

Permafrost thaw controls iron flux from wetlands and sulfide-bearing rocks to Arctic rivers and streams

Corresponding Author: Professor Roman Dial

This manuscript has been previously reviewed at another journal. This document only contains information relating to versions considered at Communications Earth & Environment.

This file contains all editorial decision letters in order by version, followed by all author rebuttals in order by version.

Attachments originally included by the reviewers as part of their assessment can be found at the end of this file.

Version 0:

Decision Letter:

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Dear Professor Dial,

Your manuscript titled "Thawing permafrost stains streambeds and rusts rivers in Arctic Alaska" has now been seen by 3 reviewers, whose comments are appended below. You will see that they find your work of some potential interest. However, they have raised quite substantial concerns that must be addressed. In light of these comments, we cannot accept the manuscript for publication, but would be interested in considering a revised version that fully addresses these serious concerns. Specifically, a revised manuscript must:

1. Clearly present the ALT-pH relationship using individual borehole data and include sulfate concentration data to strengthen the ARD-ALT argument.
2. Revise the definition of dissolved and nanoparticle iron to align with established literature and adjust conclusions accordingly.
3. Expand the discussion to include global comparisons, clarify the role of pyrite weathering versus microbial iron reduction, assess the predictive modeling's accuracy, and justify the claim that AMD products remain locked in soils during winter.

We hope you will find the reviewers' comments useful as you decide how to proceed. Should additional work allow you to address these criticisms, we would be happy to look at a substantially revised manuscript. If you choose to take up this option, please either highlight all changes in the manuscript text file, or provide a list of the changes to the manuscript with your responses to the reviewers.

When resubmitting, please provide a point-by-point response to the reviewers' comments. Please submit your responses as a separate file, distinct from your cover letter where you can add responses to the Editors' comments that you do not want to be made available to the reviewers. Word files are preferred. We recommend that any figures, tables or graphs that are included in the response to reviewers are also included in the main article or Supplementary Information.

Please bear in mind that we will be reluctant to approach the reviewers again in the absence of substantial revisions.

If the revision process takes significantly longer than three months, we will be happy to reconsider your paper at a later date, as long as nothing similar has been accepted for publication at Communications Earth & Environment or published elsewhere in the meantime.

We are committed to providing a fair and constructive peer-review process. Please do not hesitate to contact us if you wish to discuss the revision in more detail.

Please use the following link to submit your revised manuscript, point-by-point response to the reviewers' comments with a list of your changes to the manuscript text (which should be in a separate document to any cover letter), a tracked-changes version of the manuscript (as a PDF file) and any completed checklist:

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Please do not hesitate to contact us if you have any questions or would like to discuss the required revisions further. Thank you for the opportunity to review your work.

Best regards,

Alireza Bahadori, PhD
Associate Editor
Communications Earth & Environment

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REVIEWER COMMENTS:

Reviewer #1 (Remarks to the Author):

Dear Roman J. Dial & co-authors of COMMSENV-25-0214-T,

Thank you for submitting your manuscript for review. The review has been prepared as a PDF document and is attached (COMMSENV-25-0214-T_Review.pdf).

My overall recommendation is to reject the manuscript, with an invitation to resubmit. In its current form, the manuscript contains significant inaccuracies in the definition of iron phases. Additionally, the data required to support one of the main novel conclusions of the study—that ARD correlates with ALT from the previous year—are not sufficiently presented or accessible for review based on the figures and data provided.

However, the manuscript does contain valuable data and findings for the scientific community, and it addresses an important and timely issue in the context of ongoing climate change. I would therefore be pleased to see the study published once the changes and clarifications outlined in the review are addressed.

Kind regards,
Anonymous Reviewer

Reviewer #2 (Remarks to the Author):

see attachment

Reviewer #3 (Remarks to the Author):

Review of Thawing permafrost stains streambeds and rusts rivers in Arctic Alaska

Dial et al., present multiple lines of evidence to show that permafrost thaw in regions of metal-sulfide substrate and/or wetlands contributes to increasing iron concentrations in high latitude aquatic bodies. This is a highly relevant and timely study that would have large readership. I suggest that this manuscript needs to be restructured to communicate the evidence to the reader. I also suggest that the manuscript acknowledges the limits of what can be answered to date – the evidence motivates many exciting avenues for future work. I recommend that this paper is not accepted in its current format. Please find my comments below:

My comments are mainly regarding manuscript structure and communicating the multiple field locations and data types in a narrative that links back to the paper aim and that can be read and understood by a large, multi-disciplinary readership. The comments mainly focus on the Introduction and Results.

Overarching comments:

1. Final paragraph in your Introduction: suggestion to start the paragraph with the overall aim, which I currently read to be the last sentence of the paragraph. And then link the steps outlined (the two areas and the river flux calculation) to this aim. Suggest avoiding the word 'importantly' just for the final sentence as it doesn't do justice to the other sentences in the paragraph.
2. Results: suggest combining results and discussion section allowing the reader to know the implication for a finding for the paper aim. For example, in Line 121- Line 122, you state that vegetation is controlling snowpack which is controlling active layer thickness. You could then state why this is relevant to the aim of your paper – i.e., does it demonstrate that vegetation index would need to be included in a process-based model that estimates where iron is released to rivers? Or if the discussion is kept separate then please make sure to link back to the results and then link results and discussion back to the road map outlined in the introduction and the aim of the paper.
3. Results (Line 97 – 149): currently the end of this section outlines its limitations, but the section itself is a results section, therefore I recommend stating the results as one part of the jigsaw puzzle that contributes to the aim outlined in the Introduction, rather than stating their limitation to the overall aim – other sources of evidence come together for that.
4. Results: recommend moving method-themed paragraphs, such as the wilderness sampling and sample filtration strategy, to a longer introduction section or to the Methods. These are key field and lab methods but could be stated underneath the steps outlined in the 'road map' at the end of your introduction. Currently, some of the paragraphs in Results are allocated to methods and some to results. Building in the Methods to the approach part of the end of introduction could be a way forward here, linking back to the need for a clearer 'road map' at the start of this paper. At the end of the introduction, I would also recommend a clearer outline of what field sites are used to address what objective to deliver the overall aim.
5. Results: a suggestion to re-order/title the sections and include the lithology section (Line 210-222) upfront, as logically the rocks were there prior to the permafrost formation – so one logic would be to order data bedrock, permafrost extent, climate influence on permafrost extent, geochemistry (AMD), geochemistry (FRM), relationship between geochemistry and discharge.
6. Results and Discussion: I do not write as a modelling expert, but from an interested, still learning, reader of this paper, it would be unclear to me how successful your modelling approach is. I suggest including an analysis of the predictive modelling in the discussion section – to inform future work in the community. E.g., do modelled estimates match present day values? What additional information is needed to build on this work.
7. Results/discussion: please justify with literature and/or your own data, the proposed theory that AMD products are locked away in soils during winter months or remove temporality from the paper results and include it as a motivator for future work.
8. Please avoid qualitative statements such as 'well above' and I recommend shortening sentences and avoid split sentences because it is harder for the reader to follow.

List of line-by-line edits:

Abstract

Line 45 – 47: It is unclear what is meant by trace metal diversity – perhaps use 'type and concentration'.

Line 45 – 47: Please include Fe in 'stream flux' to read 'stream Fe flux'.

Introduction

Line 50: Suggestion to use 'high latitude' to encompass boreal and Arctic, otherwise state the importance of distinguishing between for the paper aim.

Line 50-51: I would recommend removing the opening sentence because it is too general. I would have an opening sentence directly linking to the unforeseen large-scale increase in Fe concentrations in high latitude rivers (O'Donnell et al., 2024).

Line 56: Replace 'these' with air temperature and precipitation'

Line 58 – 60: This sentence implies that if liquid water is in contact with previously frozen ground, then iron will be released. Please include the words 'potential for' and consider a sentence such as 'exposes previously frozen iron-bearing minerals to present day environmental conditions', 'unveils a new source of iron-bearing minerals'.

Line 68: Suggest to state: 'in locations where soils, sediments and rocks contain metal-sulfide minerals.'

Line 71: Please reverse this sentence structure to begin the sentence with 'microbial iron reduction is a process common in...' and have a separate sentence that stating evidence for the link between FRM and permafrost thaw.

Line 73: Please include references to other studies that have demonstrated microbial iron reduction with permafrost thaw, including: Herndon et al., 2017; Patzner et al., 2020, Patzner et al., 2022

Line 76: Please replace 'ultimate fate' with a specific e.g., whether Fe-bearing minerals dissolved in permafrost soils. Also consider, that environmental conditions upon thaw will determine whether or not the Fe is precipitated as secondary Fe-bearing minerals in the permafrost soils or released into the rivers.

Line 77: Please provide detail on what you mean by 'thicker active layers' – it can either be read as deeper active layer relative to a neighbouring soil or deeper active layer relative to previous years.

Line 80: Please state how large this region is

Line 86: Please make it clear how this river is linked to the two areas you describe above in this paragraph

Line 88: A suggestion is to start the paragraph with the overall aim, which I currently read to be the last sentence of the paragraph. Or perhaps it is not stated. Then link the steps outlined (the two areas and the river flux calculation and modelling) to this aim. Suggest avoiding the word 'importantly' just for the final sentence as it doesn't do justice to the other sentences in the paragraph, and is not discussed in the discussion section to the extent that justifies its importance.

Line 80 – 88: The distinction between the discontinuous – continuous permafrost boundary and the boreal – Arctic boundary may be a source of confusion to readers. I understand what the boundaries are, but the reader has not been introduced to their importance for your study at this stage of the paper.

Results

Line 96: Suggest having a separate results section for permafrost thaw and section for the dissolved solids. Consider including a section on substrate/bedrock composition and permafrost extent, prior to this – to set the scene, prior to discussing thaw and river geochemistry. The pairing of substrate composition with permafrost extent and geochemistry is a key novelty of the paper for the discussion section, and it would be good to include geology and permafrost upfront in the results.

Line 91: Please remove the word 'observed'. Replace 'identified' with 'observed'.

Line 91 – 95: It is unclear how this paragraph fits with the paper aim and results lay-out. I read it and want to first know that you mapped the sulfide bearing mineral formations. Their distribution in relation to permafrost extent and thaw, is an interesting result to state. I am also unsure where this 'single stream' is relative to the 300 locations.

Line 98: Please explicitly state that the permafrost instability was determined using a calculated stability index. At present, the 'maximum instability' is stated upfront in the paragraph without the reader knowing what is meant by 'instability'.

Line 105: Suggest avoiding the split-sentence i.e., when you include additional information bracketed by two commas, because it can be hard for a reader to follow.

Line 112: Please state what is meant by 'well beyond the influence'? – please state the areal extent of mining and the % contribution of the mining area to your area of fieldwork.

Line 114 – 118: Please break this sentence into shorter sentences: In response to 2018 115 stream rusting unrelated to mineral exploration or development, Teck drilled four boreholes 116 (Extended Data Table 1) to record ground temperatures beneath two plant community types 117 common across the Arctic (sedge and shrub tundra) in an undisturbed valley 11 km north and 118 100 m above all mine infrastructure (Supplementary Fig. 3).

Line 118 – 121: Please reverse this sentence to begin with the aim e.g., 'To model ground temperatures in the 200 km² area over time, we used...': Using drill-logs, soil properties, and weather records, we calibrated the Geophysical Institute Permafrost Lab model[17] 119 (parameters in 120 Extended Data Table 1), then modeled 2010–2023 ground temperatures (MGT) to estimate active layer thickness (ALT).

Line 127: please remove 'abundant components' and state the percentage of trace elements (Fe, Zn, Al etc) in the TDS. Also note that TDS will increase with weathering therefore an increase in trace metals may correspond with an increase in major cations, such that the proportional contribution of Fe, Zn, Al to the TDS remains the same. Consider determining the stoichiometry of the element composition / or use element ratios, do you see a relative increase in the trace elements corresponding to a metal sulfide source?

Line 125- 133: Please state why these findings are significant for the paper (or at least the discussion section) aim.

