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Effect of past century mining activities on sediment properties and toxicity to freshwater organisms in northern Sweden

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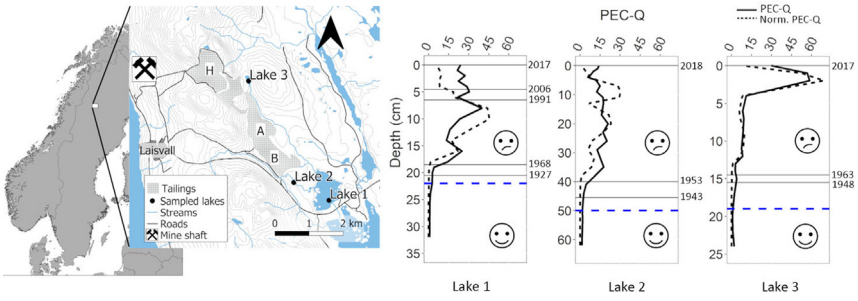
HIGHLIGHTS

- Temporal and spatial patterns of past mining were assessed using lake sediments.
- All lakes were affected, with increased metal accumulation at the onset of mining.
- The contaminant load, toxic risk and patterns of recovery differed between lakes.
- Predicted toxic effect was affected by total organic carbon content.
- There is a need of using normalized PEC-Q to better assess toxic effects.

GRAPHICAL ABSTRACT



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ABSTRACT

The release of toxic metals from local mining activities often represents a severe environmental hazard for nearby lake ecosystems. Previous studies on the impact of mining have primarily focused on single lakes, with less emphasis on spatial and temporal recovery patterns of multiple lakes within the same catchment, but with different hydrological connection and distance to the pollutant source. This knowledge gap prevents us from assessing the real environmental risk of abandoned mines and understanding ecosystem recovery. This study explores the intensity and spatial patterns of sediment contamination and the potential for ecosystem recovery in three lakes in close vicinity of a lead (Pb) and zinc (Zn) mine in Sweden that has been inoperative for >20 years. Dated (^{210}Pb and ^{137}Cs) sediment cores from each lake were used to reconstruct temporal patterns in trace element deposition and relate those with past mining activities. Results show that all lakes were affected by mining, indicated by increasing Pb and Zn concentrations and decreasing organic matter content, at the onset of mining. However, the extent and timing of mining impact differed between lakes, which was partly ascribed to differences in the historical use of tailings and settling ponds. Assessment of toxicity levels in sediments, based on normalized Probable Effect Concentration Quotient (PEC-Q) to organic matter content, provided more consistent results with the historical mining than conventional methods, showing a decreasing impact in lakes once the operations ceased. Still, sediment Pb concentrations were > 10 times higher than pre-mining values, evidencing the urgent need for remediation actions in the study lakes. This study highlights the importance of considering spatial heterogeneity in metal deposition, sediment organic matter content, and hydrological connectivity with tailings when risk assessments are performed in mining-impacted lakes. The use of normalized PEC-Q in toxic assessments is also recommended.

1. Introduction

Active and closed mines are a major threat to aquatic ecosystems (Akcil and Koldas, 2006; Beck et al., 2020; Gutierrez et al., 2016). Lakes and

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streams close to mine districts may receive large loads of metals through leakage and erosion of contaminated soils and mining material (Macklin et al., 2006; Rose et al., 2012), and atmospheric deposition (Renberg et al., 2000). These inputs of metals can result in the degradation of the ecosystem leading, for example, to decreased abundance and diversity of freshwater organisms (Clements et al., 2000; Dudgeon et al., 2006; Hogsden and Harding, 2012; Iwasaki et al., 2018), which, in turn, may harm the entire ecosystem functioning and productivity (Carlisle and Clements, 2003; Carlisle and Clements, 2005; Ferreira et al., 2016). On a landscape level, metal exposure and environmental effects in mining-impacted environments have been proven to vary at small spatial (few km) and temporal scales (years) (Cooke et al., 2017; Gonzalez-Miqueo et al., 2010; Miller et al., 2005), which challenges assessing the overall impact of mining activities in nearby ecosystems.

