soils, thereby augmenting the transfer of this long-lived radionuclide to Arctic fauna. The text has been edited to clarify this point.

Conclusions

Based on the changes made when revising the manuscript, some conclusions could be revised here (e.g. interpretations related to the organic matter content in cryoconites vs. other samples; interpretations related to Be-7 activities…)

- We don't feel that any of the revisions made warrant changes to the conclusions.

L.412: nuclear incidents >> nuclear accidents? Importantly, the thermonuclear bomb testing supplied most of these FRN (and I am not sure that they can be considered as nuclear incidents or accidents?)

- Text updated.

L.434 see the previous comment regarding the use of the 'prevalence' term

- See prior response.

Figures

Figure 1: this catchment map actually does not provide catchment delineation (to the best of my understanding of this map); could this be improved? Furthermore, there is no North arrow/ scale on the inset map of Sweden (I guess this is the map of Sweden?); it is hard to see the river network and the sample symbols are not so easy to see/understand on these maps… This is nice to see the glacier lobes on the image but the other features of interest (sampling locations and types, catchment delineation, river network…) could be presented in a much clearer way in my opinion…

- This figure has been updated to improve clarity of the sample points and labels, and we have removed the inset map of Sweden to make space for a larger figure legend. The stream network is a highly mobile, braided system which changes regularly, thus there is limited value in mapping stream channels from imagery that is not from a time period that coincides with when we were in the field. We have delineated the catchment boundary and included this in the new figure.

Figure 2: it is not easy to read/infer the FRN activities based on this map (as single values are attributed to the circles), the exact values could maybe be added near the circles on the map in red/blue?

- The key thing for readers to take away from this figure is the relative variability in activities, so we don't feel that individual values are required here. All data are publicly available should anyone want to examine this in more detail.

Figure 5: I don't understand why the core samples were split into 4 sections; in my opinion, the core samples should be merged and compared to the potential sources (maybe the continuously supplied FRN such as Pb-210 and Be-7 as they decay with time/with depth in the core and they are therefore not fully relevant for this comparison but some of the geochemical elements/ organic matter properties might be?); I would consider adding the cryoconite properties to the graphs (after normalization to the particle size/organic matter properties?)

- We split the core because the depths represent different periods of time and inferred geomorphological process relevant to changing dynamics of contaminant delivery. The top section of the core may also contain materials that are more easily remobilised, for example. We do not include cryoconite FRN values here because they are between one and three orders of magnitude higher than activities in other sediments and we are comparing material from compartments of the sediment continuum.

Figure 6 is not very easy to read neither, at least the river network should be clearly added to the map?

- The river network is a dynamic, braided, proglacial system and the streams seen on this particular satellite image will not precisely reflect how it looks today, or even at the time we sampled.

Figure 7: figure resolution seems not to be optimal, the years attributed to the peaks could be added on the Cs-137 part of the graph?

- We prefer to leave the dates off as there are inferences in the geochronological discussion, as per above response, and explained in the text. If the figure resolution is still not optimal when we upload the figures individually for the final manuscript then we can look into redrawing the figure.

Appendices

Table A.1 what was the decay-correction date? Not sure that you should provide 2 decimal digits for the Am-241 activities?

- The samples were decay corrected to the sampling dates between 7 - 17/08/2017. The decay correction approach was stated in the original text. The ^{241}Am activities recorded in Table A2 have been adjusted to one decimal place, as suggested.

Table A.2 I would remain consistent with the number of decimal digits provided in this table… Do the values after +/- refer to the SD? For the 'central forefield' samples (n=2!), this is meaningless to provide a mean/SD and I would provide the range of values instead…?

- It was not well explained in the original caption that the +/- values refer to the counting errors. Thus, for the central forefield samples the activity concentrations are reported together with their individual counting errors. We have maintained consistency with the number of decimal digits.

Adding the particle size/organic matter data to this summary table would be very useful…

- The particle size and stable isotope data are publicly available via Pangaea for those who are interested to look at this in more detail, however we feel that we have illustrated the importance of organic content within both figures 2 and 4, and would prefer that table A2 focusses on radionuclides alone. We have, however, now added correlation matrices illustrating the possible influence of these variables on FRN activity as an appendix.

Regarding the footnote on Be-7: information on the time between sampling and analysis would be particularly meaningful here…

- An appropriate comment has now been made in the footnote to table A2.

Table A.3 were the EF calculated based on the data of the upper continental crust here?

- The EFs were estimated using the data for the upper continental crust (Wedepohl, 1995). The original caption has been updated to include the source of sediment quality guidelines.