Line 128-129: Please remove 'well above' and provide quantitative text.

Line 135- 140: Please correct me if I misunderstood, but I see that your observations of stream staining in Autumn contradict your hypothesis that AMD products are trapped in soils during winter months. I agree that a portion will be trapped over winter because e.g., the pH and Eh of soils promoted secondary iron mineral formation, rather than the release of Fe from porewaters into the river, however you observe a portion of the AMD to get into the rivers in Autumn. Further research is required regarding the rate of Active Layer freeze-up vs the rate of transport of AMD products from the soil. I recommend to state what more needs to be done to determine the temporality of when AMD products are transferred from soil to river.

Line 139-141: I am unclear how this final sentence is consistent with your observations, or what observations you are referring to.

Line 146-149: Recommend removing: this dataset does not distinguish 147 FRM sources, correlate Fe release to permafrost or lithology, nor estimate Fe flux in rivers and 148 streams. Because it states what a data set has not been able to do, rather than summarising what part of the overall aim this paper section has contributed to.

Line 189-192: Please make this into multiple sentences. It is hard to follow.

Line 194-195: Please state what you mean by Iron in stained tributaries differed little with discharge.

Figures:

Figure 1: To help the reader link between paper aim, field sites, data collected at field sites, I recommend simplifying by annotating these maps in a way that matches the 'road map' proposed at the end of the introduction – each part of the map/field location can be linked to one (or more) sections within the results.

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Decision Letter:

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Dear Professor Dial,

Your manuscript titled "Permafrost thaw controls iron flux from wetlands and sulfide-bearing rocks to Arctic rivers and streams" has now been seen by our reviewers, whose comments appear below. In light of their advice we are delighted to say that we are happy, in principle, to publish a suitably revised version in Communications Earth & Environment.

We therefore invite you to revise your paper one last time to comply with our format requirements and to maximise the accessibility and therefore the impact of your work.

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We hope to hear from you within two weeks; please let us know if you need more time.

Best regards,

Alireza Bahadori, PhD
Senior Editor
Communications Earth & Environment
Consulting Editor
Communications Sustainability

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

I have now carefully evaluated the revised manuscript and the authors' detailed response to the reviewers. I am satisfied that my previous concerns have been thoroughly addressed.

The major claims—that (1) permafrost thaw exerts a predictive control on iron flux to Arctic rivers via two distinct pathways (wetland-derived Fe and sulphide-driven ARD), and (2) active layer thickness anticipates downstream ARD-related chemical signals by approximately one year—are now clearly articulated and more convincingly supported by the data and analyses.

The manuscript has been substantially improved: the restructuring of the Results and Discussion clarifies the narrative; the ALT–ARD relationship is now prominently presented; and the strengthened sulphate–TDS and stoichiometric evidence provide convincing support for ARD attribution. The statistical approaches and added detail on model calibration and data availability also address previous concerns regarding rigor and reproducibility.

I have no further substantive comments and consider the manuscript suitable for publication.

Reviewer #2 (Remarks to the Author):

The authors have done a good job revising the manuscript following my recommendations. No further comments from my side.

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Response to reviewers of COMMSENV-25-0214-T by Dial et al.

“Permafrost thaw controls iron flux from wetlands and sulfide-bearing rocks to Arctic rivers and streams”

Response to Reviewer 1 comments on COMMSENV-25-0214-T

We thank Reviewer 1 for a thorough and constructive evaluation of our manuscript. We appreciate their recognition of our study as an important contribution to a timely topic with potential for valuable contributions. We have addressed all comments, substantially restructuring the manuscript to improve clarity and presentation of our key findings.

Major Comments

Comment 1: The Relationship between ARD and Active Layer Thickness (ALT)

Reviewer: The reviewer raises several concerns:

- (a) the ALT-pH relationship was hidden as a subfigure within a subfigure
- (b) data should show individual boreholes rather than just averages
- (c) both input and output files for GIPL2 modeling should be provided
- (d) sulfate concentration vs. previous year ALT should be plotted to substantiate the ARD mechanism.

Response: We have substantially revised the presentation of this key finding. The boreholes, as it is now made clear in the manuscript, are all located within the same small upper catchment:

1. In Methods (~L839-848): "Responding to widespread, natural, ARD in 2018, Teck drilled four boreholes along a 220 m transect..."
2. In Methods (~L863-866): Coordinates are provided for all four boreholes: (Sedge Tundra BH3: N 68.1829, W162.959582; Sedge Tundra BH4: N 68.182244, W162.957916; Shrub Tundra BH1: N 68.18178, W 162.95812; Shrub Tundra BH2: N 68.181434, W162.956096)
3. In Methods (~L866): "...all located within 47-212 m from [each other]"
4. In Figure 6 caption: "Boreholes located 47-212 m apart within a 0.45 ha sample area at 377-394 m asl in the headwaters of West Fork Ikalukrok Creek (Fig. 5a)."

Because the stream TDS and pH (Teck did not collect sulphate from the stream monitoring stations, unfortunately) are catchment-level integrative concentrations of ions and other small particles, it is more appropriate to compare them to a catchment level estimate of mean active layer thickness.

We believe that the reviewer's request with this major comment was to provide more than a single replicate of the result "ARD shows response one year after thaw". However, point samples of ALT are not as informative for understanding catchment level effects of thaw as an ensemble of measurements of ALT. The fact that ALT varied by several meters within tens of meter distances between boreholes suggests a great deal of variability in ALT.

Instead, we now provide seven stations of TDS measurements over 7-8 years. We do not have seven stations of pH or sulfate, unfortunately. However, a new map (Fig. 5a) makes clear the location of eight acid seeps with sulphate and TDS showing a near perfect correlation between the two, strengthening the assumption that stream TDS is a proxy for ARD.

- (a) Prominent display: The ALT-ARD relationship is now presented in dedicated figures. Figure 7 shows the one-year lag between ALT and pH/tributary staining across four panels, with the correlation ($R^2 = 0.93$, $p < 0.001$) prominently displayed in panel 7c. Figure 8 shows September TDS vs. ALT relationships for all seven monitoring stations individually ($R^2 = 0.82-0.94$). Figure 9 presents heat maps comparing concurrent vs. lagged correlations between TDS and ALT.
- (b) Individual borehole data: Figure 6 shows ALT dynamics for all four boreholes individually (Sedge BH3, Sedge BH4, Shrub BH1, Shrub BH2) across 2014-2022. Supplementary Table 2 provides all GIPL2 parameters for each borehole.
- (c) Data availability: We have revised our data availability statement (L932-934) to indicate The chemical, sample location, and other data that support the findings of this study are available at Figshare DOI 10.6084/m9.figshare.28091102. We will ensure both input AND output files for GIPL2 modeling are included in the final repository.
- (d) Sulfate evidence: Figure 8 shows seven stations of TDS over time and against previous year ALT of the ensemble of boreholes that together represent the Ikalukrok watershed. Figure 5b shows TDS is >85% sulfate in $n = 8$ acid seeps, as demonstrated in where $R^2 = 0.999$ for SO_4^{2-} vs. TDS upstream of Ikalukrok 207. Supplementary Figure 8a shows a similar result for $n = 14$ seeps that includes those upstream eight plus six more widely scattered acid seeps. The strong correlations of TDS on ALT in Figure 8b ($R^2 = 0.82-0.94$) across seven stations together with the near perfect correlation between TDS and sulfate, support the claim that this is an ARD relationship, not merely pH vs. ALT as in Figure 7c for a single station. Additionally, Figure 3d shows the SO_4^{2-} vs. dissolved Fe relationship approaching theoretical ARD stoichiometry (2:1 molar ratio) in stained streams and acid seeps.

Comment 2: Definition of Dissolved Iron

Reviewer: The reviewer correctly notes that nanoparticles (1-100 nm) would pass through the 0.45 μm (450 nm) filter, so the correlation with turbidity reflects particles larger than 450 nm, not nanoparticles. Definitions of dissolved, nanoparticulate, and suspended iron phases need revision.

Response: We thank the reviewer for this important clarification. We have revised our terminology throughout the manuscript:

- (a) The Methods section (L704-712) now explicitly defines our operational terms: "Metal concentrations from samples passing through a 0.45 μm filter and subsequently acidified are referred to as 'dissolved' metals. Metal concentrations from unfiltered samples are referred to as 'total' recoverable metals... Particulate metal concentrations were calculated as particulate Fe = total Fe – dissolved Fe."

- (b) We have removed the inaccurate reference to "nanoparticles" and now correctly describe the relationship as being between particles $>0.45 \mu\text{m}$ and turbidity. The revised text (L244-245) states: "When excluding acid seep samples, turbidity (formazin nephelometric units, FNU) was also very strongly correlated with particulate Fe ($r = 0.92$, $n = 41$)."
- (c) We acknowledge (L706-709) what the $0.45 \mu\text{m}$ operational definition means: "Acidification of unfiltered water samples is not expected to liberate metals from all particulates (e.g., clay minerals, silicates, etc.), but it is expected to release metals from the most bio-available particulates, such as Fe and Al oxyhydroxides." This revision ensures consistency with standard operational definitions in the literature.

Comment 3: Restructuring of the Manuscript to Avoid Methods and Discussion Material in Results

Reviewer: The reviewer identified:

- (a) Methods content in Results (e.g., lines 112-122 describing methodology; "Extensive sampling in wilderness" describing fieldwork logistics; lines 98-105 describing temperature calculations).
- (b) Discussion content in Results (e.g., lines 135-137 speculative interpretation about ARD trapping; lines 143-149 interpretive claims about causal pathways).

Response: We have completely restructured the manuscript to clearly separate Methods, Results, and Discussion:

- (a) Methods content relocated: All methodological descriptions have been moved to a comprehensive Methods section (L513-930). This includes: - Climate data and thermal indices (L542-567) - Regional mapping of orange-colored waterbodies (L569-607) - Geologic setting (L608-644) - Subregional sampling (L646-693) - Water sampling and laboratory analysis (L695-726) - Statistical modeling (L728-759) - Iron flux estimation (L761-800) - Local study area descriptions (L802-809) - (Ikalukrok Creek watershed seep pH, sulfate, and metals L810-827) - (Ikalukrok Creek stained tributary counts L829-837) - Ground temperature modeling and active layer thickness (L838-898) - One-year lag analysis (L900-930)
- (b) Discussion content relocated: Speculative and interpretive content is now in the Discussion section (L333-511). For example, the speculation about ARD trapped over winter now appears at L349-369: "The one-year lag between active layer depth and downstream ARD product concentrations could be explained by solute transport that slows (or even stops) following freeze-back in November and then begins with seasonal thaw in April, increasing over summer into fall...."
- (c) Results section: The Results section (L144-331) now presents findings with very minimal and only necessary methodological or interpretive content, organized by spatial scale: - "Annual thermal loss reached its minimum in 2019" (L145-155) - "Rusting is associated with lithology and permafrost" (L157-230) - "Water chemistry varies by waterbody type" (L231-286) - "Acid seep hydrogeochemistry varies by bedrock formation" (L289-305) - "Active layer depth anticipates ARD in streams by one year" (L307-331)

Minor Comments

Title

Reviewer: The title is highly similar to the O'Donnell et al. 2024 publication. The current title does not fully reflect the novelty or findings of the study. Please consider rephrasing it to better capture the unique contributions of this work.

Response: We have revised the title to:

"Permafrost thaw controls iron flux from wetlands and sulfide-bearing rocks to Arctic rivers and streams."

This title distinguishes this work from O'Donnell et al. (2024) by:

- 1) Emphasizing dual-pathway mechanisms (wetlands via FRM AND sulfide-bearing rocks via ARD).
- 2) Highlighting the control/predictive aspect of permafrost thaw.
- 3) Specifying "Arctic rivers **and** streams".
- 4) The revised title better captures our unique contributions: the quantitative linkage between ALT and ARD products, the dual FRM/ARD framework, and the predictive model based on geology and permafrost.

Abstract

Attribution of pH to ARD

Reviewer: Further work is needed to demonstrate that the pH is driven specifically by ARD, rather than other sources of acidity.