Current environmental risk assessments for contaminated sediments generally focus on measurements of metal concentrations and their comparison to threshold limits. This approach, however, does not adequately assess toxic effects as i) only a minor fraction of metals present in sediments is bioavailable for uptake by exposed organisms and ii) it assumes that organisms are exposed to individual toxic metals, which rarely occurs in nature (Méndez-Fernández et al., 2014; O'Connor and Paul, 2000; Roosa et al., 2016; Zhang et al., 2014). Several factors may affect metal bioavailability, such as metal speciation, organic matter content, pH, and alkalinity (Simpson and Batley, 2007). In addition, these factors may all vary over time due to mining activity (Belzile et al., 2004). Among the approaches used to assess sediment toxicity, the Probable Effect Concentration Quotient (PEC-Q) enables toxicity estimations for contaminant mixtures in relation to sediment quality criteria (SQC), where the latter is based on toxicological laboratory studies in sediments (Besser et al., 2015; Ingersoll et al., 2001; MacDonald et al., 2000).

In addition to metal toxicity, knowledge about the ecosystem recovery rate is crucial to developing appropriate management and remediation strategies once mining activity has stopped. Similar to metal concentrations, the recovery rate is influenced by catchment characteristics. For example, vegetation cover in the catchment controls soil and sediment transport to freshwaters (Anderson, 2014; Rathburn et al., 2018), which often continues after mining activity has ceased (Bindler et al., 2008). Consequently, legacy metal contamination associated with this transport can continue to expose organisms for centuries (Bindler et al., 2009). Unfortunately, long-term monitoring data is often missing, which hampers the assessment of natural background conditions and, thus, long-term impact and recovery of contaminated environments. In this context, sediment records are a powerful tool that can provide valuable information about historical (>100 years) and recent (<100 years) environmental conditions (Renberg et al., 2001), such as contaminant fluxes (Couillard et al., 2004), changes in sediment toxicity (Cheney et al., 2020; Juracek, 2008; Rose et al., 2018), and changes in geochemical properties (Jarsjo et al., 2017; Page et al., 1994; van der Post et al., 1997).

In this study, we used geochemical records and analyses of toxicity to examine differences in the assessment of long-lasting effects of past century-scale mining activities, including recovery, in three lakes situated in the vicinity of one of the largest Pb mines in Europe. Studying lakes at different locations from the pollutant source and with different hydrological connectivity to the tailings offered an excellent setup to assess variations in spatiotemporal patterns in metal sediment contamination. We hypothesize that i) lakes receiving direct water discharge from the tailings will have higher contamination than isolated lakes, leading to a higher impact assessment when using their sediment record as a proxy for the whole area, ii) all lakes will show similar temporal contamination patterns that match with the historical exploitation of the mine, and iii) all lakes might have started to recover, although their recovery rates will be highly dependent on water and thus, pollutant, discharge.

2. Method

2.1. Site description

The study was conducted in the proximity of a closed Pb and Zn mine at the east border of the Caledonian mountain range near Laisvall, northern Sweden (Fig. 1). Climate is characterized by an average annual temperature of 1.4 °C and average annual precipitation of 724 mm (Boliden, 2018). The vegetation is dominated by old-grown spruce (*Picea abies*) and pine (*Pinus*

