Manuscript review: Glacial controls on iron and manganese concentration and lability in an Artic fjord

In this study, the authors perform a suite of measurements of dissolved iron (Fe) and manganese (Mn) and suspended particles that were collected across two glacier systems with varying properties. Specifically, the authors investigated systems containing a large marine-terminating glacier (Hansbreen) and a smaller land-terminating glacier (Gåsbreen). A major fraction of the dataset from both systems (aside from CTD) included dissolved Fe and Mn concentrations, as well as measurements of Fe and Mn derived from chemical extractions of the SPM using ascorbic acid (FeA and MnA) and dithionite (FeD and MnD). Other parts of the dataset included some X-ray diffraction patterns and macronutrient measurements. The authors plot the Fe and Mn measurements in a number of ways, including via several "geochemical indices", such as FeA/FeD, and FeA/dFe. Using the large amount of data presented in the manuscript, the authors find considerable differences in Fe and Mn behavior across the different glacier systems. The major trends in the dataset pointed to the system containing the larger marine-terminating glacier having substantially higher inputs of reactive particulate Fe and Mn, as well as dMn. This same system was also characterized by a significantly higher sedimentation rate than the land-terminating glacier. These results are placed generally in the context of the transition from marine-terminating to land-terminating glaciers, which would "progressively reduce glaciofluvial connectivity, erosive and weathering potential of the ice covered part of the catchment, and direct sediment export to the ocean".

This study has several strengths. The introduction is organized and clear. The methods section is very detailed, with the sample collection protocols and analytical measurements appearing to be performed with great care. The results are also organized sufficiently and the figures are simple and clear. The discussion is also sufficient, with particularly good contextualization of specific measurements with previous investigations of similar types glacier systems. In general, the manuscript is written well and has few errors in spelling or syntax, though some mistakes show up more at the end of the manuscript. I also could not personally detect any obvious use of LLMs in the writing, which I have come to really appreciate in the current state of scientific writing.

Authors: Thank you.

The major issue I have relates mostly to the structural analysis of the suspended particulate matter (SPM), which I thought was weak. First, the XRD and SEM analysis in the results section reads largely as inconsequential and is poorly integrated in the manuscript. This part is short, is presented in a kind of off-handed way and has almost no bearing on how the manuscript is otherwise presented. I think this part can be removed without any impact to the remaining manuscript.

Authors: We thank the reviewer for this comment and agree that, in the original manuscript, the pXRD and SEM-EDS results were insufficiently integrated with the broader geochemical interpretation. Should a revised version of the manuscript be invited, we would substantially rewrite this section to strengthen its focus on Fe and Mn cycling and to better integrate the presented results with the overarching aims and conclusions of the study.

The mineralogical differences between the two SPM samples provide important context for interpreting variations in Fe and Mn concentrations. Both catchments deliver similar mineral groups, including detrital silicates, phyllosilicates and carbonates, but in different proportions,

reflecting the contrasting bedrock geology of the two catchments (Birkenmajer, 1990). pXRD indicates a relatively greater contribution of carbonate minerals in Gåshamna and a higher abundance of quartz and phyllosilicates in Hansbukta. This distinction may partly influence sediment-bound Fe concentration and speciation. Consequently, part of the lower Fe content observed in Gåshamna may reflect differences in sediment composition rather than solely differences in weathering intensity or Fe mobilization processes. The mineralogical data therefore help distinguish lithological controls from weathering-related controls on the observed Fe distribution.

The pXRD and SEM-EDS results also provide insight into possible sources of reactive Fe within the Hansbreen catchment. Previous geological investigations have documented the occurrence of chlorite-bearing metasedimentary rocks together with accessory sulphides, including pyrite and pyrrhotite, in the Hansbreen drainage basin (Czerny et al., 1992a; Kieres and Piestrzyński, 1992; Spolaor et al., 2021). SPM samples from Hansbukta have a higher content of phyllosilicates (mica and probably chlorite). Sulphides were not identified by pXRD, which is unsurprising given their likely low abundance, the limited detection capability for trace phases, and the strong Fe fluorescence associated with Cu-radiation analyses. Nevertheless, oxidation of Fe-bearing sulfides represents a plausible source of Fe (oxyhydr)oxides in glacial sediments at Hansbukta.