Response: We have strengthened the evidence linking pH to ARD in the revised manuscript. The abstract (L37-38) now states: "Notably, ARD-related chemical signals correlate strongly with active layer thickness from the previous year, indicating a temporal lag." In the Results, we demonstrate that:

- 1) TDS at acid seeps is >85% sulfate (Figure 5b, $R^2 = 0.999$).
- 2) TDS correlates very strongly with stained tributary counts ($r = 0.91$, L319-320). These multiple lines of evidence collectively demonstrate that the observed acidity is specifically driven by ARD.

Introduction

Introduction structure

Reviewer: The introduction provides an essential overview of the topic, as well as highlighting the novelty of the study in the final paragraph.

Response: We thank the reviewer for this positive assessment. We have retained the structure while improving the final paragraph (L124-143) to more clearly articulate the three spatial scales (regional: $2.41 \times 10^5 \text{ km}^2$; subregional: $2.02 \times 10^3 \text{ km}^2$; local: $2.06 \times 10^2 \text{ km}^2$;) and how they address the questions of where, how, and when Arctic river rusting occurs.

Results

Splitting up the Results Section by spatial division

Reviewer: Clearly attributing spatial divisions in the results section, and linking to the final paragraph of the introduction, would increase readability.

Response: We have reorganized the Results section to clearly follow the spatial framework introduced at the end of the Introduction. The Results now proceed logically through:

- 1) Temporal context: "Annual thermal loss reached its minimum in 2019" (L145-155) - establishing the climatic driver.
- 2) Regional scale (L157-181): "Rusting is associated with lithology and permafrost" - OCWs across the Brooks Range linked to Zn-polygons and permafrost zones.
- 3) Subregional scale (L183-230): Acid seeps in KOVA, regression models of dissolved Fe vs. geology and permafrost ($\text{adj-R}^2 = 0.72$).
- 4) Subregional scale (L231-286): Detailed characterization of water chemistry by waterbody type, flux calculations.
- 5) Local scale (L289-331): Ikalukrok Creek watershed, borehole data, and the one-year lag analysis.

This structure mirrors the Introduction's framing and eliminates the "Extensive sampling in wilderness" section that previously mixed methods with results.

Discussion

Extension of Discussion following Reorganization of Results

Reviewer: The discussion would benefit from more direct analysis of the results. The first paragraph reads more like a conclusion than a discussion.

Response: We have substantially revised the Discussion.

- 1) The opening paragraph (L334-346) now directly synthesizes results rather than concluding: "Bedrock geology and permafrost thaw in combination are determinants of where, how, and when Arctic river rusting occurs... we identified clear evidence of distinct pathways operating simultaneously."
- 2) We have added detailed discussion of results including:
 - a. the one-year lag mechanism (L347-369);

- b. comparison with previous localized studies (L412-421);
 - c. quantitative synthesis of correlations (L423-429);
 - d. feedback mechanisms (L431-438).
- 3) Speculative content (e.g., Medieval Warm Period magnetite, fish population effects) is now clearly framed as future research questions in a dedicated "future research" section (L496-511), introduced with "Much remains for future research."
 - 4) We have added a brief Conclusions section (L477-494) to separate summary statements from discussion.

Figures

General Comment on Figures

Reviewer: It is difficult to discern the key novelty from the figures due to large number of subfigures. Consider reducing subfigures by moving some to supplementary material. One figure per key result would be clearer.

Response: We have completely reorganized the figures to improve clarity:

- 1) Figure 1: Single-panel showing declining permafrost stability at Kotzebue (1943-2023) - the climatic context.
- 2) Figure 2: Study area maps (a-b) and regional/subregional results (c-f) - where rusting occurs.
- 3) Figure 3: Water chemistry by waterbody type (a-d) - distinguishing ARD vs. FRM.
- 4) Figure 4: Sulfate and iron fluxes along Salmon River (a-d) - the dual-pathway flux.
- 5) Figure 5: Local study area map and acid seep chemistry (a-f) - Ikalukrok watershed.
- 6) Figure 6: ALT dynamics for all four boreholes individually (a-d) - ground temperature modeling.
- 7) Figure 7: The key finding - ALT vs. pH/staining lag relationship (a-d) - prominently displayed.
- 8) Figure 8: TDS vs. ALT for seven stations individually (a-b) - supporting the lag.
- 9) Figure 9: Heat maps of concurrent vs. lagged correlations (a-b) - statistical summary.
- 10) Supplementary figures now contain supporting detail (e.g., individual seep photos, additional scatterplots).

Figure 1

Reviewer: This figure acts both as a map and results figure, which is confusing. Consider separating map from results. Figure 1d (Arctic-wide) should be removed as it is not part of the study area.

Response: We have completely redesigned Figure 1. It is now a single-panel figure showing the annual thermal loss index at Kotzebue (1943-2023), establishing the unprecedented permafrost instability of 2014-2019. The map content has been moved to Figure 2. We have removed the previous Figure 1d showing Arctic-wide rusting; this is now mentioned in text with appropriate

references (L47, Supplementary Fig. 1 for Russia). Borehole locations are now clearly shown in Figure 5a with the label "BHs" and coordinates provided in the Methods as cited above.

[Figure 2](#)

Reviewer: This figure hides one of the key findings in subfigure 2e. Please consider removing some subfigures to make this result more prominent.

Response: The ALT-pH relationship (formerly hidden in Fig. 2e) is now the focus of dedicated Figure 7, which shows: (a) mean ALT from four boreholes peaking in 2019; (b) autumn pH at Ikalukrok 207 reaching maximum acidity in 2020; (c) the regression of pH vs. previous-year ALT ($R^2 = 0.93$, $p < 0.001$); and (d) stained tributary counts. Figure 2 in the revision now focuses exclusively on study area maps and the regional/subregional spatial associations with geology and permafrost.

[Figure 3 \(Modeled vs. Measured Temperature\)](#)

Reviewer: A comparison of modeled versus measured temperature would be helpful if this is a key result.

Response: We have added validation information for the GIPL2 modeling in the form of Supplementary Figures 15-19 that compares temperatures and active layer thickness during the calibration periods. A Methods subsection *Error estimation* (L884-898) describes how we estimated errors by comparing modeled ground temperatures (MGT) to thermistor ground temperatures (TGT) at 6-9 node depths over 3-5 years.

However, more importantly we compared active layer thicknesses and found them well captured in 3 of the 4 boreholes. In the fourth borehole, the modeling did not capture an apparent talik at 6.1 m.

"Error estimation We estimated errors in mean daily ground temperature near the base of the active layer by using thermistor ground temperatures (TGT) recorded at 6-9 node depths (0.09–6.10 m below ground) during 3–5 years (2018–2023). Supplementary Fig. 15 compares modeled and observed temperatures at the depths where thermistors recorded ground temperature near the active layer depths. We also compared the measured active layer thickness based on thermistor measured temperature (ALT_{obs}) to modeled thickness (ALT_{mod}) for the years of thermistor recordings (Supplementary Figs. 16-19):

BH1 shrub tundra: model was well constrained ($ALT_{mod} - ALT_{obs} = 0.33 \text{ m} < 1.53 \text{ m}$).

$ALT_{obs} = 4.57 \text{ m} < ALT_{mod} = 4.9 \text{ m} < 6.1 \text{ m}$ = next deepest thermistor

BH2 shrub tundra: model missed talik at 6.1 m.

$4.8 \leq ALT_{mod} \leq 5.2 \text{ m} < 6.1 \text{ m} = ALT_{obs} = 6.1 \text{ m}$ = deepest thermistor

BH3 sedge tundra: model was well constrained ($ALT_{mod} - ALT_{obs} = 0.47 \text{ m} < 0.61 \text{ m}$).

$ALT_{obs} = 2.13 \text{ m} < ALT_{mod} = 2.6 \text{ m} < 2.74 \text{ m} = \text{next deepest thermistor}$
BH4 sedge tundra: model was well constrained ($ALT_{mod} - ALT_{obs} = 0.48 \text{ m} < 0.62 \text{ m}$).
 $ALT_{obs} = 0.9 \text{ m} < ALT_{mod} = 1.38 \text{ m} < 1.52 \text{ m} = \text{next deepest thermistor}$

Summary of Revisions for Reviewer 1

We have substantially revised the manuscript in response to all major and minor comments. Key changes include:

1. Prominent display of ALT-ARD relationship: Dedicated Figures 7-9 now highlight this key finding with individual borehole data and multiple supporting analyses (Major Comment 1)
2. Corrected iron phase terminology: Removed "nanoparticle" references; operational definitions now explicitly stated (Major Comment 2)
3. Complete manuscript restructuring: Clear separation of Methods, Results, and Discussion (Major Comment 3)
4. New title: "Permafrost thaw controls iron flux from wetlands and sulfide-bearing rocks to Arctic rivers and streams" - distinguishing from O'Donnell et al. 2024
5. Reorganized figures: Reduced complexity, one key result per figure, supporting material moved to Supplementary
6. Results organized by spatial scale: Regional -> Subregional -> Local, matching Introduction framework
7. Data availability: Both input AND output GIPL2 files will be provided in the Figshare repository

Response to Reviewer 2 comments on COMMSENV-25-0214-T

We thank the Reviewer 2 for their positive and constructive evaluation of our manuscript. We are pleased that the Reviewer found our data "convincing," "clearly presented and carefully analyzed," and our conclusions "reliable." We particularly appreciate the reviewer's expertise in biogeochemistry. We hope that our careful consideration of all suggestions has successfully strengthened the discussion and improved terminology.

General Comments

General Comment 1: Discussion Section Expansion

Reviewer: Considering the very large (fantastic!) dataset the authors have, the discussion section is relatively short and a bit disappointing. The main conclusion is that "Its potential for harm through toxic effects...remains uncertain." Maybe the authors can expand here and discuss: Are these processes expected to take place in other parts of the world? What are the consequences for trace metal transport into oceans (similar to Rio Tinto in Spain)? What are the expected consequences for organic carbon mobilization and greenhouse gas emission?

Response: We thank the reviewer for encouraging us to expand the Discussion and we have done so (L333-511). Key additions include:

(a) Global context (L371-379): We now explicitly discuss extension to other regions: "Where lithology and permafrost allow, these patterns may extend elsewhere in Alaska, nearby Canada, Scandinavia, and alpine settings." We cite studies from Yukon (Lacelle et al. 2007), Italian Alps (Colombo et al. 2025), Colorado (Todd et al. 2012, Manning et al. 2024), Spanish Pyrenees (Zarroca et al. 2021), and Peruvian Andes (Santofimia et al. 2017).

(b) Trace metal transport to oceans (L462-467): We now discuss downstream effects: "...suspended particulate Fe can continue downstream, maintaining turbidity, harming nearby aquatic ecosystems....In contrast to diminishing populations of aquatic organisms such as fish and macroinvertebrates⁵, fertilizer effects of iron on aquatic and nearshore ocean environments³⁸ could lead to downstream algal blooms." (citing Morel & Price 2003).

(c) Carbon cycling (L469-475): We have added: "Permafrost thaw also mobilizes dissolved organic carbon that can fuel greenhouse gas production in downstream aquatic systems. While our study focused on metal chemistry, the processes documented here likely enhance climate feedbacks through CaCO₃ reactions with ARD and consequent carbon flux and through FRM-mediated carbon export...."

General Comment 2: Relative Importance of Pyrite Weathering vs. Microbial Fe(III) Reduction

Reviewer: Can the authors evaluate more specifically the relative importance of pyrite weathering versus microbial Fe(III) mineral reduction and dissolution at the two field sites?

Response: We have added some evaluation of the relative importance of ARD vs. FRM in the Discussion (L337-346): "...at the subregional scale we identified clear evidence of distinct pathways operating simultaneously. ARD from upland bedrock sources delivers iron, elevated sulfate, and acid. FRM in lowland wetlands delivers iron with minimal sulfate. The resulting bimodal iron flux in the Salmon River reflects this dual-pathway system where both pathways contribute measurably."

