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Answer to editor

Pb and Fe flow through the mire-lake complex of Skogaryd catchment - a system under anthropogenic influence

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Editor decision: Publish subject to minor revisions (review by editor)

Editor comments:

The authors have satisfactorily addressed most of the 3 reviewer comments, although there is some variability in the completeness of responses. The Response to Reviewer 1 includes the revised text and its location, but this was not necessarily the case for the responses to Reviewers 2 and 3. Please see below where further clarification is desirable – the list may not be complete. There were additional minor errors introduced or retained in the revised version.

In general

Typos, grammar and word choice have been corrected/changed a few places.

Response to reviewer 1.

Question 1.3: The revised text addresses the reviewer comment, but the response to reviewer text is slightly different than the actual revised text.

Answer

The response to the reviewer and what has been written in the manuscript is now identical:

L329: Peat Fe concentrations at the top of the mire were between 606 and 1237 mg kg⁻¹ and were measured to between 1434 and 1474 mg kg⁻¹ in 120cm depth (Fig 4, Table S2).

Question 1.4: The authors address the query, but is the answer also explained in the text?

If so, please indicate where to find it.

Answer

Following have been added to the answer to reviewer 1: The motivation for including the lake measurements was to touch upon the environmental consequences of the metals leaving Mycklemossen.

The following is found in the manuscript L103: A large proportion of pollution derived Pb deposited in Swedish peatlands will eventually end up in downstream ecosystems such as lake sediments (Brännvall et al., 2001), where it will accumulate and potentially reach harmful concentrations for benthic living organisms.

Also, the inclusion of the lake measurements was necessary for the mixing model that was used to determine the fate of the anthropogenic Pb that leaves Mycklemossen mire, as stated in the discussion from L565 to L573.

Response to Reviewer 2

Question 2.1: The answer provided in the Response is well-written and clearer than the revised text in section 4.2. Which part of the revised text specifically addresses this question? Please also clarify what is meant by the term “hydrological connectivity”, since it is a key explanatory factor in this manuscript but is not defined.

Answer

The revised text about the underlying chemical mechanisms and whether the observed Fe-DOC co-variation results primarily from colloidal co-transport, redox-driven Fe-organic complexation or other processes is discussed from L445 to L459. The asynchronous peak is discussed in L460 to L479.

Based on editor’s comment, we have used more of the text from the response instead of what was written in the manuscript to improve the clarity and language of the manuscript.

The revised text in the manuscript is very similar but not identical to the text-response for reviewer 2.

The term hydrological connectivity is now defined in L105: Several key processes exhibit control on metal export from peatlands, such as pH and the different metal’s binding affinity to DOC, but hydrological connectivity that refers to the transport of water from peat to stream, and seasonality, which includes drought and precipitation events seems to be most important (Broder & Biester, 2015, 2017; Prijac et al., 2023)

Question 2.3: The answer provided in the Response here is also clear. Please specify the corresponding lines/text in the manuscript that provide this answer.

Answer

The geochemical associations of Pb and assessment of Pb mobility and ecological risk is discussed from L496 to L509. The specific text is included in the response to reviewer 2.

Question 2.4. Please specify text and lines in the manuscript where it is discussed.

Answer

How traits related to *Sphagnum* might govern the tight Fe–DOC correlation and the substantial Pb retention observed in this system can be found in the introduction from L122

and in the discussion from L448. The specific text has been included in the response to reviewer 2.

Question 3.4. This comment belongs to the reviewer 3 response.

Response to reviewer 3

Answer

It has been moved to the section for reviewer 3

Question 3.2. “These known drivers... also discussed in the manuscript”. – Where?

Answer

These known drivers are almost certainly also key processes in our mire-lake complex and is now better introduced from L106 to L116. The specific text has been included in the response to reviewer 2.

Question 3.3. It is not too important, but I see that the methods still include Hg, As, Cd, although it was said to be omitted.

Answer

It has been removed from L223 in the methods.

New sentence: The concentration of Pb that is considered one of the highest ranked heavy metals to consider for public health (Tchounwou et al., 2012) and Fe were quantified in technical triplicates of the peat samples at 10-15cm, 15-20cm, 20-25cm, 120-125cm and 420-425 cm depth with inductively coupled plasma-mass spectrometry (ICP-MS) (iCAP, ThermoScientific) (SI Table 2).

Question 3.4. “will clarify this better in the text”. Where?

The general answer to reviewer 3 has been modified to the following:

The catchment areas have been generated based on detailed topographic maps in relation to the location of the measuring stations (out-flow) of the mire and the lake Erssjön. Thus, all the water that are measured at these locations are from these two areas. We do not think we need to add more maps to the manuscript, but this is now better clarified in the text. Specifically, in the methods from L164 to L171 and from L271 to L280.

Specific line numbers have been provided for the sub-questions.

Question 3.8. Please specify where the new text can be found, especially the “Pb concentrations for water streams will be better explained... in the next version of the manuscript...”

Answer

The question regarding whether Pb is bioavailable to microorganisms is discussed from L496 to L509, with some attention to where Pb is located.

Also, the Pb concentrations for water streams in relation to toxicity and bioavailability is discussed from L523 to 535.

The specific text is included in the response to reviewer 3.