This interpretation is consistent with the SEM-EDS observations, which revealed Fe-enriched sub-micron aggregates associated with silicate particles. Although SEM-EDS does not permit definitive mineral identification, the morphology and chemical composition of these particles are consistent with poorly ordered Fe-bearing phases and provide independent support for the presence of reactive Fe pools in Hansbukta SPM. Mineralogical differences may also contribute to the contrasting Mn distribution observed between the two catchments. The relatively greater abundance of dolomite in Hansbukta SPM, together with reported occurrences of siderite- and ankerite-bearing veins in the surrounding bedrock, suggests a potential lithological source of Mn (Czerny et al., 1992a; Czerny et al., 1992b).

An error occurred during the preparation of the previous version of figure showing pXRD results (Fig. S.5), and the peak labels and relative intensities did not correspond to the original pXRD data. Should a revised version of the manuscript be invited, the original diffraction patterns and phase identifications would be restored. The corrected Fig. S.5 indicate that SPM from Gåshamna and Hansbukta contains broadly similar mineral assemblages, consistent with the original interpretation, although the relative abundances of individual mineral groups differ between the two catchments, reflecting differences in bedrock geology.

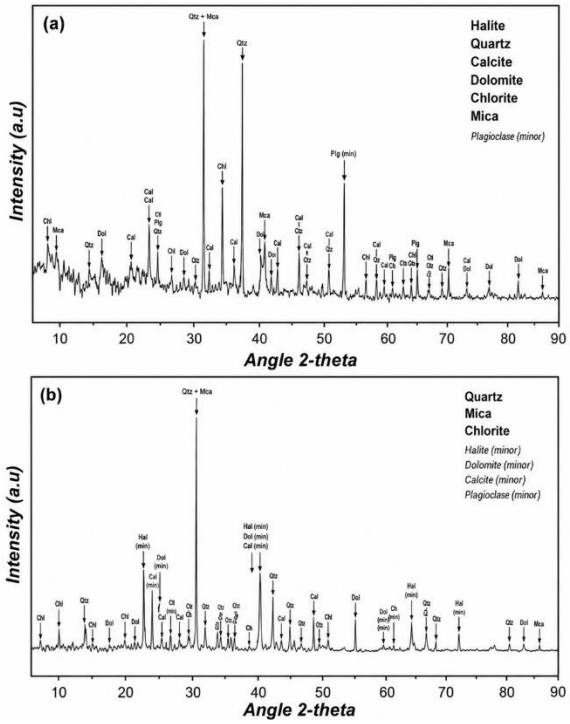


Figure S.5 (corrected). X-ray diffraction patterns of suspended particulate matter from (a) Gåshamna Gashamna and (b) Hansbukta, Hornsund fjord, Svalbard.

Similarly, the section on macronutrients and organic carbon, which is actually given in the discussion even though it reads as a result in my view, is poorly integrated in the manuscript, has little consequence on the conclusions of this work as-written, and can also be removed without changing almost anything else about the manuscript. This is not to say that the XRD and macronutrient information is without benefits, rather, these data are simply not integrated in a meaningful way in the rest of manuscript, which leads the reader to ask why the data were collected and presented. This aspect of the manuscript should be improved by reframing some of the work or by handling these data in a better way.