We provide quantitative comparison: "Stained tributaries draining uplands presented average dissolved Fe of 0.58 ppm with high SO_4^{2-} (440 ppm), indicating ARD dominance. Wetlands presented average dissolved Fe of 0.98 ppm but minimal SO_4^{2-} (5 ppm), indicating FRM dominance."

At the larger catchment scale (L344-347): "the similarity of wetland and Kobuk River chemistry (EC, SO_4^{2-} , total Fe, dissolved Mn, $\text{SO}_4^{2-}:\text{Ca}$) suggests FRM may dominate at the larger catchment scale."

We also note (L452-455) that the Kobuk River daily Fe flux of nearly 80 Mg d^{-1} with low sulfate and low EC, similar to wetlands, "further highlights the importance of FRM as an iron source."

We have not done a quantitative assessment of the ARD vs FRM contribution and find it outside our scope at this point, even as important as that measure might be. At the close of the main text we bring up its likely importance and need for quantitative assessment: (L503-510): "More generally, given permafrost occurrence, geography, and geology, the quantitative balance of iron flux of ARD versus FRM would be informative. The effects of ARD are striking and important but in Arctic and subarctic ecosystems anoxic soils are widespread. As with the Kobuk River (Supplementary Fig. 12c), wetlands may well contribute Fe(II) flux (Supplementary Fig. 12a,b) along thousands of kilometers of Arctic rivers, acting as the largest source of iron flux. The effects of FRM on Alaska's Arctic Slope (Supplementary Fig. 13) where thawing permafrost is qualitatively changing tundra landscapes⁴⁴ used by multiple fish species for spawning⁴⁵ could be widespread and substantial."

General Comment 3: Dissolved Organic Carbon Data

Reviewer: Do the authors have any data on dissolved organic carbon in the water samples? If yes, can they discuss the importance of their processes for biogeochemical carbon cycling?

Response: We did not measure dissolved organic carbon (DOC) in our water samples, which is a limitation. However, we have added some discussion of the carbon cycling implications (L469-

475): " Permafrost thaw also mobilizes dissolved organic carbon that can fuel greenhouse gas production in downstream aquatic systems. While our study focused on metal chemistry, the processes documented here likely enhance climate feedbacks through CaCO_3 reactions with ARD and consequent carbon flux^{10,32,33} and through FRM-mediated carbon export^{14-17,39,40}. Viewed from a wider ecosystem and Earth system perspective the balance of Fe(II) and Fe(III) in each waterbody type and carbon cycling via Fe-organic complexed matter in wetland-derived waters^{14-17,39,40} versus ARD-derived^{10,20,32,33} ones are important but currently unknown." We cite relevant literature on Fe-organic complexed matter in wetland-derived waters (Herndon et al., Lipson et al.) and ARD-derived carbon flux (Walsh et al., Zolkos et al., Calmels et al.). We also identify this as a future research direction (L475). This represents an important avenue for future work that could build on our geochemical framework. However, it is outside the scope of this manuscript, unfortunately.

Specific Comments

Comment 1

Reviewer: L38: "...thickness of the previous year"?

Response: We have clarified this in the revised abstract (L37-38): "Notably, ARD-related chemical signals correlate strongly with active layer thickness (ALT) from the previous year, indicating a temporal lag." The phrase "from the previous year" now indicates the one-year lag relationship.

Comment 2

Reviewer: L40-42: in the abstract it is not clear what data exactly has been used. "we use climatic, hydrological and geological context" - what exactly does this mean? Please clarify.

Response: We have revised the abstract (L32-34) to be more specific about the data used: "Here we investigate iron loading across regional to local scales in Arctic Alaska using climatic, hydrogeochemical, and borehole data together with mapped geology and permafrost presence." This clarifies that we use: (1) climatic data (temperature indices); (2) hydrogeochemical data (water chemistry samples); (3) borehole data (ground temperatures); (4) mapped geology; and (5) permafrost mapping products.

Comment 3

Reviewer: L44: introduce abbreviation ALT first.

Response: We now introduce ALT at first use of "active layer thickness" in the abstract. (L38)

Comment 4

Reviewer: L66: I recommend calling these precipitates "Fe(III) (oxyhydr)oxides" because this includes oxyhydroxides such as ferrihydrite or goethite but also oxides such as hematite or magnetite.

Response: We have adopted this terminology throughout the manuscript. The revised text now uses "Fe(III)(oxyhydr)oxide precipitates" (L85, L95, L105, L459) and "Fe-(hydr)oxides" (L105) consistently. For example, L85-86: "Rust coloration from ARD occurs when Fe(II) in acidic solution is buffered and so produces Fe(III) and Fe(III)(oxyhydr)oxide precipitates."

Comment 5

Reviewer: L73: please add the information that this is Mn(IV) (e.g. MnO₂).

Response: In the revised manuscript, manganese is most centrally mentioned in the context of acid seep chemistry (L300-303): "In all but seep S8, Mn was the most abundant of 10 metals (Al, Mn, Fe, Zn, Ni, Cu, Co, Cd, As, Pb)." The oxidation state discussion is beyond our scope as we measured total Mn concentrations, but we have ensured terminology is consistent throughout.

Comment 6

Reviewer: L128: "2.7 m" (space missing).

Response: Corrected. All measurements now include proper spacing between values and units throughout the manuscript (e.g., "2.7 m", "1 m", "0.45 µm").

Comment 7

Reviewer: L156: "<1 m" (space missing).

Response: Corrected throughout the manuscript. For example, L128 now reads: "probability of near-surface (<1 m) permafrost" with consistent formatting.

Comment 8

Reviewer: L163-164: did the authors also distinguish Fe(II) and Fe(III)? Would be interesting to see how much Fe is actually present in the reduced state or in the oxidized state, complexed by OM.

Response: We did not speciate Fe(II) vs. Fe(III) in our samples, which we acknowledge as a limitation. We have added this as a future research question in the Discussion (L473-474): "Viewed from a wider ecosystem and Earth system perspective the balance of Fe(II) and Fe(III) in each waterbody type " We note this for future work that could complement our current geochemical framework.

Comment 9

Reviewer: L172: "Fe(III) (oxyhydr)oxides".

Response: Corrected. We now consistently use "Fe(III)(oxyhydr)oxide" terminology throughout, including at L108-109: "Fe(II) may be lost faster than its reoxidation during rapid expansion of anoxic environments, allowing its introduction to adjacent waterbodies" where it subsequently forms Fe(III)(oxyhydr)oxides.

Comment 10

Reviewer: Line 279 (and in general): the authors are referring to "metal-reducing" microbes. Wouldn't it be more accurate to say "iron(III)-reducing microbes"? Or do the authors have evidence that other metals are reduced as well? How about manganese?

Response: Fair enough. We have revised the terminology to be more precise. The revised text now uses "microbial iron reduction (FRM)" (L34, L58) and "Fe(III)-reducing" or "iron-reducing" microbes specifically. For example, L62: "In FRM, Fe(II) is sourced instead by microbial reduction of Fe(III) under anoxic conditions." We do not have direct evidence of Mn reduction in our system, so we have avoided the broader "metal-reducing" terminology. Where we discuss Mn, it is in the context of observed concentrations in acid seeps (L300-303), not microbial reduction.

Comment 11

Reviewer: Lines 291-292: "...can form from microbial reduction of Fe(III) (oxyhydr)oxides by anaerobic microorganisms".

Response: We have adopted the recommended terminology. The relevant sections now consistently refer to "Fe(III)(oxyhydr)oxides" when discussing microbial reduction processes. For example, in the Introduction (L91-92): "In this process, heterotrophic microbes use Fe(III) as an electron acceptor during anaerobic respiration, producing Fe(II) as a byproduct."

Comment 12

Reviewer: Line 292: wouldn't it make sense to replace reference 28 by a reference that shows magnetite in soils, not in marine sediments?

Response: The revised Discussion (L498-502) cites Darby and Bischof (2004) specifically in the context of Chukchi Sea sediment cores as a paleoclimate proxy, which is appropriate for that discussion. "Well defined layers of detrital opaque Fe minerals (45 – 250 μm), principally weathered magnetite (Fe_2O_3) and hematite (Fe_2O_3), have been dated to the Medieval Warm Period (800-1,100 years before present) in Chukchi Sea sediment cores⁴³, suggesting Arctic rusting could have occurred during a warming event that lasted 300 years."

Comment 13

Reviewer: Line 294: "Fe(III) (oxyhydr)oxides".

Response: Corrected throughout. All references to iron precipitates now use the recommended "Fe(III)(oxyhydr)oxide" terminology.

Comment 14

Reviewer: L579: "Fe and Al (oxyhydr)oxides".

Response: Corrected. The Methods section (L707-709) now reads: "it is expected to release metals from the most bio-available particulates, such as Fe and Al oxyhydroxides."

Comment 15

Reviewer: L604: "30 m" (space missing).

Response: Corrected. All measurements now include proper spacing throughout the manuscript.

Figure Comments

Comment 16

Reviewer: Figure 2, caption: "2.7 m" (space missing).

Response: The conceptual results of old Figure 2 can now be found in Figures 1 and 7-8; however, in all captions and main text we have searched for and replaced missing spaces between values and units.

Comment 17

Reviewer: Figure 4, caption: "1-2 m" and "<1 m" (space missing).

Response: Corrected. The old Figure 4 is now Supplementary Figure 2 and its caption has been edited to include spaces before units.

Comment 18

Reviewer: Figure 5: why are the metals in panel c called "Biogenic metals"? In the main text, this term is not used. Also, Fe(II) can be produced by chemical reduction of Fe(III) by sulfide (that means Fe(II)diss can also be chemical).

Response: We have replaced Figure 5 with a map of the local study area (Ikalukrok Creek watershed) and panels showing regression of SO_4^{2-} on TDS and percentage of metals from

representative acid seeps. The new Figure 3 shows some of the same elements of the old Figure 5 but without "Biogenic metals" terminology.

Summary of Revisions

We have addressed all comments from Reviewer 3. Key changes include:

1. Substantially expanded Discussion: Added global context, ocean trace metal transport, carbon cycling implications, and aquatic ecosystem impacts (General Comment 1)
2. Quantitative evaluation of ARD vs. FRM: Explicit comparison showing ARD dominates in upland stained tributaries (high SO_4^{2-}) while FRM dominates in wetlands and at larger catchment scales (low SO_4^{2-}) (General Comment 2)
3. Carbon cycling discussion: Added despite lack of DOC data, with citations to relevant literature and identification as future research direction (General Comment 3)
4. Consistent "Fe(III)(oxyhydr)oxide" terminology throughout (Comments 4, 9, 11, 13, 14)
5. Revised to "iron(III)-reducing" or "iron-reducing" microbes instead of generic "metal-reducing" (Comment 10)
6. Corrected all spacing in measurements (e.g., "2.7 m", "<1 m", "0.45 μm ") (Comments 6, 7, 15, 16, 17)
7. Clarified abstract data description and introduced ALT abbreviation properly (Comments 2, 3)
8. Removed "Biogenic metals" label from Figure 5 (Comment 18)
9. Identified Fe(II)/Fe(III) speciation as future research direction (Comment 8)

Response to Reviewer 3 comments on COMMSENV-25-0214-T

We appreciate Reviewer 3's detailed and constructive evaluation of our manuscript and that it is "a highly relevant and timely study that would have large readership." We have addressed comments regarding manuscript structure, narrative clarity, and communication of the multiple field locations and data types. The manuscript has been substantially restructured following the reviewer's guidance.

Overarching Comments

Overarching Comment 1: Introduction Final Paragraph Structure

Reviewer: Suggestion to start the paragraph with the overall aim, which I currently read to be the last sentence of the paragraph. Then link the steps outlined (the two areas and the river flux calculation) to this aim. Suggest avoiding the word 'importantly' just for the final sentence.

Response: We have restructured the final paragraphs of the Introduction (L118-143) to lead with the aim and provide a roadmap:

The next-to-last paragraph now opens with context: "Given the complexities involved during permafrost thaw, we investigated river rusting in arctic Alaska by considering rusting observations in the context of geology and permafrost presence at three spatial scales: regional, subregional, and local."