OTHER

Other minor edits to consider (Line numbers refer to manuscript v2):

Line 50: “in different forms” instead of “on different forms” **Corrected**

Line 53 : “changes ...are” instead of “changes... is” **Corrected**

Line 59: “Sphagnum-derived” **Corrected**

Line 80 “Fe’s ability to redox cycle” – is this the ability to cycle between different redox states?

Answer: Modified L80: These reactions depend on Fe’s ability to cycle between redox states

Line 107: “seem” instead of “seems” **Corrected**

Line 117 “such as Fe” instead of “as Fe”? **Yes, corrected**

Line 117: “redox conditions that occur” instead of “redox conditions as occur” **Yes, corrected**

Line 119: “are commonly” instead of “is commonly” **Corrected**

Line 138: “surmount to ” should be either “amount to or exceed” ? **Corrected to “amount”**

Line 160: “intermediate” can be in quotes here. **Corrected**

Line 443: “likely representative for the rest of Sweden” is probably not what you mean here.

Answer

What we meant is: Changed to “likely represents similar ecosystems in Sweden”.

Line 447: Despite **Corrected**

Line 448: “2018” instead of “of2018” **Corrected**

Line 458 “Notably” instead of “noteably” **Corrected**

List of substantial changes

In addition to the changes marked in the manuscript with “track changes”, please find the list below, showing an overview of relevant changes as requested together with Author’s response.

- The introduction has been amended to accommodate the general comments from all reviewers. In particular, the focus has been to present the study objectives in a more logic structure. In addition, the last paragraph of the introduction has been elaborated and now more clearly states main study objectives and expectations of the study.
- All paragraphs in the discussion have been amended, and now all include a deeper analysis as requested by the reviewers and associate editor. Emphasis has been on deeper discussion of the controlling mechanisms behind DOC, Fe and Pb export.
- New SI table (table S1)
- New title for section 4.1
- Separation of section 4.4 into 4.4: Pb pollution status of stream water and 4.5: Export and fate of Pb in the mire-lake complex.
- Re-calculation of the mixing model

Reviewer 1

Question 1.1

The study objectives are not entirely clear upon reading the manuscript. The research questions are introduced without a clear structure, and the final paragraph of the introduction provides only a very general statement of the aim of the study. I recommend that the authors present the study objectives more explicitly and potentially include specific hypotheses/predictions in the introduction to better guide the reader.

Answer

The first paragraph in the introduction has been amended to better introduce the overall research question of the study. Changes include more context of the relationship between export of DOC and metals and why that is important in peatlands containing heavy metals.

The last paragraph of the introduction has been rewritten. The study objectives are now more elaborated, and we have put up expectations for the study to better guide the reader.

Question 1.2

The manuscript lacks clear definitions of the topography types and information regarding their spatial scale. It is specified that the topography types were sampled 1-2 m apart, but the typical size or extent of these structures is not described. I furthermore lack a discussion of the observed differences in the physical and chemical characteristics among different topography types.

Answer

Section 2.1 in the manuscript “Site description”, has been amended, and now the below text is included:

Mycklemossen is composed of a mosaic of hummocks and hollows with transitions zones, called intermediate in our study, but mostly dominated by hummock (Rinne et al., 2022). The hummocks are mainly dominated by *Eriophorum vaginatum*, *Calluna vulgaris* and *Erica tetralix* and the hollows consists of different *Sphagnum* species, mainly *S. rubellum*, *S. fallax* and *S. austinii* as well as *Rhyncospora alba*, and the peat samples were sampled approximately within a square meter plots (Kelly et al., 2021).

The observed differences in SOM and N% (the only significantly different chemical parameters) are now included in the discussion, section 4.3, in relation to decomposition.

Question 1.3

The depth resolution of the measured variables, especially for the metals, is coarse. Consequently, statements such as that on line 292; “Peat Fe concentrations at the top of the mire were between 606 and 1237 mg/kg and barely changed until below 120” are problematic, as no data are available for the interval between 50 and 120 cm depth. This limitation should be acknowledged in the manuscript.

Answer

Yes, there will be variation in concentration of metals in the peat cores we do not see with our depth resolution (quite large std. on concentrations, SI table 2), and we acknowledge this. The text has been rephrased to a more neutral language.

L329: Peat Fe concentrations at the top of the mire were between 606 and 1237 mg kg⁻¹ and were measured to between 1434 and 1474 mg kg⁻¹ in 120cm depth (Fig 4, Table S2).

Question 1.4

The introduction currently lacks a clear motivation for including the lake measurements. The objectives stated at the end of the introduction are rather general and focus solely on the mire.

Interestingly, the substantially higher export of C and Fe from the lake compared to the inflow to the lake points to other sources than the mire. This observation could be explored further in the manuscript, and the relative contribution of the mire to the overall hydrological inflow to the lake could be clarified if this data is available.

Answer

The motivation for including the lake measurements was to touch upon the environmental consequences of the metals leaving Mycklemossen. It is explained in: L103 A large proportion of pollution derived Pb deposited in Swedish peatlands will eventually end up in downstream ecosystems such as lake sediments (Brännvall et al., 2001), where it will accumulate and potentially reach harmful concentrations for benthic living organisms. Also, the inclusion of the lake measurements was necessary for the mixing model that was used to determine the fate of the anthropogenic Pb that leaves Mycklemossen mire, as stated from L565 to L573.