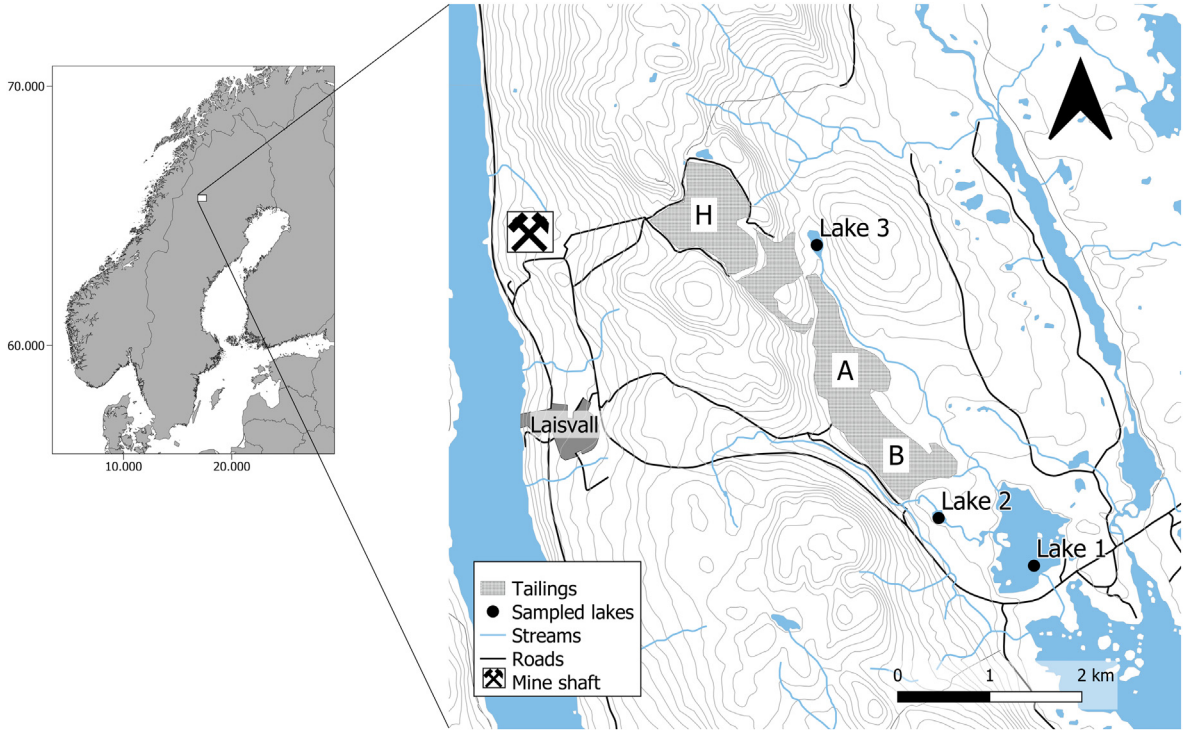

Fig. 1. Map of the study area in northern Sweden including the location of the mine shaft, mine tailings (H, A, and B) and sampled lakes (Lake 1–3).

sylvestris) trees, including also deciduous trees such as birch (*Betula* sp.) and willows (*Salix* sp.). Deciduous trees are more abundant in riparian zones disturbed by mining.

With a deposit of about 64 Mt. of 4 % Pb (Saintilan et al., 2016), the underground mine in Laisvall was the largest Pb mine in Europe at the time (production period 1943–2001) (Widmark, 1983). The overbearing mineral contained galena (PbS) and sphalerite (ZnS) (Rickard et al., 1975), justifying the focus of Pb and Zn in this study. Excavated material was processed in a concentrator at the site, and the separate tailings were kept water-covered in tailing ponds to prevent oxidation. Tailing ponds were established east of the mine: pond A in 1952, pond B in 1965, and pond H in 1976, in total covering approximately 3.5 km^2 (Fig. 1). The tailing ponds were later dewatered and covered with till, and replanted with vegetation as part of the post-treatment: pond A and B during 1978–1982, and pond H during 1997–2002. Today, the tailing ponds are sparsely covered by vegetation dominated by willows (*Salix* sp.).

