

## ***Interactive comment on* “Enrichment of trace metals from acid sulphate soils in sediments of the Kvarken Archipelago, eastern Gulf of Bothnia, Baltic Sea” by Joonas J. Virtasalo et al.**

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Received and published: 21 September 2020

General comments:

This work contributes to important scientific questions regarding the transport and fate of metals mobilised from oxidised acid sulfate soils, which fall within the scope of Biogeosciences.

While not a novel concept, per se, the results and conclusions of this paper are important contributions to understanding the source-sink transport of contaminants in estuarine-marine systems which, to date, is poorly constrained due to complex biogeo-

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chemistry and the interconnectivity of sediment transport processes, geomorphology, hydrology, and climate.

The sampling design, analytical methods, and statistical analyses are robust and appropriate.

Specific comments:

Research aims should be more explicitly presented in the final paragraph of the introduction.

The role of Fe-oxyhydroxides in metal sequestration is mentioned in line 48, however the discussion of Fe is absent from this paper. Data from Australia point to Fe species as a major product of acidic drainage, and a major sink for mobilised metals (see Bush et al., 2004; Mosely et al., 2018; Job et al., 2020). I recognise that some of the papers you reference do not observe high levels of dissolved Fe in stream waters affected by acidic drainage, but Fe could be transported in solid phases (near-source precipitation), and subsequently still be accumulating at elevated levels in this terminal system. If you have data to the contrary, stating this would be a helpful insight to international audiences.

Bush, R. T., Fyfe, D. and Sullivan, L. A. (2004) 'Occurrence and abundance of mono-sulfidic black ooze in coastal acid sulfate soil landscapes', *Australian Journal of Soil Research*, 42(5–6), pp. 609–616. doi: 10.1071/sr03077.

Mosley, L. M. et al. (2018) 'Fate and dynamics of metal precipitates arising from acid drainage discharges to a river system', *Chemosphere*, 212, pp. 811–820. doi: <https://doi.org/10.1016/j.chemosphere.2018.08.146>.

Job, T., Penny, D. and Morgan, B. (2020) 'Geochemical signatures of acidic drainage recorded in estuarine sediments after an extreme drought', *Science of The Total Environment*, 749, p. 141435. doi: <https://doi.org/10.1016/j.scitotenv.2020.141435>.

Regarding the correlation of trace metals Cd, Co, Cu, Ni, and Zn to the grain size

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fraction (2–6  $\mu\text{m}$ ), and to C&N, which you conclude to represent metal-organic matter aggregates; can you please clarify how this relationship can be established when the grain-size analysis method digests organic matter? Also, why might the relationship between C&N with trace metals be less evident in the robust PCA analysis?

In lines 265-285, when discussing the Manganese data, the impact of redox transformations on the down-core element profile is mentioned. Do you have any data on redox conditions in the cores? The colour change in the core photos is notable. An acknowledgement of how redox transformations may be impacting the geochemical profiles of the other elements would be of benefit. The redox transition appears to be at notably shallower core depths than the 1986 temporal marker.

Redox states are also of significance to the Risk Assessment section where redox conditions have implications for bioavailability (the presence of AVS for example). I suggest it is acknowledged in this section that further data is required to determine the bioavailability of these metals (dilute acid-extractable, for example). The magnitude of enrichment is certainly sufficient to flag potential ecotoxic risk however.

A brief description of changes in lithology observable in the cores would be of benefit in the primary manuscript (I recognise that core photos are included in the supplementary material – this could be referenced in-text), at the very least to exclude sedimentological changes as a primary driver of geochemical variability.

It is identified that the 2–6  $\mu\text{m}$  grain-size fraction is positively associated with trace metal loading – how variable is this grain-size fraction down-core? Regarding sediment transport, OM accumulating with 2–6  $\mu\text{m}$  siliciclastics or carbonates would presumably be coarser in grain-size due to density differences. A down-core log of any grain-size variability may similarly help interpret controls on down-core geochemical variability.

An assessment of experimental precision should be included. Were replicates analysed?

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Lines 43-44: Provide a reason why climate change might increase acid release from AS Soils.

Lines 59-67: I think the importance of understanding contaminant dynamics should be more explicit / clear – it is a valuable contribution of this research.

Line 108: ... cool and dark conditions - include the approximate temperature of storage, as well as time until laboratory analysis. Where/how were the samples stored in the shore-based laboratory?

Line 171: the grain-size distributions are described as poorly sorted but from a narrow grain-size range. This sounds contradictory (poor sorting implies wide grain-size range) however I think the intention is that the median grain-sizes exhibit low variability. Please clarify, and consider including the range of sorting measurements.

Line 334: precipitation does not only occur 'out to sea' - explicitly identify neutralization, which can coincide with reaching seawaters.

Technical Comments:

The term contents is regularly used, when perhaps concentration would be more explicit and clear. Consider amending.

Line 12: which is the recipient system of the Laihianjoki and Sulvanjoki ...

Line 14: ... landscape. Metal deposition has remained at high levels since ...

Line 26: ... low pH conditions in pore- and surface-waters ...

Lines 28-30: Due to the uncertainty, perhaps simplify these two sentences into one.

Line 54: deteriorative\*

Line 63: in the long term

Line 111: Define 'fresh'.

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Line 118: ... classified as those ...

Line 128: ... dissolved in 1 M HNO<sub>3</sub>...

Line 130: Instead identify which element was analysed by which technique inside brackets.

Line 168: ... in the R software environment.

Line 195: Mn contents are low except for a strong increase

Line 241: the pattern of decreasing metal contents with increasing distance

Line 242: The median metal contents ...

Line 251: Reword this sentence as it implies uniformity both laterally and vertically, which you go on to clarify is not the case.

Lines 287-300 – here, and some other sections of the discussion, are moderately convoluted with data, impacting readability. Some of this could be shifted into the results section, or just simplified.

Line 307: begin to decrease

Line 312: as a result of, for example, ... (the abbreviation here breaks readability)

Line 312-313: rather than a decrease

Lines 316-323 – I do not think this paragraph is necessary. It is mostly already explained earlier in the manuscript.

Line 401: ecotoxicological\*

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Interactive comment on Biogeosciences Discuss., <https://doi.org/10.5194/bg-2020-231>, 2020.

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