General answer

The lake has more than twice as large catchment area than the mire and also the discharge is substantially higher. Hence, a lot of water is coming from the forested land around the lake, and this can be expected to be high in DOC (maybe also Fe, as there are e.g. Podzols in the area). That means the lake receives a lot of DOC and Fe from the forested land, but not Pb. This corresponds with the low Pb in all other streams in the catchment (mainly forest dominated), where the mire-streams are the exception.

The relative DOC and Fe export from station 1 relative to station 6 follows the ratio of catchment area, see table below (question 2.2).

Question 1.5 – technical comments

- Line 54: Replace “binds” with “bind”. **Done**
- Line 57: Replace “peatland” with “peatlands”. **Done**
- Line 68: Remove “and” before “can be traced...”. **Done**
- Line 337: Remove “at” before “from Mycklemossen”. **Done**
- Line 402: Incomplete sentence starting with “The strong correlation...” **Done**
- Line 407: I suggest adding “The year of” or something similar before 2017 to avoid beginning the sentence with a number. **Done**

- Line 270: I cannot see that the change in N with depth was more extreme for hummock compared to intermediate and hollow. This is not obvious looking at Fig. 3. Should it be the other way around?

Answer

Yes, it should be the other way around, and “more extreme” has been changed to “less extreme”. N content increases in a somewhat linear manner with depth in hummock, while for intermediate and hollow, in particular for hollow, N content decreases from the top of the mire to 200 cm depth, from where it increases to 400 cm depth.

- Line 286: Intermediate generally had the highest Pb content, although the largest concentration was found in hummock at 25-50 cm (Table S1).

Yes, that is correct.

New sentence: Intermediate generally had highest Pb content that was more than twice as high as for hollows, while the highest Pb content was measured in hummock at 25-50 cm depth

- Line 288 – 290: Make sure that the correct numbers are presented here. According to Table S1, intermediate has the Pb content of 64.25 mg/kg, and hollow that of 32.21 mg/kg, and not the other way around. Pb contents of 4.41 and 0.05 mg/kg in the 25-50 cm interval do not match with the data in Table S1, nor with Fig. 4. This has been corrected
New sentence: In intermediate and hollow topographies Pb content was highest in 15-20 cm: 64.25 and 32.21 mg kg⁻¹ and decreased to 58.5 and 14.89 mg kg⁻¹ at 25-50 cm depth, respectively (p = 0.02, Fig 4, Table S7).

- Line 319: It is not clear why data points for Fe and Pb were removed when discharge was low? It would have been informative to include this data.

New sentence: Data points for Fe and Pb were removed when discharge <0.0001 (m³ s⁻¹), which occurred during summer when the water level was too low to measure discharge, making calculation of export impossible.

Question 1.6 and 1.7

Line 390: Please elaborate on what type of interaction with Fe that stabilizes peat. Also in the same sentence, that most Fe in Mycklemossen is placed in deep anoxic peat layers does not rule out that this stabilizing effect of Fe on C is important.

Line 392: What is the “C destabilizing mechanism of Fe”. Please clarify.

Answer

Following text has been included in the discussion from L429, to clarify both the peat stabilising and destabilising effect of Fe.

Static oxic conditions during summer could also stabilise peat OM and DOC through adsorption and complexation interactions with Fe (Chen et al., 2020; Riedel et al., 2013; Song et al., 2022), though most Fe in Mycklemossen is placed in deep anoxic peat layers. However, regardless of redox regime, the majority of total Fe in peat will interact with OM, and Fe-OM complexes are formed in both oxic and anoxic peat (Bhattacharyya et al., 2018). Thus, the stabilising effect of Fe might not be limited to the oxic layer. But under these aerobic conditions, oxidative reactions catalysed by Fe can lead to production of hydroxyl radicals that can promote degradation of peat (Qin et al., 2022; Trusiak et al., 2018) (Qin et al., 2022; Trusiak et al., 2018). Such reactions might be driven by small concentrations of Fe (between 280 - 2.300 mg Kg⁻¹ peat; Curtinrich et al., 2024), which is in the range of the content in the top part of Mycklemossen. The importance of the stabilizing interactions contra the destabilizing reactions for peatland C dynamics need further investigation.

Question 1.8

Line 420: Could this be assessed if there are CO₂ flux measurements from the site?

Answer

The CO₂ flux data we have available show that in 2018 that was the very dry year, soil respiration was higher compared to “normal” years (Keane et al., 2021).

This is now commented on in the discussion in line 498.

Reviewer 2

Question 2.1

First, the reported Fe-DOC correlation ($R^2 = 0.96$) is striking but insufficiently explained. The term “hydrological connectivity” alone does not capture the underlying chemical mechanisms. Please discuss whether the observed Fe-DOC co-variation results primarily from colloidal co-transport, redox-driven Fe-organic complexation, or other processes. In addition, the asynchronous timing of Fe (2018) and DOC (2019) peaks require clarification. Does this temporal offset reflect delayed microbial production, hydrological hysteresis, or differences in source zones within the peat profile?