In the south-east (Lake 1) and east (Lake 3) of the tailing ponds, natural lakes were used as clearing ponds to settle particles and decrease the input of metal-rich particles to adjacent lakes. Lake 1 (maximum depth 4.8 m and approximate area of 12 ha at present) was used as a clearing pond between 1966 and 2001. Lake 3 (maximum depth 4.8 m and approximate area of 4 ha at present) was used as a clearing pond between 1997 and 2001 (Fig. 1). A dam was built in 1966 to increase the capacity and retention time of the south clearing pond (Lake 1) by increasing the water level. When the mine closed in 2001, the dam was removed, rendering a water level similar to pre-mining conditions. Lake 2 (maximum depth 2.5 m and approximate area of 2 ha at present) was not used as a clearing pond (Fig. 1), although it is situated within the catchment area of the tailing ponds and has received runoff from the tailings. The surface of the lake catchments consists mainly of lodgment till, with small areas of exposed bedrock and peat. The studied lakes are hydrologically open, and stream outlets of Lake 2 and 3 are connected to Lake 1. Groundwater inputs likely affect metal budgets in all lakes. No other regional or local metal contamination sources have been identified within this remote and sparsely populated area.

2.2. Sediment sampling

Sediment cores were collected in winter of 2017 from the ice at the deepest part of each lake (Fig. 1; Table 1) using a Kajak sampler (Renberg, 1991). The core from Lake 2 was not deep enough to reach pre-mining sediments, and the lake was sampled again in 2018 using a Russian peat corer. A complementary core for the surface sediments was collected with a Kajak sampler in 2019. All cores were sliced into 1 cm segments. Samples were stored at −18 °C until further analyses. Frozen samples were freeze-dried, weighted, and ground prior to analysis. Further analyses of sediment layers (metal, organic content, and radionuclide analyses) were performed in every cm of the top 5 cm (except one sample at 3–4 cm depth from Lake 3) followed by every second cm. For the Russian core from Lake 2, samples were analyzed every second to third cm. For dating purposes (see below), additional layers below this depth were analyzed.

2.3. Metal concentrations and organic content in sediments

Sediment metal concentrations were analyzed by X-ray fluorescence (XRF), using a Bruker S8-Tiger WD-XRF with a detection limit of 0.7 and 0.9 mg kg^{-1} for Zn and Pb, respectively (Rydberg, 2014). Certified reference material (NCS DC-70314, NCS DC-73048 and NCS DC-73310, Chinese National Analysis Center for Iron and Steel) were included in the analysis, providing 2–10 % accuracy for Pb concentrations between 23 and 285 mg kg^{-1}, and 0.3 to 2 % for Zn concentrations between 52 and 500 mg kg^{-1}.

Organic matter content was determined by loss-in-ignition (LOI) in those samples analyzed for metal concentrations. Approximately 3.5 cm^3 of sediment material was combusted at 550 °C for 4 h (Heiri et al., 2001). LOI was converted to total organic carbon (TOC) using a conversion factor of 2, based on the assumption that organic matter is 50 % carbon (Pribyl, 2010). This factor is slightly higher than the traditional conversion factor of 1.724, which has been questioned in more recent literature (Pribyl, 2010). We acknowledge that this conversion provides a rough estimate of TOC, and parameters such as sediment grain size can limit the accuracy of TOC conversion (Vereș, 2002).

To match the Kajak and Russian cores from Lake 2, we used non-metric multidimensional scaling (NMDS, package: Vegan in R). We included metal concentrations, LOI, and PEC-Q (see Section 2.5 Sediment toxicity) for the top 9–11 cm from the two respective cores and estimated their similarity using Mahalanobis distance, combining all dimensions of the NMDS. The results indicated that the highest similarity (a difference of 0.05) occurred between 3 and 5 cm depth in the Russian and Kajak core, respectively (Fig. 2). Hence, in this paper, results from the upper 0–5 cm interval in Lake 2 sediment core originates from the Kajak core while the remaining profile (5–63 cm) originates from the Russian core.

2.4. Sediment chronology

Sediment chronologies were established using ^{210}Pb. ^{210}Pb is a naturally occurring radionuclide from the ^{238}U decay series with a half-life of 22.3 years. The distribution of the atmospheric or unsupported ^{210}Pb in the sediment can be used to derive an age-depth model covering the accumulation for the past 100–150 years (Appleby and Oldfield, 1992). The artificial radionuclide ^{137}Cs was used to constrain the derived ^{210}Pb chronology. ^{137}Cs has a half-life of 30 years and was spread globally through atmospheric fallout from nuclear testing in 1954, with a maximum emission in 1963. Later, and mainly in Europe, the Chernobyl accident in 1986 introduced significant amounts of ^{137}Cs into the atmosphere. Clear

Table 1
Sampling depth and core length for each site and core type.