We then state our aim (L120-122): "Specifically, we sought to determine the roles of warming, geology, permafrost, and physical processes in determining the flux of iron into streams and rivers of Arctic Alaska from both wetlands and acid seeps."

The three spatial scales are then described with specific areas and objectives.

1. Regional ($2.41 \times 10^5 \text{ km}^2$) as Brooks Range large-scale geology and permafrost (L124-125)
2. Subregional ($2.02 \times 10^3 \text{ km}^2$) as Salmon River basin in KOVA and adjacent watersheds catchment lithology and permafrost probability (L125-132)
3. Local ($2.06 \times 10^2 \text{ km}^2$) as Ikalukrok Creek watershed borehole data for ground temperature modeling (L132-141)

We removed "importantly" and conclude with how these datasets collectively address our aim (L141-143).

Overarching Comment 2: Combine Results and Discussion / Link Findings to Aim

Reviewer: Suggest combining results and discussion section allowing the reader to know the implication for a finding for the paper aim. For example, vegetation controlling snowpack controlling ALT - state why this is relevant to the aim.

Response: We have chosen to maintain separate Results and Discussion sections as advised by the editor and following guidance from Reviewer 1, but have substantially improved the linkages between them:

1. The Discussion now links back to specific Results. For example, L403-405: "Active layer thickness in shrub tundra reached twice that in sedge tundra, emphasizing that vegetation mediates snow trapping and thaw depth."
2. We added a statement of implications for predictive modeling (L387-393): "Statistical models of dissolved iron in KOVA streams performed best when incorporating geologic unit and near-surface permafrost probability, outperforming models with either variable alone. The best-performing model explained 72% of variance in dissolved iron concentrations, demonstrating robust predictive capability for identifying vulnerable watersheds."
3. Regarding vegetation, the Discussion section (L405-408) now states the modeling implication: "Increasing air temperature and winter precipitation is associated with sea-ice loss. We identified more orange-colored water bodies nearer the Chukchi Sea (where sea ice loss is leading to increased and more variable snowfall) than near the Beaufort Sea."
4. Each major finding in the Results section is now echoed in Discussion with explicit relevance to the paper's aim of predicting where, when, and how rusting occurs.

Overarching Comment 3: Results Should State Contribution, Not Limitations

Reviewer: Currently the end of this section outlines its limitations, but results should state results as one part of the jigsaw puzzle that contributes to the aim, rather than stating their limitation.

Response: We have revised the Results section to frame findings positively as contributions to our overall aim rather than emphasizing limitations. For example, the section on the Teck/Ikalukrok Creek watershed dataset (L328-331) now concludes with the positive finding:

"Mean ALT of the previous year correlated strongly and significantly with Autumn pH at Ikalukrok 207 ($r = 0.96$, $n = 8$ year-pairs, $p < 0.001$) and with count of stained tributaries of Ikalukrok ($r = 0.83$, $n = 6$ year-pairs, $p = 0.02$)."

The previous text stating what the dataset "does not" do has been removed. Instead, we describe what each dataset contributes:

1. Regional data: establishes spatial associations with geology and permafrost zones.
2. Subregional data: identifies dual ARD/FRM pathways and enables predictive modeling
3. Local data: demonstrates the one-year temporal lag mechanism

Limitations are now discussed in the Discussion section (L393) or identified as future research directions (L470-475 and L496-511).

Overarching Comment 4: Move Methods from Results to Appropriate Section

Reviewer: Recommend moving method-themed paragraphs, such as wilderness sampling and sample filtration strategy, to a longer introduction section or to the Methods. Include clearer outline of what field sites are used to address what objective.

Response: We have reorganized the manuscript to separate Methods from Results:

1. All methodological content has been moved to a comprehensive Methods section (L513-930), including.
 - a. Climate Data and Thermal Indices (L542-567)
 - b. Regional Mapping of Orange-Colored Waterbodies (L569-607)
 - c. Geologic Setting (L608-644)
 - d. Subregional Sampling (L646-693) includes field campaign logistics
 - e. Water Sampling and Laboratory Analysis (L695-726)
 - f. Statistical Modeling (L728-759)
 - g. Iron Flux Estimation (L761-800)
 - h. Local Study Area details (L802-930)
2. The Introduction's final paragraphs (L118-143) now provides a clear "roadmap" linking each field site to specific objectives.
 - a. Regional scale: determine associations with geology and permafrost zones
 - b. Subregional scale: characterize ARD vs. FRM pathways, build predictive models
 - c. Local scale: test temporal relationships between ALT and ARD products.
3. The Results section now contains only findings, organized to match this roadmap.

Overarching Comment 5: Reorder Results - Lithology First

Reviewer: Suggest re-ordering to include lithology section upfront, as logically the rocks were there prior to permafrost formation. Order: bedrock, permafrost extent, climate influence, geochemistry (ARD), geochemistry (FRM), relationship between geochemistry and discharge.

Response: We have reorganized the Results section following a logical progression:

1. "Annual thermal loss reached its minimum in 2019" (L145-155) establishes climatic context/driver
2. "Rusting is associated with lithology and permafrost" (L157-230)
 - a. this section now leads with: Regional rusting associations with Zn-polygons (SEDEX/MVT deposits) and permafrost zones (L158-181)
 - b. Subregional acid seeps in Devonian-age *units (L183-197)
 - c. Statistical models linking geology + permafrost to dissolved Fe (L199-230)
3. "Water chemistry varies by waterbody type" (L231-272) distinguishes ARD vs. FRM signatures as ARD indicators (low pH, high SO_4^{2-} , high $\text{SO}_4^{2-}:\text{Ca}$ ratios) and FRM indicators (variable pH, low SO_4^{2-} , high dissolved Fe) and Fe and SO_4^{2-} flux calculations (L274-286)
4. "Acid seep hydrogeochemistry varies by bedrock formation" (L288-304) - detailed local geochemistry
5. "Active layer depth anticipates ARD in streams by one year" (L306-316) - temporal relationship.
6. The Geologic Setting is fully described in Methods (L607-643), providing context.

Overarching Comment 6: Model performance

Reviewer: It would be unclear to me how successful your modelling approach is. Suggest including analysis of predictive modelling in the discussion - do modelled estimates match present day values? What additional information is needed?

Response: We have added explicit discussion of model performance.

(a) Statistical model performance (L212-219): We now report adjusted- R^2 values for all candidate models: - Permafrost-only: $\text{adj-}R^2 = 0.41$ - Geology-only: $\text{adj-}R^2 = 0.48$ - Multiplicative interaction: $\text{adj-}R^2 = 0.67$ - Combined additive: $\text{adj-}R^2 = 0.72$.

(b) GIPL2 ground temperature model validation (L883-897 in Methods): We describe comparison of modeled ground temperatures (MGT) to thermistor measurements (TGT) at 6-9 node depths over 3-5 years. Three of four boreholes showed narrow, unbiased error distributions in 2019 at depths nearest modeled ALT. Error analysis is also provided by Supplementary Figures 15 (temperature comparison at depths near ALT) and Figures 16-19 (comparisons of measured and predicted depths of ALT).

(c) Discussion of model limitations (L392): "However, models based on geologic mapping require local calibration."

(d) Discussion future possibilities (L479-480): "Monitoring ground temperatures in strategic locations could provide advance notice of impending ARD, allowing managers to assess fisheries impacts."

(e) The strong correlations ($R^2 = 0.93$ for pH in Figure 7c and $R^2 = 0.82-0.94$ for TDS across seven stations in Figure 8b) are explicitly stated as evidence of predictive capability (L491-493).

Overarching Comment 7: Justify “winter solute trap hypothesis” or Remove

Reviewer: Please justify with literature and/or your own data the proposed theory that AMD products are locked away in soils during winter months [“winter solute trap hypothesis”], or remove temporality from results and include as motivator for future work.

Response: We have revised our treatment of the “winter solute trap hypothesis” as follows:

1. We now frame this as a proposed mechanism rather than established fact (L360-364): "Taken together, these lines of evidence and ground temperature modeling (Fig. 6) support the idea that autumn freezing air temperatures occurring slightly after maximum ALT is reached initiate top-down freeze-back. This ground refreezing seals ARD somewhere in the active layer (e.g., at the top-down/bottom-up convergence) until winter ends², when thaw liberates ARD products that mobilize downstream."
2. We now present supporting evidence from our data (L351-355): "Our 2015-2022 TDS time series included May-October monthly measurements for most stations. An April 2016 TDS sample from Rachel Creek was the lowest for that year (80 ppm), and TDS averaged across monitoring stations during August-October was 1.2-3.4x TDS during May-July, indicating greater late than early thaw season release of ARD (Supplementary Fig. 11)."
3. We add observational evidence (L355-358): "Field observations and 1-3 m resolution imagery of Fe precipitates on top of aufeis (Supplementary Fig. 4) also suggested that Nov-Mar ARD release is minimal, except where taliks occur in some areas during some years²⁸ (e.g., Kutuk River; Dirk Nickisch, fixed-wing aircraft pilot at Coyote Air Services, oral communication, 2021)."
4. We cite relevant literature (L364-366): "One-year time lags between warming and aufeis formation in Arctic Alaskan rivers have been related to talik development and subsurface hydrology transport and connectivity³⁶." (Koch et al. 2024)
5. And we acknowledge other factors may play a role (L366-368): "Similar processes may occur with ARD, as well as buffering capacity of surrounding rocks, travel distance from subsurface to stream, and the mechanical frost fracturing of rock in the deepened active layer."

Overarching Comment 8: Avoid Qualitative Statements, Shorten Sentences

Reviewer: Please avoid qualitative statements such as 'well above' and recommend shortening sentences and avoiding split sentences.

Response: We have revised the manuscript throughout to: (a) Replace qualitative statements with quantitative text. For example: - "well above" replaced with specific values and comparisons - "abundant" replaced with percentages or concentrations (b) Shortened complex sentences. For example, the original L114-118 describing borehole drilling has been broken into shorter, clearer sentences in the Methods section. (c) Removed or restructured split sentences (those with information bracketed by commas). The revised text uses more direct sentence structures throughout. (d) Specific examples of revisions: - L299-302: Now reads simply: "Among the $n = 8$ acid seeps sampled upstream of Ikalukrok 207, Mn was the most abundant of 10 metals (Al, Mn, Fe, Zn, Ni, Cu, Co, Cd, As, Pb) in all but seep S8 (100 m downhill from Kuna Formation rocks²⁴) where Fe and Zn were most abundant (Fig. 5c-f)." - Statistical results are presented with exact values: $r = 0.96$, $R^2 = 0.93$, $p < 0.001$

Line-by-Line Comments

Abstract

L45-47: "trace metal diversity"

Reviewer: It is unclear what is meant by trace metal diversity - perhaps use 'type and concentration'. Also include Fe in 'stream flux' to read 'stream Fe flux'.

Response: The abstract has been revised. We no longer use "trace metal diversity." The revised abstract (L30-42) focuses on the key findings: dual ARD/FRM mechanisms, correlation with geology and permafrost, and the temporal lag. References to metal flux are now specific: "iron flux" (title), "iron loading" (L32), "iron influx" (L36).

Introduction

L50: "high latitude"

Reviewer: Suggestion to use 'high latitude' to encompass boreal and Arctic, otherwise state the importance of distinguishing between for the paper aim.

Response: The revised Introduction (L46-48) now uses broader framing: "One emerging threat—the widespread occurrence of metal precipitates discoloring formerly clear waterbodies in northern Canada², Alaska³⁻⁵, Russia (Supplementary Fig. 1), and temperate⁶⁻⁸ and tropical⁹ mountains—is visibly dramatic." We use "Arctic" when specifically referring to our study area and "high-latitude" or "northern" for broader context.

L50-51: Opening sentence too general

Reviewer: Recommend removing the opening sentence because it is too general. Have an opening sentence directly linking to the unforeseen large-scale increase in Fe concentrations.

Response: The revised Introduction now opens (L45-47): "Rapid warming is fundamentally altering Earth system processes and so threatening ecosystem functioning worldwide¹. One emerging threat—the widespread occurrence of metal precipitates discoloring formerly clear waterbodies..." This directly connects warming to the phenomenon of interest without excessive generality.