Answer

The text below about the underlying chemical mechanisms and whether the observed Fe-DOC co-variation results primarily from colloidal co-transport, redox-driven Fe-organic complexation or other processes is discussed from L445 to L459. The asynchronous peak is discussed in L460 to L479.

The revised text in the manuscript is similar, but not identical to the text-response found below.

In general, Fe in boreal rivers is primarily transported on colloidal form, for instance as Fe-DOC complexes with humic substances, and Fe as Fe^{2+} or Fe(oxy)hydroxides (Heikkinen et al., 2022). The transport of these forms of Fe is primarily affected by redox conditions (oxic/anoxic) and pH, whereas oxic conditions favor co-precipitation of DOC and Fe (Riedel et al., 2013) and acidic pH (4-5) favors soluble Fe-DOC complexes, while a pH of 6 and higher favors precipitation of Fe-DOC complexes (Neubauer et al., 2013).

We believe our reported Fe-DOC relationship is mostly explained by colloidal transport of Fe-DOC complexes, with essentially all Fe bound to an organic ligands and a small proportion as Fe(oxy)hydroxides. It has been reported in a recent study that Fe in northern rivers is dominantly transported on colloidal form (between 1kDa and $0.22\mu m$), and otherwise and to a much less extent found as “truly dissolved” ($<1kDa$) (Aleshina et al., 2024). Björnerås et al., (2021) investigated Fe inflow to a lake from a Mire, in the south of Sweden. Here, Fe was found bound mainly on colloidal form, but also a noteworthy proportion as Fe(oxy)hydroxides, but here the inflow water had a pH of 6.7 and above, which favors precipitation of Fe-DOC complexes. At the pH we measured (4-5), we would expect a higher fraction of soluble Fe-DOC complexes.

The asynchronous timing of Fe (2018) and DOC (2019) peaks is probably due to a higher soil respiration of DOC in 2018. Both Fe and Pb concentrations were highest in 2018, so the peat degradation was likely highest that year. In 2018, the soil respiration was 15% higher compared to “normal” years (Keane et al., 2021). The year of 2018 was an extreme warm and dry summer in Scandinavia, and therefore it is likely that the larger soil respiration was due to mineralization of DOC, leading the lower DOC peaks in 2018 compared to 2019, which would also explain the higher release of Pb and Fe in 2018.

The large DOC peak in 2019 is harder to explain. One possible explanation is that the stream was supplemented by DOC rich water from the catchment, as supported by Fig. S2, that shows the the pH in the stream water in 2019 was notably higher than 2018 (pH 5-6.5) in the summer period. The high pH is not what we expect from water coming from the mire. Furthermore, there was a much higher precipitation in 2019 compared to 2018 (see table below), that could lead to a higher runoff, rich in DOC, from the forested catchment.

| SMHI data from Vänersborg | |
|---------------------------|--------------------|
| Year | Precipitation (mm) |
| 2015 | 875 |
| 2016 | 655 |
| 2017 | 708 |
| 2018 | 599 |
| 2019 | 911 |
| | |

Question 2.2

Second, the Pb isotope work is technically robust, and the evidence for anthropogenic Pb contamination derived from gasoline combustion is convincing. Nevertheless, I recommend that the authors provide propagated uncertainties for the isotope ratios and isotope-mixing model outputs and compare their measured isotope values with established European reference baselines (these appear to be missing from the manuscript). Moreover, please include confidence intervals for the estimated ~33% Pb retention in the lake to substantiate this quantitative conclusion.

Answer

- We have added the paragraph below to the method section 2.8.
- Regarding the mixing model, line 286-289 now describes calculation of the uncertainties and the new table S1 shows the Pb export values used for the calculations – Table S1 table is also inserted below.
- Northern European isotope ratios is now included in line 567 -571
- The 33% has been recalculated and is now 19%

Uncertainties of the Pb isotopic composition are reported at 2s and include quadratic addition of the standard error for the 10 replicates, the excess scatter of the primary reference solution NIST SRM 981 for the respective ratio from the measurement session and the uncertainty in the ²⁰⁴Hg correction. The uncertainties from the published ratios for the primary reference material (approximately 0.04%; NIST SRM 981, Cantanzaro et al., 1968) would need to be propagated to add systematic uncertainties. These uncertainties are considerably smaller than the random uncertainties (approximately 0.30%) and thus do not significantly contribute to the total uncertainties when added in quadrature.

| | Station 1 | Station 6 | St1 / St6 |
|---|------------------|---------------------------------|-----------|
| Catchment area (km ²) | 0.595 | 1.337 | 0.44 |
| Annual discharge (m ³ yr ⁻¹) | 136 155 ± 35 168 | 412 878 ± 181 006 | 0.33 |
| DOC export (kg yr ⁻¹) | 5834 ± 1674 | 12616 ± 9293 | 0.46 |
| Fe export (kg yr ⁻¹) | 151 ± 39 | 325 ± 232 | 0.46 |
| Pb export (kg yr ⁻¹) | 0.705 ± 0.193 | 0.681 ± 0.520 | 1.04 |
| of which from St. 1 | | 84.2 ± 2.6% = 0.573 ± 0.438* | |