Site	Core type	Sampling depth (m)	Core length (cm)
Lake 1	Kajak	3.8	33
Lake 2	Kajak	1.15	12
Lake 2	Russian	1.15	61
Lake 3	Kajak	4.8	25

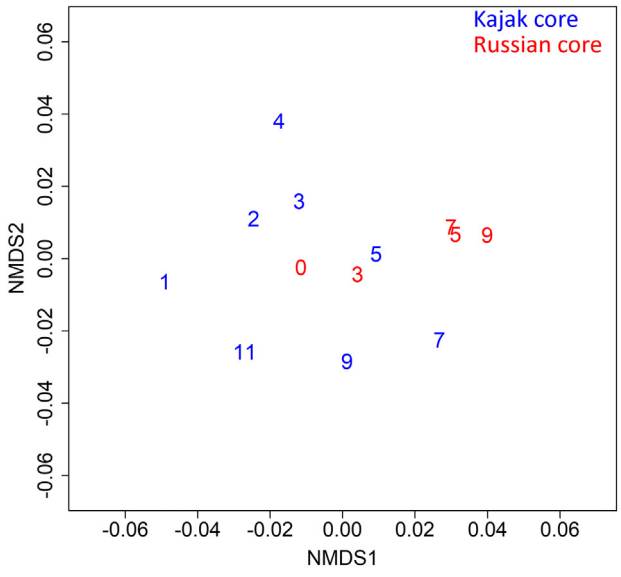

Fig. 2. Ordination plot of the NMDS analysis made to correlate the two cores (Kajak core in blue and Russian core in red) from Lake 2. Numbers indicate depth of the sediment layer for each core.

peaks of ^{137}Cs concentration in sedimentary deposits can be used to identify the occurrence of those incidences and validate ^{210}Pb-derived ages (Smith, 2001).

Total ^{210}Pb and ^{137}Cs concentrations were measured by gamma spectrometry using a high-purity Small Anode Germanium (SAGe) well detector (Canberra, Model GSW120). To do this, approximately 4.8 cm^3 of dry ground, homogenized sediments were placed into calibrated geometries. Samples were sealed for three weeks prior to analyses to ensure ^{222}Rn/^{226}Ra equilibration. Total ^{210}Pb concentration was measured via its gamma emissions at 46.5 keV. ^{226}Ra content, used to estimate the supported ^{210}Pb fraction, was determined by measuring the concentration of its daughter radionuclide ^{214}Pb measured at its gamma emission at 295 keV and 351 keV. Unsupported ^{210}Pb concentration was calculated as the difference between total and supported ^{210}Pb concentration. ^{137}Cs concentrations were determined by measuring its gamma emission at 662 keV.

The determination of a ^{210}Pb age-depth model for modern (< 150 years) sediments in mine-impacted sites can be challenging, mainly because the ^{210}Pb record is often altered by intermediate layers of mine material from the tailings (see Results and discussion section). Therefore, due to the difficulties of accurately quantifying the total unsupported ^{210}Pb content, accurate ^{210}Pb dating models such as the Constant Rate of Supply (CRS) model (Appleby and Oldfield, 1978) cannot be applied. Similarly, surface ^{210}Pb concentrations are commonly affected by mining and other processes, such as bioturbation or mixing, invalidating the use of the Constant Initial Concentration (CIC) model (Robbins, 1978). Therefore, mean sediment accumulation rates (SAR) over the past 100–150 years were estimated using the Constant Flux: Constant Sedimentation model (CF:CS) (Krishnaswamy et al., 1971). Unlike the CRS, the CF:CS model assumes that SAR does not vary with time. However, the model can be applied in a piecewise way using the ^{137}Cs date as a reference point (Appleby, 2001) to interpret deviations of ^{210}Pb concentrations from the typical exponential decline as the result of mining activities.