L56: Replace 'these'

Reviewer: Replace 'these' with 'air temperature and precipitation'.

Response: We have revised to be more specific throughout and made an effort to replace pronouns with unclear antecedents by explicit terms.

L58-60: Implies iron always released

Reviewer: This sentence implies that if liquid water is in contact with previously frozen ground, then iron will be released. Please include 'potential for' or consider 'exposes previously frozen iron-bearing minerals to present day environmental conditions'.

Response: Revised. The text (L51-54) now reads: "New pathways in thawing permafrost allow liquid water and its dissolved constituents to interact with previously isolated substrates, mobilizing iron (Fe) and other metals from terrestrial sources to aquatic sinks." The word "mobilizing" implies potential rather than certainty, and "previously isolated substrates" is more precise.

L68: Specify locations with metal-sulfide minerals

Reviewer: Suggest to state: 'in locations where soils, sediments and rocks contain metal-sulfide minerals'.

Response: Revised. The Introduction (L56-58) now states: "Two main processes facilitate Fe mobilization as ferrous iron (Fe(II)) following thaw: oxidation of iron-sulfide minerals (e.g., pyrite, FeS₂, the most abundant iron-sulfide) as acid rock drainage (ARD)^{2,4-11} and anaerobic microbial respiration as iron-reducing metabolism (FRM)^{3-5,12-17}." The geologic context is established clearly.

L71: Reverse sentence structure for FRM

Reviewer: Please reverse this sentence structure to begin with 'microbial iron reduction is a process common in...' and have a separate sentence stating evidence for the link between FRM and permafrost thaw.

Response: Revised. The FRM description (L90-96) now begins with the process: " Anaerobic metabolism by microbes in saturated environments (like wetland soils) initiate FRM, mobilizing Fe(II) in anoxic solution^{12,13}. In this process, heterotrophic microbes use Fe(III) as an electron acceptor during anaerobic respiration^{12,13}, producing Fe(II) as a byproduct¹²⁻¹⁷:" Evidence linking FRM to permafrost thaw follows (L111-114).

L73: Add references for FRM and permafrost thaw

Reviewer: Please include references to other studies that have demonstrated microbial iron reduction with permafrost thaw, including: Herndon et al., 2017; Patzner et al., 2020, Patzner et al., 2022.

Response: Thank you for that suggestion. We have added these references. The revised text (L111-114) cites: "Examples of FRM associated with ongoing deepening of active layers³ or with active layers deeper relative to nearby habitats (space-for-time substitution studies¹⁴⁻¹⁷) have been documented in high-latitude wetlands. Extensive areas of high-latitude permafrost consist of poorly draining, carbon-rich, reducing soils²²." with references to Herndon et al. (2017, 2020) and Patzner et al. (2020, 2022) in the reference list.

L76: Replace 'ultimate fate'

Reviewer: Please replace 'ultimate fate' with a specific outcome. Consider that environmental conditions upon thaw will determine whether Fe is precipitated as secondary minerals in soils or released into rivers.

Response: Revised. The text (L103-107) now describes the two fates: " After thaw, environmental conditions determine whether the Fe(II) produced by FRM is oxidized and precipitated as Fe(III)-bearing minerals within permafrost soils or is released into surface waters where it is oxidized and precipitated as Fe-(hydr)oxides^{16,17}. In oxygenated conditions, lithotrophic microbes can oxidize Fe(II) back to Fe(III) before Fe(II) is flushed from wetland soils¹²."

L77: Clarify 'thicker active layers'

Reviewer: Please provide detail on what you mean by 'thicker active layers' - deeper relative to neighboring soil or deeper relative to previous years?

Response: We now clarify this distinction. For temporal comparisons (L111): " Examples of FRM associated with ongoing deepening of active layers³" refers to year-over-year changes. For spatial comparisons (L11-112): " active layers deeper relative to nearby habitats (space-for-time substitution studies¹⁴⁻¹⁷)" explicitly labels this approach.

L80: State region size

Reviewer: Please state how large this region is.

Response: The revised Introduction (L122-124) now explicitly states all study area sizes: "At the regional scale ($2.41 \times 10^5 \text{ km}^2$), we focused on Alaska's Brooks Range mountains with its large-scale geology and permafrost conditions. At the subregional scale ($2.02 \times 10^3 \text{ km}^2$)... At the local scale ($2.06 \times 10^2 \text{ km}^2$)...."

L86: Link Salmon River to study areas

Reviewer: Please make it clear how this river is linked to the two areas you describe above.

Response: The revised text (L124-129) now clearly situates the Salmon River: ". At the subregional scale ($2.02 \times 10^3 \text{ km}^2$), we examined water samples collected in the basin of the Salmon River⁵ within the roadless Kobuk Valley National Park (KOVA) and adjacent watersheds in the context of watershed lithology²⁴ and near-surface (<1 m) permafrost probability²⁶." The Salmon River is explicitly identified as part of the subregional study.

L80-88: Permafrost boundary vs. biome boundary confusion

Reviewer: The distinction between the discontinuous-continuous permafrost boundary and the boreal-Arctic boundary may be a source of confusion. The reader has not been introduced to their importance.

Response: We have clarified this in the revised Introduction. The Methods section (L528-530) now explains: "Alaska's Brooks Range (<2,800 m asl) separates Arctic from interior climates, tundra from boreal ecosystems⁴⁶, and continuous (90-100% land area frozen) from discontinuous (50-90%) permafrost²²." The Introduction focuses on geology and permafrost zones as the key variables.

Results

L96: Separate results sections

Reviewer: Suggest having separate results sections for permafrost thaw and dissolved solids. Consider including bedrock/permafrost extent section first to set the scene.

Response: The Results section has been reorganized with clear subsections:

1. "Annual thermal loss reached its minimum in 2019" (L145-155)
2. "Rusting is associated with lithology and permafrost" (L157-230) includes regional and subregional spatial associations
3. "Water chemistry varies by waterbody type" (L231-286)
4. "Acid seep hydrogeochemistry varies by bedrock formation" (L288-304)
5. "Active layer depth anticipates ARD in streams by one year" (L306-330)
6. Geologic setting is in Methods (L607-643), providing detailed context.

L91: Word choice

Reviewer: Please remove the word 'observed'. Replace 'identified' with 'observed'.

Response: Revised throughout for consistent, precise word choice.

L91-95: Unclear how paragraph fits aim

Reviewer: It is unclear how this paragraph fits with the paper aim. I want to first know that you mapped sulfide-bearing mineral formations. Also unclear where this 'single stream' is relative to the 300 locations.

Response: The revised Results now leads with climatic data (L145-155) followed immediately by regional mapping (L157-181), establishing the 205 orange-colored waterbodies and their association with Zn-polygons and permafrost zones. The "single stream" (Ikalukrok Creek) is now clearly introduced in the local scale section (L288-330) with explicit geographic context ("5 km downstream of the boreholes", Figure 5a).

L98: Explain permafrost instability

Reviewer: Please explicitly state that permafrost instability was determined using a calculated stability index. 'Maximum instability' is stated without the reader knowing what is meant.

Response: The revised text (L146-150) now introduces the concept more clearly: "Permafrost stability is maintained through heat loss from the ground. A sufficient reduction in annual ground heat loss destabilizes permafrost by substantially deepening ALT and forming taliks. Heat loss is calculated as the balance between accumulated freezing (AFI) and thawing (ATI) air temperature indices as absolute values that provide an annual thermal loss index (AFI – ATI)." The index is defined before results are presented.

L112: 'Well beyond the influence'

Reviewer: Please state what is meant by 'well beyond the influence'? State the areal extent of mining and % contribution to fieldwork area.

Response: The revised Introduction (L132-136) now states “At the local scale ($2.06 \times 10^2 \text{ km}^2$), we analyzed water samples and ground temperatures in a landscape within the Ikalukrok Creek²⁷ watershed during a record warming period²⁸ (Fig. 1). There, Teck Alaska Incorporated (Anchorage, Alaska) drilled four boreholes within a single small catchment 11 km upstream of the Pb-Zn sulfide deposit Red Dog Mine (RDM).” And the revised Methods (L839-840) now states explicitly: “four boreholes along a 220 m transect (Fig. 5a, 377–394 m asl) in an upper catchment of Ikalukrok Creek, 11.2 km NW and 100 m above the mine’s roads and infrastructure” and “11 km upstream of the Pb-Zn sulfide deposit Red Dog Mine (RDM)” and (L846-847) “TDS was also measured at six other stations in the Ikalukrok Creek watershed upstream of RDM (Fig. 5a).” Figure 5a is a map showing all the mentioned locations and RDM.

L127: TDS composition

Reviewer: Please state the percentage of trace elements in TDS. Consider determining stoichiometry - do you see a relative increase in trace elements corresponding to a metal sulfide source?

Response: The revised text addresses this. Figure 5b shows that SO_4^{2-} comprises >85% of TDS in eight acid seeps ($R^2 = 0.999$ for SO_4^{2-} vs. TDS, L303). We also present molar SO_4^{2-} :Ca ratios (Figure 3b) showing values $\gg 1$ in acid seeps and stained tributaries, indicating sulfate from ARD (metal-sulfide source) rather than carbonate dissolution. The stoichiometric approach is used in Figure 3d showing SO_4^{2-} :Fe approaching theoretical ARD ratio of 2:1.

L135-140: Autumn observations vs. winter trapping hypothesis

Reviewer: Your observations of stream staining in Autumn seem to contradict your hypothesis that AMD products are trapped during winter. Further research is required regarding rate of freeze-up vs. transport rate.

Response: We have clarified this apparent contradiction (L348-368). Autumn staining does not contradict winter trapping - rather, autumn (Aug-Oct) is when that year's ALT reaches maximum depth, generating ARD that appears in streams before freeze-back seals the system. The one-year lag means the strongest ARD signal in autumn of year T reflects ALT from year T-1. We state (L353-355): “TDS averaged across monitoring stations during August-October was 1.2-3.4x TDS during May-July, indicating greater late than early thaw season release of ARD (Supplementary Fig. 11).” We acknowledge (L366-368) that rate of freeze-up vs. transport is an area for future research. Figure 9 shows correlations across 7 stations and 6 months of TDS with concurrent year ALT and previous year ALT.

L146-149: Remove statement of what dataset cannot do

Reviewer: Recommend removing text that states what the dataset has not been able to do. Instead summarize what part of the overall aim this section has contributed to.

Response: This text has been removed. The Results section now concludes each subsection with positive statements of what was found. For example, the local study section (L327-330) concludes: "Mean ALT of the previous year correlated strongly and significantly with Autumn pH at Ikalukrok 207 ($r = 0.96$, $n = 8$ year-pairs, $p < 0.001$) and with count of stained tributaries ($r = 0.83$, $n = 6$ year-pairs, $p = 0.02$)."

Figures**Figure 1: Simplify and link to roadmap**

Reviewer: Recommend simplifying by annotating maps in a way that matches the 'road map' at end of introduction - each part can be linked to one or more sections within the results.

Response: We have completely reorganized the figures:

Figure 1: Now a single panel showing the annual thermal loss index (1943-2023) as the climatic driver.

Figure 2: Regional and subregional study area maps with clear annotations linking to Results sections (panel a = regional OCWs, panels c-d = subregional acid seeps, panels e-f = dissolved Fe model).

Figure 5: Local study area (Ikalukrok Creek) with borehole locations clearly marked ("BHs").

Figures 6-9: Dedicated to the ALT-ARD relationship, a key finding.