* High uncertainty due to the variability in Pb export from Station 6

| | Northern Europe | | |
|--------------------------------------|-----------------|--------|---------|
| | Minimum | Median | Maximum |
| Pb | 1.6 | 9.6 | 52 |
| ²⁰⁶ Pb/ ²⁰⁷ Pb | 1.143 | 1.258 | 1.727 |
| ²⁰⁷ Pb/ ²⁰⁸ Pb | 0.287 | 0.397 | 0.414 |
| ²⁰⁸ Pb/ ²⁰⁶ Pb | 1.477 | 2.017 | 2.702 |

Question 2.3

Third, the finding that surface peat Pb concentrations exceed ecotoxic thresholds (>90 mg kg⁻¹) is both important and policy-relevant. However, the manuscript does not adequately discuss Pb speciation or its geochemical associations, which are critical for assessing Pb mobility and ecological risk.

Answer

The Pb mobility and ecological risk is discussed from L496 to L509. The text is inserted below:

Lead is toxic for mosses (Choudhury & Panda, 2005) and hampers the growth of microbial communities associated with *Sphagnum fallax* (Nguyen-Viet et al., 2007). Lead contents of 80 – 300 mg kg⁻¹ soil can affect microbial decomposition of C and nutrient turnover (Bååth, 1989; Zheng et al., 2017). The Pb content of 92 mg kg⁻¹ in hummocks at 25-50 cm depth are thus likely to affect the microbial community and the biomass turnover rate. The studies Nguyen-Viet et al., 2007, Bååth, 1989 and Zheng et al., 2017 reports effects of Pb based on contents (mg kg⁻¹) and not bioavailable Pb. Therefore, the high Pb content measured in hummock in this study, could have a potentially harmful effect on microbes. The bioavailability of Pb can be assessed in different ways, but requires some form of bioassay (Fleming et al., 2013; Hoang et al., 2025). In peatlands, Pb is mostly found strongly bound to organic matter and minerals with a low available fraction and is generally very immobile due to high binding affinity for OM (Hou et al., 2019; Lu et al., 2025; Novak et al., 2011; Vile et

al., 1999). The bioavailable Pb in hummock is therefore certainly lower than the 92 mg kg⁻¹, but Pb can be made available by microbial decomposition and accumulate in organisms over time.

Question 2.4

Given that the studied mire is strongly Sphagnum-dominated, the biochemical characteristics of Sphagnum mosses likely play a central role in the observed Fe-DOC-Pb interactions. Sphagnum tissues contain abundant polyphenolic compounds and organic acids, all of which can influence Fe and Pb cycle and modulate DOC chemistry. Could the authors elaborate on how these unique Sphagnum traits might govern the tight Fe-DOC correlation and the substantial Pb retention observed in this system? For example, does the acidity and high ligand density of Sphagnum-derived organic matter affects the stability of Fe-DOC-Pb complexes? A brief discussion along these lines would substantially enhance the ecological and mechanistic relevance of the study.

Answer

We are not aware of the specific type of DOC that leaves the mire contra the lake, but the relationship between Fe and Pb with DOC seems a little stronger in the stream leaving the mire contra the lake.

Characteristics related to *Sphagnum* that might govern the tight Fe-DOC relationship is introduced in the introduction from L122:

Dissolved organic matter from peatlands might even have a higher binding affinity to metals due to high contents of polyphenolic compounds, seemingly derived from Sphagnum, compared to DOC supplied from other types of catchments (Kaal et al., 2017). The strong correlation between Pb and DOC is likely important for the total export of Pb from peatlands, but it could also have implications for the retention of Pb in downstream systems.

L448 in the discussion: In addition, water that leaves Sphagnum-dominated bogs are typically rich in polyphenolic DOM that is seemingly derived from Sphagnum and binds strongly to metals (Kaal et al., 2017). The transport of the different forms of Fe is primarily affected by redox conditions (oxic/anoxic) and pH, whereas oxic conditions favor co-precipitation of DOC and Fe (Riedel et al., 2013) and acidic pH (4-5) favors soluble Fe-DOC complexes, while a pH of 6 and higher favors precipitation of Fe-DOC complexes (Neubauer et al., 2013)

Reviewer 3

General Comments

The manuscript investigates the mobilization of Pb, Fe, and DOC from a mire–lake system in southern Sweden, as well as the system’s carbon storage, over a four-year period marked by drought events that affected ecosystem dynamics.

The topic is relevant for understanding ecosystem responses to climate change and for the preservation of peatland–lake systems in Europe. However, the scientific conclusions are not entirely novel. As the authors acknowledge, the role of drought–rewetting cycles and hydrological connectivity in controlling DOC, Fe, and Pb export is already well established (e.g., Broder & Biester 2015, 2017; Rezaeehad et al., 2016).

The study presents a comprehensive and valuable dataset, supported by an extended four-year sampling period, which surpass typical studies based on shorter (1–2 year) campaigns. The comparison between the mire and lake compartments provides a broader perspective on the functioning of these common northern European ecosystems.

Overall, the topic is suitable for publication, but several revisions are recommended to improve the presentation and contextualization of the results. The discussion, in particular, needs a deeper and more critical analysis of the controlling processes and factors. Emphasis should also be placed on highlighting the novel aspects of this work and implications.