Authors: We agree that the macronutrient and organic carbon data, as presented in the original manuscript, were not sufficiently integrated with the central objectives of the study. Rather than substantially expanding these aspects, we would streamline the manuscript to maintain a clearer focus on the key processes controlling Fe and Mn distributions. Consequently, we would propose removing Section 5.1.3 ("Role of macronutrients and organic matter distributions: implications for micronutrient cycling") from the main text. The corresponding supplementary discussion (Text S1.3) and associated figures (Figs. S.7 and S.8) would also be removed. Related references would be deleted and supplementary figure numbering adjusted accordingly. The relevance of the XRD analyses would be addressed separately in response to the previous reviewer comment.

Second, I think there is only limited benefits of applying chemical extractions of SPM to create operationally defined pools of FeA, FeD, MnA and MnD. I realize that extraction protocols have been useful in previous decades to operationally differentiate solid-phase Fe and Mn species.

However, I think there has been enough evidence to show that results and conclusions based on chemical extractions alone can be confusing and prone to error and misinterpretations. Considering that the authors also mention the challenges of using extractions to identify Fe phases in the manuscript, I think the authors are also aware of these issues.

I also think that the broader biogeochemical field is starting to move away from deriving conclusions based solely on extractions, particularly given that synchrotron methods are much more effective at measuring (directly) SPM speciation and are becoming more and more accessible, with a growing number of beam lines offering mail-in service for standard samples, such as these. I have the impression that the dataset is already quite complete and that performing an additional set of measurements to directly identify Fe and Mn speciation in the SPM is likely beyond the scope of the study. In that case, the authors must be more careful with their language to avoid overinterpreting the extraction results.

Authors: We disagree with the reviewer. Although studies (including our own) have demonstrated the value of synchrotron approaches (e.g., XAS/XANES/EXAFS) for resolving the oxidation state and mineralogy of particulate Fe and Mn in glacial and fjord environments (Hawkings et al., 2018; Jones et al., 2025; van Genuchten et al., 2022; van Genuchten et al., 2021), they are also prone to misinterpretation and model fitting (particularly when it comes to oxyhydroxide pools in complex samples), do not produce reliable concentration data, and, in our opinion, should be considered complementary to rather than instead of operationally defined extractions. Additionally, synchrotron-based techniques are also somewhat challenging in low Fe/Mn environments such as fjords and coastal systems, and not all beamlines are suitable for this kind of analysis. We are also not aware of any "easily accessible" beamlines – all beamlines are subject to increasingly competitive application procedures with turnaround times of at least several months, and in some cases much longer.

Sequential extractions remain highly valuable and widely used for iron and manganese speciation work. Indeed, extractions using buffered ascorbate and dithionite solutions have been widely applied to quantify reactive and more crystalline Fe and Mn phases in marine and glacial environments precisely because they are well calibrated to know mineral pools (Hawkings et al., 2018; Hawkings et al., 2014; Herbert et al., 2022; Herbert et al., 2020; Laufer-Meiser et al., 2020; Lenstra et al., 2021; Lenstra et al., 2022; Michaud et al., 2020; Piret et al., 2026; Raiswell et al., 2018; Raiswell et al., 2010; Stachnik et al., 2025; Wehrmann et al., 2014; Wunder et al., 2024). While we acknowledge that these fractions are associated with uncertainties, they provide a well-calibrated methodology for assessing contributions from various mineral phases (Lenstra et al., 2021; Raiswell et al., 2010). This study is also, to our knowledge, the first to use these extractions on SPM in marine settings and concentration data cannot be derived directly from synchrotron-based measurements. Similarly, the extraction approach allows direct comparison with previous studies investigating reactive Fe and Mn fractions in glacier-fed rivers, meltwaters, and sediments (Hawkings et al., 2014; Pryer et al., 2020; Stachnik et al., 2025), thereby placing our results within a broader biogeochemical context.

The following statement is a good example, "The greater contribution of highly reactive particulate Fe to the Fe pool is not confined to the immediate vicinity of the glacier but extended across the bay and persisted beyond the sill, indicating substantial lateral transport…" In this statement, the

authors use the operationally defined FeA fraction too loosely, almost as though "highly reactive particulate Fe" was a directly measured and unambiguously defined mineral.