Summary of Revisions

We have substantially restructured the manuscript in response to Reviewer 4's detailed comments with changes that include:

1. Clear "roadmap" in Introduction: Final paragraph now leads with aim, explicitly links three spatial scales to specific objectives (Overarching 1, 4)
2. Complete separation of Methods and Results: All methodological content moved to comprehensive Methods section (L512-906) (Overarching 4)
3. Results reorganized logically: Climate -> Geology/Permafrost -> Chemistry (ARD) -> Chemistry (FRM) → Temporal lag (Overarching 5)

4. Results state contributions, not limitations: Positive framing of findings as contributions to aim (Overarching 3)
5. Model validation added: R^2 values, comparison of modeled vs. measured temperatures, discussion of predictive capability (Overarching 6)
6. Winter trapping hypothesis justified: Supporting data (seasonal TDS patterns, aufeis observations) and literature cited (Overarching 7)
7. Qualitative language removed: "Well above" and similar phrases replaced with quantitative values throughout (Overarching 8)
8. Figures reorganized: Each figure linked to specific Results section and Introduction roadmap

Author responses

The reviewers required no changes:

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

I have now carefully evaluated the revised manuscript and the authors' detailed response to the reviewers. I am satisfied that my previous concerns have been thoroughly addressed.

The major claims—that (1) permafrost thaw exerts a predictive control on iron flux to Arctic rivers via two distinct pathways (wetland-derived Fe and sulphide-driven ARD), and (2) active layer thickness anticipates downstream ARD-related chemical signals by approximately one year—are now clearly articulated and more convincingly supported by the data and analyses.

The manuscript has been substantially improved: the restructuring of the Results and Discussion clarifies the narrative; the ALT–ARD relationship is now prominently presented; and the strengthened sulphate–TDS and stoichiometric evidence provide convincing support for ARD attribution. The statistical approaches and added detail on model calibration and data availability also address previous concerns regarding rigor and reproducibility.

I have no further substantive comments and consider the manuscript suitable for publication.

Reviewer #2 (Remarks to the Author):

The authors have done a good job revising the manuscript following my recommendations. No further comments from my side.

Review of COMMSENV-25-0214-T

February 2025

1 Reviewer Decision

This manuscript addresses an important and timely topic in light of ongoing climate change. The study has the potential to make a valuable contribution to the scientific community; however, several significant issues need to be addressed before it can be considered for publication. These issues are outlined in the major comments below. Therefore, the decision is to reject the manuscript, with an invitation to resubmit.

2 Major Comments

1. **The Relationship between ARD and Active Layer Thickness:** One of the key claims of the paper, presented in lines 40 to 42 of the abstract, states: “Here we use climatic, hydrological, and geologic contexts in northwest Alaska, together with modeled ground temperatures at the continuous-discontinuous permafrost boundary, to show ARD correlates with active layer thickness the previous year.” Given the importance of this finding, it is surprising that the relationship between modeled active layer thickness (ALT) and water pH is only presented as a subfigure within a subfigure (Fig. 2e). While space constraints are understandable, if this is to be a central result of the study, the data supporting this claim needs to be more prominently and clearly displayed.

Furthermore, it is concerning that the line of best fit is based solely on the average of all boreholes ($N = 8$ or $N = 9$ — the exact number is difficult to determine given the figure’s size) and the average of pH data from August to October. Providing data from individual boreholes, rather than averaging, would allow readers to assess site-specific relationships and variability. This would not preclude the presentation of an overall trend line on the same figure, but it would add depth and transparency to the analysis.

In attempting to reproduce these relationships, the following statement was noted in the supplementary data: “All modeled ground temperatures used the online GIPL2. Rather than provide the output files, we provide the input files which can be applied to replicate the modeled ground temperature values.” It is essential that both input and output files be provided. Reviewers and readers should not be required to reprocess critical data underpinning the study’s conclusions — that choice should remain optional for those who wish to explore the modeling independently.

Finally, to substantiate the assertion that this is a relationship between ARD and active layer thickness — rather than simply between water pH and ALT — the pH should be contextualized alongside other reaction products. Given that ARD releases sulfate ions into water, as stated in lines 62 to 66, a plot of sulfate concentration against the previous year’s ALT would strengthen this key finding and provide more robust evidence for the proposed mechanism.

2. **Definition of Dissolved Iron:** On lines 169–170, dissolved iron is operationally defined as material that passes through a $0.45\ \mu\text{m}$ filter. It is then stated that “The strong correlation of Fe_{SUS} with turbidity ($r = 0.77$, $n = 42$) in the Salmon River and its tributaries (Extended Data Fig. 4) implies that suspended nanoparticles of Fe(hydr)oxide color the Salmon River and its stained tributaries orange.” However, this statement is inaccurate. Nanoparticles are typically defined as particles between 1 and 100 nm in diameter, while the $0.45\ \mu\text{m}$ ($450\ \text{nm}$) filter used in this study would allow nanoparticulate iron to pass through. Therefore, the observed relationship is actually between iron particles larger than 450 nm and turbidity.

To maintain accuracy and consistency with the literature, the definitions of dissolved, nanoparticulate, and suspended iron phases need to be revised. Several conclusions and statements in the manuscript also require adjustment to reflect this distinction correctly.

3. **Restructuring of the Manuscript to Avoid Methods and Discussion Material in the Results Section:** The readability and clarity of the manuscript are hindered by the presence of extensive methods and discussion content within the results section. Moving this material to the appropriate sections would greatly improve the manuscript’s structure and flow.

Examples of methods appearing in the results section include:

- Lines 112 to 122, in the section titled “Dissolved solids and warming-induced permafrost thaw”, which consist entirely of methodological description without presenting any results.
- The section “Extensive sampling in the wilderness”, which largely describes fieldwork logistics rather than findings.
- Lines 98 to 105, which detail how daily measured air temperatures were used to calculate temperature-related parameters (freezing and thawing indices, thermal balance) — material that clearly belongs in the methods section.

Similarly, several instances of discussion appear in the results section:

- Lines 135 to 137, where it is stated: “Because ALT and talik formations reach their maximum dimensions in Sep and Oct, we speculate that, unless sub-aerial taliks[4] persist to the ground surface, ARD produced by substrates newly exposed to water is trapped over winter at depth, surfacing the following thaw season if ALT is sufficiently deep to allow its release.” This is a key claim of the study (as noted in lines 42 to 45 of the abstract) and is clearly interpretive, as signaled by the phrase “we speculate”.
- Lines 143 to 149, which include statements such as “The Teck dataset provides compelling evidence of causal pathways leading from climatic drivers to ARD in a known metal-sulfide-containing formation.” This level of interpretation belongs more appropriately in the discussion section.

A clearer distinction between methods, results, and discussion would enhance the overall structure and readability of the manuscript, making it easier for readers to follow the narrative and evaluate the study’s findings.

3 Minor Comments

3.1 Title

1. **Title Similarity:** The title is highly similar to the O’Donnell et al. 2024 publication, which is extensively referenced throughout the manuscript. The current title does not fully reflect the novelty or findings of the study. Please consider rephrasing it to better capture the unique contributions of this work.

3.2 Abstract

1. **Attribution of pH to ARD:** The correlation between acid-rock drainage (ARD) and pH is mentioned. However, further work is needed to demonstrate that the pH is driven specifically by ARD, rather than other sources of acidity. This needs to be addressed before making the claim. Please refer to Major Comment 1 for further clarification.

3.3 Introduction

1. The introduction provides an essential overview of the topic, as well as highlighting the novelty of the study in the final paragraph.

3.4 Results

1. **Splitting up the Results Section by Spatial Division:** As introduced in the final paragraph of the introduction, there is a spatial division of the results: one set of results from time-series monitoring of borehole studies and another set from repeated spot-sampling campaigns in the national park. Clearly attributing these spatial divisions in the results section, and linking it to the final paragraph of the introduction, would increase the readability of the manuscript. This has been partially attempted by the inclusion of the section “Extensive sampling in wilderness”; however, this section detracts from the results by introducing a paragraph on methods without presenting any results.

3.5 Discussion

1. **Extension of Discussion following Reorganization of Results:** A large portion of the discussion is currently dedicated to speculative content (e.g., potential associations of marine magnetite with permafrost melting during the medieval warm period, despite the iron-oxyhydroxide composition not being well known; and declines in fish populations). While such speculations are valid, the discussion section would benefit from more direct analysis of the results presented in the manuscript. For example, the first paragraph of the discussion reads more like a conclusion than a discussion of the study’s findings.

3.6 Figures

1. **General Comment:** It is difficult to discern the key novelty of the study from the current manuscript figures due to the large number of subfigures and their interrelationships. Please consider reducing the number of subfigures by moving some of them to the supplementary material, and referencing them as necessary. One figure showing the study location and one figure per key result would be more appropriate and clearer.
2. **Figure 1:** This figure acts both as a map to show the study areas and as a results figure displaying the relationship between seep count and length in a separate subfigure. This dual purpose is confusing for the reader. Please consider creating one figure to show the two field areas (1a, 1b), linked to the results section, and including the borehole locations (their location in supplementary Figure 3 is unclear). Figure 3c should be a separate figure. If there is insufficient space for this subfigure in the manuscript, it may suggest that there are too many results to convey a clear message. The results from this part of the study occupy only a single sentence: “Over six years, we also observed variability both within and between years in the rusting of a single stream and the number of its tributaries that were stained orange (Fig. 1c).” Figure 1d does not need to be included and should be removed, as it is not part of the study area and merely states that this is an Arctic-wide phenomenon, which can be referenced instead.
3. **Figure 2:** This figure hides one of the key findings of the study in subfigure 2e. Please consider removing some subfigures to make this result more prominent.
4. **Figure 3:** A comparison of modeled versus measured temperature would be helpful if this is a key result of the manuscript. This would not detract from the other finding that the type of vegetation impacts ALT.

References

Jonathan A. O'Donnell, Michael P. Carey, Joshua C. Koch, Carson Baughman, Kenneth Hill, Christian E. Zimmerman, Patrick F. Sullivan, Roman Dial, Timothy Lyons, David J. Cooper, and Brett A. Poulin. Metal mobilization from thawing permafrost to aquatic ecosystems is driving rusting of Arctic streams. *Communications Earth & Environment*, 5(1):1–10, May 2024. ISSN 2662-4435. doi: 10.1038/s43247-024-01446-z. URL <https://www.nature.com/articles/s43247-024-01446-z>. Publisher: Nature Publishing Group.

Review of manuscript COMMSENV-25-0214-T “Thawing permafrost stains streambeds and rusts rivers in Arctic Alaska” by Dial et al.

Overview

In this article, the authors use a combination of a very large set of field data and modelling approaches to discuss sources of input of metals (and sulfate, etc.) into streams and rivers (the occurrence of “orange streams and acid seeps”) in northwest Alaska. Specifically, they consider the contributions of pyrite weathering (leading to acidification and Fe/sulfate release) and microbial iron mineral reduction and dissolution. At the end of their manuscript (discussion section), the authors very briefly discuss the relevance of these processes for water quality (use as drinking water, fishing, etc.).

Rusting streams and rivers in permafrost areas have been discussed in several publications in the last few years, in particular their relevance for organic carbon mobilization leading to increased greenhouse gas formation and emission. Therefore, this publication is timely and relevant and will be of interest to the readership of Communications Earth & Environment.

The data provided in the paper are convincing although I have to note that I am not an expert in the modelling approaches that have been used. My expertise is more the biogeochemistry part and is mostly well described (I added a few questions and suggestions below). Overall, my impression is that the data is clearly presented and carefully analyzed and interpreted. In my opinion, the conclusions drawn from the data are reliable. The discussion at the end of the manuscript can be improved.

Below, I provide some general comments and specific line-by-line suggestions for revising the manuscript and I hope that these are helpful to the authors.

General comments and questions

1. Considering the very large (fantastic!) dataset the authors have, the discussion section is relatively short and a bit disappointing. The main conclusion is that “Its potential for harm through toxic effects...remains uncertain” (lines 299-301). Maybe the authors can expand here and discuss their data a bit more, also in the context of other publications? Are these processes expected to take place in other parts of the world? What are the consequences for trace metal transport into oceans (similar to Rio Tinto in Spain where a lot of natural pyrite weathering occurs)? What are the expected consequences for organic carbon mobilization and greenhouse gas emission? These are just some ideas that came into my mind.
2. Also regarding the discussion and interpretation of the data: can the authors (in the discussion) evaluate more specifically the relative importance of pyrite weathering versus microbial Fe(III) mineral reduction and dissolution at the two field sites?
3. Finally, do the authors have any data on dissolved organic carbon in the water samples that they analyzed? If yes, can they discuss the importance of their processes (pyrite weathering, iron mineral reduction/dissolution) for biogeochemical carbon cycling?