Specific Comments

Question 3.1

The introduction would benefit from a clearer and more concise presentation of the current state of knowledge regarding Fe, DOC, and Pb export from peatlands—particularly the roles of hydrological connectivity, drought, and precipitation events (see Broder et al.). This section could be shortened by summarizing previously established processes collectively, allowing the focus to shift toward how these factors specifically affect the studied system and its long-term dynamics.

Answer

We agree. The paragraph about hydrological connectivity, drought, and precipitation has been substantially amended. It has not been shortened, but we have tried to describe the processes in a manner that more specifically address our studies system.

Question 3.2

The statement that “how the export of DOC and hydrology affect the transport of metals is unknown for most peatlands” is somewhat overstated. While some mechanistic details remain uncertain, several key processes are already considered common to peatlands (see previous comment). If previous research cannot be considered indicative for this system, it becomes

difficult to reconcile this with the claim in Line 366 that the study site represents northern European mires in the temperate–boreal transition zone. Please clarified.

Answer

Yes, several key processes include hydrological connectivity, precipitation and drought. The statement “how the export of DOC and hydrology affect the transport of metals is unknown for most peatlands” have been removed. These known drivers are almost certainly also key processes in our mire-lake complex and is now better introduced from L106 to L116. See text below:

Several key processes exhibit control on metal export from peatlands, such as pH and the different metal’s binding affinity to DOC, but hydrological connectivity that refers to the transport of water from peat to stream, and seasonality, which includes drought and precipitation events seem to be most important (Broder & Biester, 2015, 2017; Prijac et al., 2023). In bogs, DOC is primarily produced from decomposition, which is likely the main mobilisation process of Fe and Pb (Broder & Biester, 2017). Temperature is important for DOC production as it affects decomposition rates (Rosset et al., 2022), but the most important factor is likely oxygen availability that is linked to water table depth and enables degradation of accumulated lignin-like compounds in the peat (Fenner & Freeman, 2011; Freeman et al., 2001). Water table decline primarily occurs during warm and dry periods during summer and early fall. Typically, at this time of year the highest export and stream water concentrations of DOC, Fe and Pb is observed and particularly in relation to high discharge events where hydrological connectivity between peatland and stream re-establishes (Broder & Biester, 2015, 2017; Clark et al., 2007; Prijac et al., 2023).

Question 3.3

Several trace metals (Pb, Hg, As, Cd) were analyzed in peat cores as part of previous work (Tchounwou et al., 2012). Since only Pb is discussed in the current manuscript, I recommend omitting mention of the other trace metals in both the methods and the introduction to maintain focus and clarity. If not, I recommend explaining in more detail the implication for other trace metals analyzed. For example, considering the Pb behaviour.

Answer

We have omitted the parts about Hg, As and Cd and keep focus on Pb.

Question 3.4

The Pb isotope dataset is valuable, covering mire, lake, and forest samples. However, its interpretation is limited. The isotope data suggest a mixing line between European gasoline, coal, and natural geogenic sources, with the lake outflow positioned between the mire and forest endmembers. This pattern likely reflects contributions from both local (forest) and upstream (mire) sources, implying that the lake catchment exerts additional influence (indicated by the forest).

In this regard, please consider:

General answer

The catchment areas have been generated based on detailed topographic maps in relation to the location of the measuring stations (out-flow) of the mire and the lake Ersjön. Thus, all the water that are measured at these locations are from these two areas. We do not think we need to add more maps to the manuscript, but this is now better clarified in the method section. Specifically:

L164 to L171 and Table S1: Two streams lead away from Mycklemossen. One runs from the north into the lake Ersjön (this stream is hereafter referred to as S1) and the other runs into a neighbouring catchment. Ersjön is 0.061 km², 4.5 m deep in the deepest part and has characteristics of a mesotrophic lake. Ersjön is sourced from Mycklemossen and from a neighbouring mostly forested catchment and drains through a stream (hereafter referred to as S6). The total catchment area of Mycklemossen (S1) is 0.595 km² and the catchment of Ersjön (S6) is 1.337 km² (Table S1). The sizes of the catchments have been generated based on detailed topographic maps in relation to the location of the measuring stations (out-flow)

L271 to L280: To determine the fate of anthropogenic Pb in the mire-lake complex, we applied an isotope mixing model (Eq. 1). Assuming Ersjön receives Pb from two different sources, water from Mycklemossen and other sources from the surrounding forest with distinct isotopic value the mixing model used is:

$$\text{Equation 1.} \quad R_{\text{(Ersjön)}} = f_1 \times [R]_{\text{(Mycklemossen)}} + f_2 \times [R]_{\text{forest}}$$

With R being the isotope ratio of Pb and f being the relative contribution of the two sources, with $f_1 + f_2$ equal to 1. The model was applied both with the ²⁰⁶Pb/²⁰⁷Pb and ²⁰⁸Pb/²⁰⁶Pb ratios, using the IsoError tool (Phillips & Gregg, 2001) that provides mean contributions in addition to an uncertainty estimate. 11 samples from a forest stream within the Skogaryd catchment were analysed in 2023.