Authors: We agree that, if interpreted in terms of rapid environmental transformation or dissolution during transport, the term may be misleading. However, FeA clearly refers to an operationally defined ascorbate-extractable Fe fraction that is predominantly associated with, and is calibrated to, poorly crystalline Fe and Mn oxyhydroxides, particularly 2-line ferrihydrite (Lenstra et al., 2021; Raiswell et al., 2010), which are known to be highly reactive and at least partially bioavailable (Kranzler et al., 2016). The selectivity of the ascorbate extraction for these mineral phases is well demonstrated (Raiswell et al., 2016; Raiswell et al., 2010). Amorphous and poorly ordered iron phases have larger surface areas and higher kinetic lability than more crystalline phases. Additionally, the terms "reactive Fe" or "highly reactive Fe" have been widely used in the literature to describe this operationally defined pool (e.g. (Hawkings et al., 2014; Raiswell et al., 2006; Shoenfelt et al., 2017). Our interpretation is not that FeA is immediately utilized or dissolvable upon entering the fjord (that will depend not only on the reactivity/dissolution kinetics of the Fe pool, but also of the equilibrium concentration of truly dissolved iron) nor that it is an unambiguously defined mineral (it is more helpful to imply that it is not), but rather that glaciers supply substantial quantities of an operationally reactive Fe pool to coastal waters.

To address the reviewer's concern and avoid ambiguity, we suggest revision of the manuscript to clarify that FeA represents an operationally defined ascorbate-extractable Fe fraction associated primarily with poorly crystalline Fe oxyhydroxides. We could carefully review the use of the term "highly reactive" throughout the manuscript and revised several passages to make this definition and its intended meaning more explicit.

However, it is still unclear to me what is meant by "highly reactive particulate Fe" because, in my thinking, the structure of a highly reactive solid phase is not likely to be preserved in the environment over long times and distances. Here in this example, I think the authors specifically mean FeA, but I think the behavior of this material in the investigated glacier systems is at odds with the operational definition of a highly reactive Fe phase. Perhaps an ascorbic acid-soluble phase is encapsulated in an aggregate that preserves the solid from transforming over long distances, but then I would not describe this Fe species as highly reactive.

Authors: The term "highly reactive" originates from earlier studies and refers to an operationally defined fraction of suspended particulate matter rather than to a specific rate of transformation or dissolution under given environmental conditions (which is arguably a more complex question as we detail below). The term does not imply rapid dissolution or short residence times during environmental transport, and we think our definition is clear in the methodology in that it relates to a mineral pool that is likely to include amorphous and poorly crystalline phases. The persistence of this phase during transport is not necessarily inconsistent with its operational reactivity.

Under conditions of Fe biological limitation, this fraction is generally considered potentially bioavailable. For example, biological uptake and incubation experiments conducted in the Southern Ocean have demonstrated that poorly crystalline Fe phases, including ferrihydrite, can represent an important source of bioavailable Fe for phytoplankton (Shoenfelt et al., 2018; Shoenfelt et al., 2017; Wyatt et al., 2023). Furthermore, the transport of operationally defined reactive particulate Fe over substantial distances has been documented in glacial systems. Forsch et al. (2025) showed that the

reactive (termed “labile” by the authors) Fe fraction extracted using the Berger leach, a method considerably less selective for poorly crystalline Fe phases than the ascorbate extraction used here, remained detectable up to 15 km from the glacier source. These observations indicate that operationally reactive Fe phases may persist during transport despite their potential bioavailability.