Specific comments

1. L38: “...thickness of the previous year”?

2. L40-42: in the abstract it is not clear for the reader what data exactly has been used by the authors. Here they say “we use climatic, hydrological and geological context” – what exactly does this mean? Please clarify.
3. L44: introduce abbreviation ALT first.
4. L66: I recommend calling these precipitates “Fe(III) (oxyhydr)oxides” because this include oxyhydroxides such as ferrihydrite or goethite but also oxides such as hematite or magnetite.
5. L73: please add the information that this is Mn(IV) (e.g. MnO₂).
6. L128: “2.7 m”.
7. L156: “<1 m”.
8. L163-164: did the authors also distinguish Fe(II) and Fe(III)? Would be interesting to see how much Fe is actually present in the reduced state or in the oxidized state, complexed by OM. If they don’t have it, maybe this is something they want to consider in the future.
9. L172: “Fe(III) (oxyhydr)oxides”.
10. Line 279 (and in general): the authors are referring to “metal-reducing” microbes. Wouldn’t it be more accurate to say “iron(III)-reducing microbes”? Or do the authors have evidence that other metals are reduced (and mobilized by reduction) as well? How about manganese? Any other metals?
11. Lines 291-292: “....can form from microbial reduction of Fe(III) (oxyhydr)oxides by anaerobic microorganisms”.
12. Line 292: wouldn’t it make sense to replace reference 28 by a reference that shows magnetite in soils, not in marine sediments?
13. Line 294: “Fe(III) (oxyhydr)oxides”.
14. L579: “Fe and Al (oxyhydr)oxides”.
15. L604: “30 m”.

Figures:

16. Figure 2, caption: “2.7 m” (space missing).
17. Figure 4, caption: “1-2 m” and “<1 m”. (space missing).
18. Figure 5: why are the metals in panel c called “Biogenic metals”? In the main text, this term is not used. Also, Fe(II) can be produced by chemical reduction of Fe(III) by sulfide (that means Fe(II)_{diss} can also be chemical).

Review of *Thawing permafrost stains streambeds and rusts rivers in Arctic Alaska*

Dial et al., present multiple lines of evidence to show that permafrost thaw in regions of metal-sulfide substrate and/or wetlands contributes to increasing iron concentrations in high latitude aquatic bodies. This is a highly relevant and timely study that would have large readership. I suggest that this manuscript needs to be restructured to communicate the evidence to the reader. I also suggest that the manuscript acknowledges the limits of what can be answered to date – the evidence motivates many exciting avenues for future work. I recommend that this paper is not accepted in its current format. Please find my comments below:

My comments are mainly regarding manuscript structure and communicating the multiple field locations and data types in a narrative that links back to the paper aim and that can be read and understood by a large, multi-disciplinary readership. The comments mainly focus on the Introduction and Results.

Overarching comments:

1. Final paragraph in your Introduction: suggestion to start the paragraph with the overall aim, which I currently read to be the last sentence of the paragraph. And then link the steps outlined (the two areas and the river flux calculation) to this aim. Suggest avoiding the word ‘importantly’ just for the final sentence as it doesn’t do justice to the other sentences in the paragraph.
2. Results: suggest combining results and discussion section allowing the reader to know the implication for a finding for the paper aim. For example, in Line 121- Line 122, you state that vegetation is controlling snowpack which is controlling active layer thickness. You could then state why this is relevant to the aim of your paper – i.e., does it demonstrate that vegetation index would need to be included in a process-based model that estimates where iron is released to rivers? Or if the discussion is kept separate then please make sure to link back to the results and then link results and discussion back to the road map outlined in the introduction and the aim of the paper.
3. Results (Line 97 – 149): currently the end of this section outlines its limitations, but the section itself is a results section, therefore I recommend stating the results as one part of the jigsaw puzzle that contributes to the aim outlined in the Introduction, rather than stating their limitation to the overall aim – other sources of evidence come together for that.
4. Results: recommend moving method-themed paragraphs, such as the wilderness sampling and sample filtration strategy, to a longer introduction section or to the Methods. These are key field and lab methods but could be stated underneath the steps outlined in the ‘road map’ at the end of your introduction. Currently, some of the paragraphs in Results are allocated to methods and some to results. Building in the Methods to the approach part of the end of introduction could be a way forward here, linking back to the need for a clearer ‘road map’ at the start of this paper. At the end of the introduction, I would also recommend a clearer outline of what field sites are used to address what objective to deliver the overall aim.
5. Results: a suggestion to re-order/title the sections and include the lithology section (Line 210-222) upfront, as logically the rocks were there prior to the permafrost formation – so one logic would be to order data bedrock, permafrost extent, climate influence on permafrost extent, geochemistry (AMD), geochemistry (FRM), relationship between geochemistry and discharge.
6. Results and Discussion: I do not write as a modelling expert, but from an interested, still learning, reader of this paper, it would be unclear to me how successful your modelling approach is. I suggest including an analysis of the predictive modelling in the discussion section – to inform future work in the community. E.g., do modelled estimates match present day values? What additional information is needed to build on this work.

7. Results/discussion: please justify with literature and/or your own data, the proposed theory that AMD products are locked away in soils during winter months or remove temporality from the paper results and include it as a motivator for future work.
8. Please avoid qualitative statements such as ‘well above’ and I recommend shortening sentences and avoid split sentences because it is harder for the reader to follow.

List of line-by-line edits:

Abstract

Line 45 – 47: It is unclear what is meant by trace metal diversity – perhaps use ‘type and concentration’.

Line 45 – 47: Please include Fe in ‘stream flux’ to read ‘stream Fe flux’.

Introduction

Line 50: Suggestion to use ‘high latitude’ to encompass boreal and Arctic, otherwise state the importance of distinguishing between for the paper aim.

Line 50-51: I would recommend removing the opening sentence because it is too general. I would have an opening sentence directly linking to the unforeseen large-scale increase in Fe concentrations in high latitude rivers (O'Donnell et al., 2024).

Line 56: Replace ‘these’ with air temperature and precipitation’

Line 58 – 60: This sentence implies that if liquid water is in contact with previously frozen ground, then iron will be released. Please include the words ‘potential for’ and consider a sentence such as ‘exposes previously frozen iron-bearing minerals to present day environmental conditions’, ‘unveils a new source of iron-bearing minerals’.

Line 68: Suggest to state: ‘in locations where soils, sediments and rocks contain metal-sulfide minerals.

Line 71: Please reverse this sentence structure to begin the sentence with ‘microbial iron reduction is a process common in...’ and have a separate sentence that stating evidence for the link between FRM and permafrost thaw.

Line 73: Please include references to other studies that have demonstrated microbial iron reduction with permafrost thaw, including: Herndon et al., 2017; Patzner et al., 2020, Patzner et al., 2022

Line 76: Please replace ‘ultimate fate’ with a specific e.g., whether Fe-bearing minerals dissolved in permafrost soils. Also consider, that environmental conditions upon thaw will determine whether or not the Fe is precipitated as secondary Fe-bearing minerals in the permafrost soils or released into the rivers.

Line 77: Please provide detail on what you mean by ‘thicker active layers’ – it can either be read as deeper active layer relative to a neighbouring soil or deeper active layer relative to previous years.

Line 80: Please state how large this region is

Line 86: Please make it clear how this river is linked to the two areas you describe above in this paragraph

Line 88: A suggestion is to start the paragraph with the overall aim, which I currently read to be the last sentence of the paragraph. Or perhaps it is not stated. Then link the steps outlined (the two areas and the river flux calculation and modelling) to this aim. Suggest avoiding the word ‘importantly’ just for the final sentence as it doesn’t do justice to the other sentences in the paragraph, and is not discussed in the discussion section to the extent that justifies its importance.

Line 80 – 88: The distinction between the discontinuous – continuous permafrost boundary and the boreal – Arctic boundary may be a source of confusion to readers. I understand what the boundaries are, but the reader has not been introduced to their importance for your study at this stage of the paper.

Results

Line 96: Suggest having a separate results section for permafrost thaw and section for the dissolved solids. Consider including a section on substrate/bedrock composition and permafrost extent, prior to this – to set the scene, prior to discussing thaw and river geochemistry. The pairing of substrate composition with permafrost extent and geochemistry is a key novelty of the paper for the discussion section, and it would be good to include geology and permafrost upfront in the results.

Line 91: Please remove the word ‘observed’. Replace ‘identified’ with ‘observed’.

Line 91 – 95: It is unclear how this paragraph fits with the paper aim and results lay-out. I read it and want to first know that you mapped the sulfide bearing mineral formations. Their distribution in relation to permafrost extent and thaw, is an interesting result to state. I am also unsure where this ‘single stream’ is relative to the 300 locations.

Line 98: Please explicitly state that the permafrost instability was determine using a calculated stability index. At present, the ‘maximum instability’ is stated upfront in the paragraph without the reader knowing what is meant by ‘instability’.

Line 105: Suggest avoiding the split-sentence i.e., when you include additional information bracketed by two commas, because it can be hard for a reader to follow.

Line 112: Please state what is meant by ‘well beyond the influence’? – please state the areal extent of mining and the % contribution of the mining area to your area of fieldwork.

Line 114 – 118: Please break this sentence into shorter sentences: *In response to 2018 115 stream rusting unrelated to mineral exploration or development, Teck drilled four boreholes 116 (Extended Data Table 1) to record ground temperatures beneath two plant community types 117 common across the Arctic (sedge and shrub tundra) in an undisturbed valley 11 km north and 118 100 m above all mine infrastructure (Supplementary Fig. 3).*

Line 118 – 121: Please reverse this sentence to begin with the aim e.g., ‘To model ground temperatures in the 200 km² area over time, we used...’: *Using drill-logs, soil properties, and weather records, we calibrated the Geophysical Institute Permafrost Lab model[17] 119 (parameters in 120 Extended Data Table 1), then modeled 2010–2023 ground temperatures (MGT) to estimate active layer thickness (ALT).*

Line 127: please remove ‘abundant components’ and state the percentage of trace elements (Fe, Zn, Al etc) in the TDS. Also note that TDS will increase with weathering therefore an increase in trace metals may correspond with an increase in major cations, such that the proportional contribution of Fe, Zn, Al to the TDS remains the same. Consider determining the stoichiometry of the element composition / or use element ratios, do you see a relative increase in the trace elements corresponding to a metal sulfide source?

Line 125- 133: Please state why these findings are significant for the paper (or at least the discussion section) aim.

Line 128-129: Please remove ‘well above’ and provide quantitative text.

Line 135- 140: Please correct me if I misunderstood, but I see that your observations of stream staining in Autumn contradict your hypothesis that AMD products are trapped in soils during winter months. I agree that a portion will be trapped over winter because e.g., the pH and Eh of soils promoted secondary

iron mineral formation, rather than the release of Fe from porewaters into the river, however you observe a portion of the AMD to get into the rivers in Autumn. Further research is required regarding the rate of Active Layer freeze-up vs the rate of transport of AMD products from the soil. I recommend to state what more needs to be done to determine the temporality of when AMD products are transferred from soil to river.

Line 139-141: I am unclear how this final sentence is consistent with your observations, or what observations you are referring to.

Line 146-149: Recommend removing: *this dataset does not distinguish 147 FRM sources, correlate Fe release to permafrost or lithology, nor estimate Fe flux in rivers and 148 streams*. Because it states what a data set has not been able to do, rather than summarising what part of the overall aim this paper section has contributed to.

Line 189-192: Please make this into multiple sentences. It is hard to follow.

Line 194-195: Please state what you mean by *Iron in stained tributaries differed little with discharge*.

Figures:

Figure 1: To help the reader link between paper aim, field sites, data collected at field sites, I recommend simplifying by annotating these maps in a way that matches the ‘road map’ proposed at the end of the introduction – each part of the map/field location can be linked to one (or more) sections within the results.