- Exploring implications for DOC origin: how does catchment size and type (forest, agriculture) affect lake outflow composition compared with mire outflow? Do sites S1 and S6 directly receive water from other catchment areas? Relevant literature includes Kaal et al. (2017, 2020), which highlights the contribution of forest organic matter to DOM in similar mire systems and could strengthen this interpretation.

Answer

Mycklemossen receives almost all its water from rain and some from forested soils, and as we discuss already, the Ersjön has a larger inflow from forest land (there is no agriculture to speak of in the catchment, just a tiny field).

- Clarifying the processes of Pb and in lake outflow. Does it primarily derive from the mire (directly). Estimating or discussing the lake's water residence time could help address this. What are the implications for the lake being a "sink" for Pb from the peatland?

Answer

Yes, it is primarily from the mire, which is calculated by the isotope mixing model. No previous estimates of the water residence time have been made, but given the size of the lake (6.2 ha) and the mean depth of 1.7 m (Milberg et al., 2017)) the volume would be ~100 000 m³, which gives a mean residence time of about 3 month.

This estimate is found in L565.

- Based on isotopes and Pb. Can any kind of extrapolation be made about the role played by the mire-lake system depending on the flow (precipitation, drought period, etc.)?

Answer

Except for one sampling, the 206/208Pb ratio is always higher in Erssjön than Mycklemossen, proofing that the forest land almost always contributes to the lead in Erssjön. Conducting mixing model on the individual sampling dates (N=10), Mycklemossen contributes by 68 – 100% to the lead in Erssjön outflow.

The contribution calculated by the mixing model is found from L568 to L573.

Question 3.5

Although CN ratios are presented, their implications for organic matter degradation are not discussed in sufficient depth. Given the importance of decomposition in DOM formation in peatlands, I recommend expanding on the observed CN trends and discussing how they reflect mass loss or varying degradation intensities within the cores. (See Biester et al., 2014 and Zeh et al., 2020 for comparison between proxies for OM decomposition.

And

Question 3.6

The results for $\delta^{13}\text{C}$ and $\delta^{15}\text{N}$ show clear variations, yet their interpretation remains superficial. The authors should elaborate on how these isotopic shifts relate to organic matter degradation, plant sources, and CN ratios, and what they reveal about peat formation and transformation processes. See Zeh et al., 2020; Gandois et al., 2019.

Answer

Question 3.5 and 3.6 is now discussed in the discussion, as part of section 4.1 "Assessment of decomposition and C stability in Mycklemossen". The text in the manuscript is a more concise version of the below text and contain new references. From L403 to 423.

Looking at the top part of Mycklemossen where we observe the largest differences in C/N between hollow and hummock ranging from a median value of approximately 25 to 75 for hollow and hummock respectively. (Fig. 3). First, there are differences in the vegetation between the two topographies, with the hummock being mostly dominated mainly by vascular plants (*Eriophorum vaginatum*, *Calluna vulgaris* and *Erica tetralix*) and the hollows mainly consists of different *Sphagnum* species as *S. rubellum*, *S. fallax* and *S. austinii* as well as *Rhynchospora alba* (Kelly et al., 2021). The higher median value of C/N in hummock can therefore be explained differences in the dominant vegetation, meaning the higher C/N in hummocks is likely explained by higher lignin content from vascular plants and by the presence of roots (Biester et al., 2014; Zeh et al., 2020).

The depletion of $\delta^{13}\text{C}$ in hummock contra hollow, in particular in top layer, is a common tendency and in our case, the most likely explanation is likely that hummock is dominated by vascular plants (Biester et al., 2014; Zeh et al., 2020) or a higher degree of grasses, “the Suess effect”, as mentioned in the manuscript.

The increase of $\delta^{13}\text{C}$ towards 200cm depth, where hummock reach similar values to hollow and intermediate might indicate aerobic degradation of lignin in the hummock and preservation of carbohydrates from below the water table. The lower median value of $\delta^{15}\text{N}$ in hummocks at the top of the mire contra hollow and intermediate, might be because microbial denitrification have removed light N isotope components (Biester et al., 2014). In general, the $\delta^{15}\text{N}$ increased with depth and is likely because of an uptake of the lighter N isotopes

Question 3.7

The differentiation among hollows, hummocks, and intermediate positions yields interesting insights into trace metal accumulation and peatland heterogeneity.

The discussion could be strengthened by integrating findings from Pérez-Rodríguez et al. (2025), who examined degradation dynamics under aerobic versus anaerobic conditions in similar microtopographies.

Additionally, it would be helpful to clarify whether the hollow–hummock pattern is assumed to have remained consistent throughout the peatland’s development. And what are the possible implications. See Nungesser (2003).

Answer

The most interesting results in (Pérez-Rodríguez et al., (2025) in relation to our study, is the leaching of phenolic compounds from the hummock topography, which could have implications for how metals are mobilised and transported. While data explaining the degradation patterns of hollow contra hummock seems interesting, the apparent selective preservation of lignin-like compounds in hollows seems a little curious. Bryophytes do not contain S-type lignin (Weng & Chapple, 2010), as otherwise shown by the paper, so the source of lignin-like compounds measured in hollows is a little unclear.