Experimental studies in seawater have shown that equilibrium dissolved Fe concentrations are extremely low at pH ~8. For example, Kuma et al. (1996) reported Fe(III) solubilities of 0.15 - 0.30 nM in open-ocean waters and 0.3-0.6 nM in surface waters enriched in natural organic ligands. Likewise, Liu and Millero (2002) reported Fe(III) seawater solubility of 0.2-0.5 nM, while solubility in artificial seawater lacking organic ligands was close to 0.01 nM. Unsurprisingly, the equilibrium dissolution of Fe(III) is thermodynamically constrained, allowing substantial quantities of particulate Fe to persist during transport while remaining potentially available for biological utilization if environmental conditions change. In other words, it is not surprising that we do not see a pronounced dissolution profile even with a large, highly reactive particulate pool. In Fe-limited marine systems, any dissolved Fe released from particulate phases may also be rapidly assimilated by phytoplankton, maintaining very low dissolved Fe concentrations and further limiting net losses from the particulate pool.

In any case, I think this is a problem that the discipline has, and it is not necessarily the author’s job in this work to solve it, indeed I do not have a good answer about how to interpret the extraction results through the lens of assigning an actual Fe mineral phase myself. However, I at least think the authors need to revise the manuscript to avoid over-interpreting what the extraction results actually yield.

Author**s:** We agree that operationally defined extraction methods have inherent limitations and that extraction results should not be interpreted as representing discrete mineral phases with complete certainty. Resolving the precise mineralogical composition and bioavailability of sediment-bound Fe and Mn remains an important research challenge, particularly in glacier-to-coast systems where suspended particulate matter (SPM) contains complex mixtures of poorly crystalline, nanoparticulate, and more crystalline authigenic and/or detrital mineral phases.

We also note that other approaches commonly used in glacial and marine biogeochemistry, such as the Berger leach (Berger et al., 2008) or total dissolvable trace element analyses, have different, perhaps larger limitations. The Berger leach is not calibrated against specific Fe mineral phases and therefore integrates a broader pool of unknown reactive materials and biological material, making the interpretation of the extracted pool less straightforward. Similarly, total dissolvable trace element analyses may include contributions from less reactive mineral structures in addition to potentially bioavailable particulate phases. For this reason, we compared selected samples with Berger leach results to illustrate differences among extraction approaches and provide additional context for interpreting our results.

I have some smaller comments of a technical nature below:

Line 93: Missing a space between and2020

Authors: This will be corrected as suggested.

Line 135-136: A bit unclear why depths of 5 m is listed twice

Authors: “Second” 5 m was related above bottom. It is written in the text.

"At each station, depending on the depth of the station, water samples were collected at depths of 1, 5, 15, 25, 35, 50 and **5 m over the bottom** using a bathymetric rosette equipped with 3.5 L Niskin bottles"

Line 310: I perhaps missed this detail, but it would be nice to have a short explanation about why 2024 seemed to produce more SPM in the two systems compared to other years.

Authors: Although the focus of the study is on the 2022 and 2023 sampling campaigns, we agree that a brief explanation of the elevated SPM concentrations observed in 2024 could provide useful context. In a revised version of the manuscript, we would consider adding a short discussion of the potential causes of this interannual variability, supported by a supplementary figure summarizing seasonal snow cover, air temperature, precipitation conditions and freshwater content in Hansbukta. Specifically, we would note that SPM concentrations in both Hansbukta and Gåshamna were considerably higher during summer 2024 than in the other study years. This period was characterized by relatively thick spring snow cover and elevated summer air temperatures and precipitation, while exceptionally low salinity in Hansbukta in early September suggests enhanced glacial meltwater input. Together, these conditions may have promoted greater sediment mobilization and delivery to the fjord. We suggest adding a new figure to supplementary materials (Fig. REV1).