2.5. Sediment toxicity

We used PEC-Q to estimate sediment toxicity over time. For each sediment section, PEC-Q was derived for Pb and Zn combined according to Eq. (1),

$$PECQ = \frac{\sum_i^n \frac{C_{M_i}}{PEC_{Mi}}}{n} \quad (1)$$

where C_{Mi} is the concentration of the metal M_i in the sediment section, PEC_{Mi} is the probable effect concentration for metal M_i (MacDonald et al., 2000), and n is the number of metals included in the PEC-Q estimation (here, $n = 2$). As suggested for paleotoxicity (Rose et al., 2018), a mean PEC-Q > 0.5 was used as an indication of possible biological effects and a mean PEC-Q > 2 was used as a threshold for probable biological effects. This is based on studies indicating that an increased risk of toxicity occur already at a mean quotient of >0.5 for chronic exposures (Ingersoll et al., 2001), and that biological effects of metal exposure may be expected if the mean quotient exceeds 2 (Rippey et al., 2008). Because a high organic carbon content in sediment may reduce the toxicity, we also calculated normalized PEC-Qs where C_{Mi} were normalized against TOC content (Long et al., 2006).

2.6. Statistical analyses

Spatial and temporal patterns in geochemical properties were explored using principal component analyses (PCA; *prcomp* function). We included sediment layers from all three lakes and their individual TOC content, density, Pb and Zn concentrations and the estimated age (year of formation). All variables were scaled, and the analyses were carried out using R version 3.6.1 (R Core Team, 2019).

3. Results and discussion

3.1. Sediment stratigraphy and core chronology

All sediments were characterized by a layer of dark brown gyttja at the bottom of the cores, where the lowest densities (0.35 ± 0.02, 0.12 ± 0.01, and 0.39 ± 0.03 g cm^{-3} for Lake 1, 2, and 3, respectively) and the highest TOC content (15.8 ± 0.2, 19.9 ± 0.3 and 18.0 ± 0.1 % for Lake 1, 2, and 3, respectively) were found (Fig. 3A). An intermediate grey layer was found above this horizon (transition depth at 20, 40, and 15 cm for Lake 1, 2, and 3, respectively). Densities in this intermediate layer increased up to a factor of 5 to 7 compared to the bottom layers. A drop in the TOC content below 2 % was also observed (Fig. 3A). The uppermost 2 to 5 cm of the cores were visually similar to the deepest layers for all profiles, with increasing TOC content towards the surface (up to 3.5–11 %) and twice as high in Lake 2 compared to Lake 1 and 3. Similar changes in sediment characteristics, including a visible event layer with similar trends in TOC content, are typical for mining-impacted sediments (McDonald et al., 2010). Thus, assuming that the intermediate layer (hereafter referred to as the event layer) found in our sediment cores corresponds to perturbations of the sedimentary record linked to mining activities in the area seems reasonable.

Lithological changes along the cores were also evident from the record of radionuclides (^{210}Pb and ^{137}Cs) (Fig. 3B). Unsupported ^{210}Pb concentrations do not show the exponential decrease with depth expected from radioactive decay in any core. In particular, unsupported ^{210}Pb was not detected in the event layer. On the other hand, ^{226}Ra concentrations increased significantly in the event compartment for Lake 3 (data not shown). Non-detectable unsupported ^{210}Pb concentrations, together with the significant differences in ^{226}Ra concentration between altered and non-altered layers, are a good indicator of changes in sediment composition and allow interpretation of the unsupported ^{210}Pb profile as influenced by rapid sedimentation occurring during the event layer, rather than biological perturbation (Abril and Brunskill, 2014; Arnaud et al., 2002; Huh et al., 2006). This is also confirmed by increased density and decreased TOC content in the event layer (Fig. 3A). Thus, we can confirm that rapid sedimentation is associated with increased activity in the catchment due to mining activities (Miller et al., 2019; Richerson et al., 2008), with the consequent enhancement of minerogenic inputs from the tailings.