Hummocks and hollows are generally relative resilient to climate shifts and the average environmental conditions (hydrology, temperature, species composition) over time

determines the topography, that said, hummocks and hollows are formed by the species themselves (Nungesser, 2003). The time scale for development of hummock and hollow is long compared to other ecosystems, so we would assume the topography has remained the same in Mycklemossen during the time when Pb pollution was ongoing. It could imply that Pb is better sequestered in hummocks as overall slower decomposition rate is observed compared to hollows. Also, it might be worth considering that hummock makes up most of the Mycklemossen topography (Rinne et al., 2022) and together with the height of hummocks is more exposed to atmospheric deposition.

Question 3.8

The suggestion that Pb toxicity may inhibit microbial degradation of organic matter deserves further consideration. Where is the Pb located in the moss and moss-derived organic matter, and is this Pb likely to be bioavailable to microorganisms?

Answer

We got a similar question from reviewer 2, that also got a similar answer.

The question regarding whether Pb is bioavailable to microorganisms is discussed from L496 to L509, with some attention to where Pb is located. See text below:

Lead is toxic for mosses (Choudhury & Panda, 2005) and hampers the growth of microbial communities associated with *Sphagnum fallax* (Nguyen-Viet et al., 2007). Lead contents of 80 – 300 mg kg⁻¹ soil can affect microbial decomposition of C and nutrient turnover (Bååth, 1989; Zheng et al., 2017). The Pb content of 92 mg kg⁻¹ in hummocks at 25-50 cm depth are thus likely to affect the microbial community and the biomass turnover rate. The studies Nguyen-Viet et al., 2007, Bååth, 1989 and Zheng et al., 2017 reports effects of Pb based on contents (mg kg⁻¹) and not bioavailable Pb. Therefore, the high Pb content measured in hummock in this study, could have a potentially harmful effect on microbes. The bioavailability of Pb can be assessed in different ways, but requires some form of bioassay (Fleming et al., 2013; Hoang et al., 2025). In peatlands, Pb is mostly found strongly bound to organic matter and minerals with a low available fraction and is generally very immobile due to high binding affinity for OM (Lu et al., 2025; Novak et al., 2011; Vile et al., 1999). Organic matter content (including CEC) and pH are likely the two most important factors for heavy metal availability, of which high organic matter decrease availability (Hou et al., 2019). The bioavailable Pb in hummock is therefore certainly lower than the 92 mg kg⁻¹, but Pb can be made available by microbial decomposition and accumulate in organisms over time.

Also, the Pb concentrations for water streams in relation to toxicity and bioavailability is discussed from L523 to 535. See text below:

The average Pb concentrations in stream water from Mycklemossen in the period of 2017 to 2020 was 4-fold higher than the long-term accepted values of 1.2 µg L⁻¹ and suggests that this stream, according to European standards, is of bad environmental quality (Sjöberg, 2016). In stream water from Ersjön, the concentration of Pb was drastically lower (1.1±0.1

µg L⁻¹), and can be considered good quality for long term exposure (Sjöberg, 2016). The highest concentration of Pb in stream water from Mycklemossen was measured in late 2018 and reached 14 µg L⁻¹, which is the short-term maximum limit value for fresh water (Sjöberg, 2016). As already pointed out, the high Pb concentration in S1 (14 µg L⁻¹) in 2018 was likely due to a higher degree of Pb mobilisation (Broder & Biester, 2017). Beside the European standard for Pb thresholds, a comprehensive analysis taking bioavailability into account and including 7 European surface water scenarios and 25 freshwater species, found Pb toxicity thresholds to within 6.1 and 31 µg L⁻¹ (Van Sprang et al., 2016). Therefore, Pb reached concentrations on short-term basis that can be considered potentially harmful for freshwater fauna. Furthermore, it seems that longer periods of drought in a future climate could lead to higher mobilisation of Pb and stream water concentrations of Pb exceeding toxicity thresholds (Szkokan-Emilson et al., 2013).

Question 3.9

While the cited literature on peatlands as trace metal sinks is appropriate, the authors should also consider referencing the extensive work conducted by Bindler's and Kylander's groups on Swedish peatlands, which would provide useful regional context.

Answer

We have included:

Kylander, M. E., Bindler, R., Cortizas, A. M., Gallagher, K., Mörth, C.-M., & Rauch, S. (2013). A novel geochemical approach to paleorecords of dust deposition and effective humidity: 8500 years of peat accumulation at Store Mosse (the "Great Bog"), Sweden. *Quaternary Science Reviews*, 69, 69–82. <https://doi.org/10.1016/j.quascirev.2013.02.010>

Bindler, R., Brännvall, M.-L., Renberg, I., Emteryd, O., & Grip, H. (1999). Natural Lead Concentrations in Pristine Boreal Forest Soils and Past Pollution Trends: A Reference for Critical Load Models. *Environmental Science & Technology*, 33(19), 3362–3367. <https://doi.org/10.1021/es9809307>

Question 3.10

The citation (González & Pokrovsky, 2014) in line 64 is not appropriate. Although the authors developed an excellent model to understand trace metal accumulation in mosses, their results are not specifically related to the peatland context.

Answer

Yes, that is a fair point, as the result were obtained under controlled laboratory conditions and not in a peatland – the reference has been omitted.

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