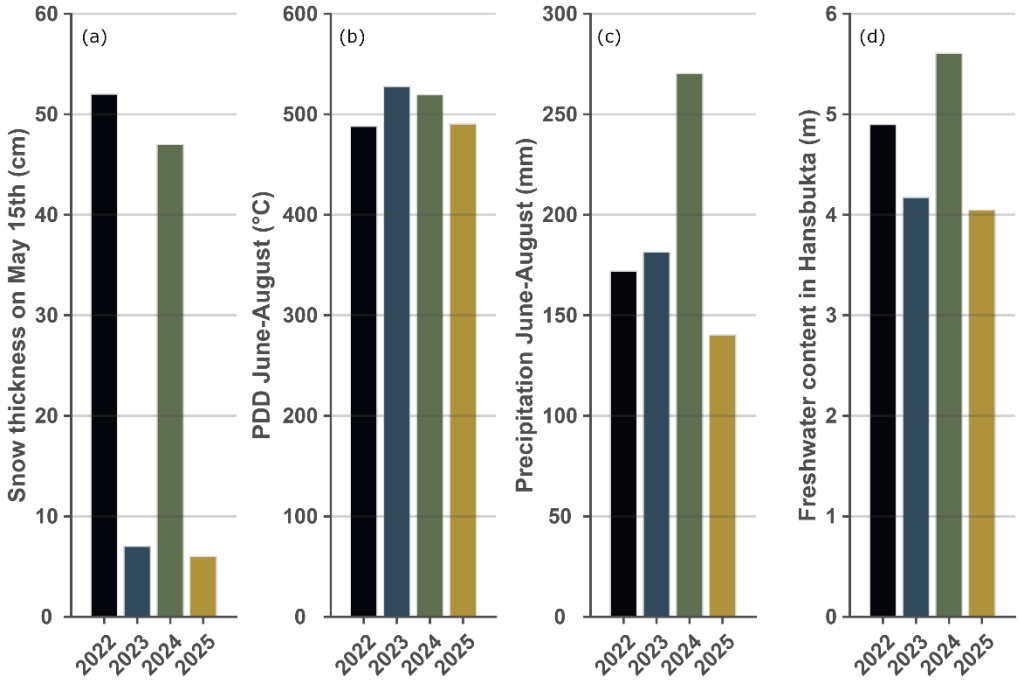


Fig. REV1. Interannual variability in spring snow cover, summer climatic conditions, and freshwater content in Hansbukta during 2022-2025: (A) snow depth on 15 May, (B) positive degree days (PDD) during June-August, (C) cumulative precipitation during June-August, and (D) freshwater content of the upper water column.

Line 439-440: This statement is written very broadly without some qualification. If the authors are referring to the systems investigated in the current work, I would make that clear, otherwise I would include some references here.

Authors: We agree that the original wording may be too broad and could be interpreted as a general statement applicable to all coastal environments. Should a revised version of the manuscript be invited, we would clarify that this observation refers specifically to the two glacier-fed bays (Hansbukta and Gashamna) investigated in this study. We would also consider revising the terminology throughout the manuscript to more consistently emphasize that FeA and FeD represent operationally defined highly reactive Fe fractions rather than direct measures of bioavailable Fe.

Line 456: "A" potential process

Authors: This would be corrected as suggested.

Line 467-468: This sentence reads as if FeA is a distinct mineral phase to me, which I think is incorrect. At the very least I would replace "is formed" with "can be formed"

Authors: We would change to "can be formed" and improve entire sentence to address issues raised by Reviewer.

Line 470: "the" Berger leach

Authors: This would be corrected as suggested.

First paragraph of section 5.1.2: Probably a writing style issue, but there are many missing parenthetical brackets in this paragraph "(110-130 nM sedimentary substratum (Yang et al. 2022)" needs a ")" at the end.

Authors: This would be corrected as suggested.

Line 507: more than "an" order

Authors: This would be corrected as suggested.

Line 518: Missing a space between Arctic and (Bhatia

Authors: This would be corrected as suggested.

Line 567: two .. at the end of the sentence.

Authors: This would be corrected as suggested.

Line 573: should probably be "promotes" instead of "promoting"

Authors: This would be corrected as suggested.

Line 582: Missing a space after the .

Authors: This would be corrected as suggested.

Line 588: Missing a space between "salinity" and "and"

Authors: This would be corrected as suggested.

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