After applying the CF:CS model in Lake 1 at the upper, non-altered compartment (0–4 cm), we obtained a SAR of 0.8 ± 0.7 g cm^{-2} year^{-1}. Measurable concentrations of ^{137}Cs concomitant with measurable unsupported ^{210}Pb below the event layer suggests that the signal of these radionuclides has been preserved (Fig. 3B). We associated the ^{137}Cs concentration peak found at 18.5 cm with the maximum atmospheric deposition of ^{137}Cs occurred in 1963. By combining the most recent SAR with the 1963-horizon, we found that the event layer occurred in the 1963–1991 period for Lake 1 (Fig. 3B). For the non-altered deepest compartment (20–30 cm), the CF:CS model provided a SAR of 0.05 ± 0.01 g cm^{-2} year^{-1}. Using this accumulation rate, the increase in ^{137}Cs concentration was located around 1954, consistent with the beginning of the nuclear weapon testing period and hence confirming the ^{210}Pb-derived chronology.

For Lake 3, a typical exponential decay of ^{210}Pb concentration with depth was observed only below the event layer (below 14 cm). For the non-altered compartment (14–18 cm), the CF:CS model estimated a SAR of 0.030 ± 0.014 g cm^{-2} year^{-1}. Using this SAR and the peak of ^{137}Cs at 14.5 cm as the 1963 horizon, we estimated that the event layer occurred during the 1948–2010 period (Fig. 3B). We acknowledge that the chronologies estimated for Lake 1 and 3 assume that the deposition of the mining material occurred at a constant rate.

We could not apply the CF:CS model in Lake 2 due to the altered distribution of unsupported ^{210}Pb (Fig. 3B). For Lake 2, we assumed that sediment accumulated up to undetectable concentrations of unsupported ^{210}Pb (60 cm) corresponded to approximately 100 years of accumulation (Appleby, 2000). A clear peak of ^{137}Cs at 33.5 cm was used as the 1963 horizon (Fig. 3B). Using these two age-depth horizons, we estimated a mean accretion rate of 0.60 cm year^{-1}, suggesting that the increase in density

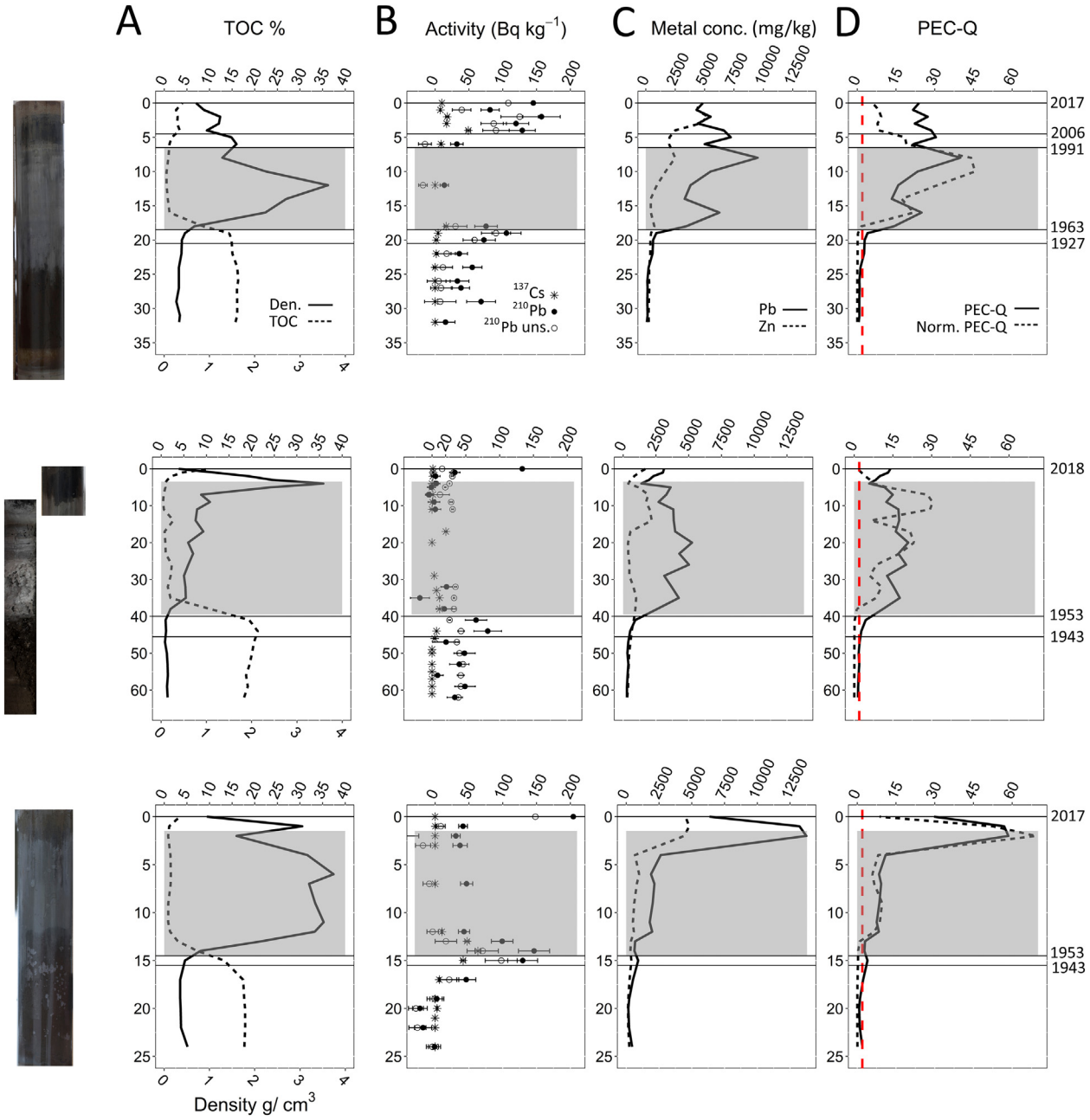

Fig. 3. Sediment profiles of Lake 1 (upper panels), Lake 2 (middle panels) and Lake 3 (lower panels and A) total organic carbon (TOC) and sediment density, B) activity of ^{137}Cs, ^{210}Pb and unsupported ^{210}Pb, C) metal (Pb and Zn) concentrations, and D) PEC-Q values with (norm) and without normalization against TOC. Photos of the sediment cores are provided in the left margin, and grey areas indicate the compartment of the cores affected by mining activities (the event layer). Approximate dates of the sediment layers, based on the radionuclides, are provided on the right y-axis. The red dashed vertical lines in plot D represents the threshold for probable biological effects (PEC-Q = 2).

and decrease in TOC content observed at 40 cm depth would correspond to 1951. Assuming that the event layer accumulated at a constant rate, we located this event to the 1951–2013 period.

3.2. Impact of mining activities: spatial and temporal patterns

Mining has affected all three lakes considerably, which is evident from the clear intermediate light grey layer found in all cores accompanied by the high increase in sediment density, the decrease in organic content (Fig. 3A), and the alteration of the ^{210}Pb record (Fig. 3B). Mining impact in the lakes is also confirmed by the significant increase in metal concentrations (Pb and Zn) (Fig. 3C). However, the impact from the mine differs both in magnitude and timing depending on the lake's spatial setting concerning the mine and tailing's location, as well as on the historical use of lakes. This conclusion is reinforced by the results from the PCA analysis, which indicated that lakes showed more similarities before the start of mining operations, compared to the period that corresponded to layers accumulated during and after mining ceased (Fig. 4). The variation of almost all variables included in the PCA is largely explained (~90 %) by the three identified axis. PC1 explained 58 % of the variance in the data and was mainly driven by the time of sediment formation (estimated year) and, to less extent, TOC and metal content. Of these variables, TOC and year correlated negatively with each other. PC2 explained 20 % of the variance and was driven by sediment density, metal content, and TOC. PC3 (not shown) explained 11.8 % of the variance and was mainly driven by year. Lakes 1 and 3 are strongly affected by metal accumulation, especially post-mining. During active

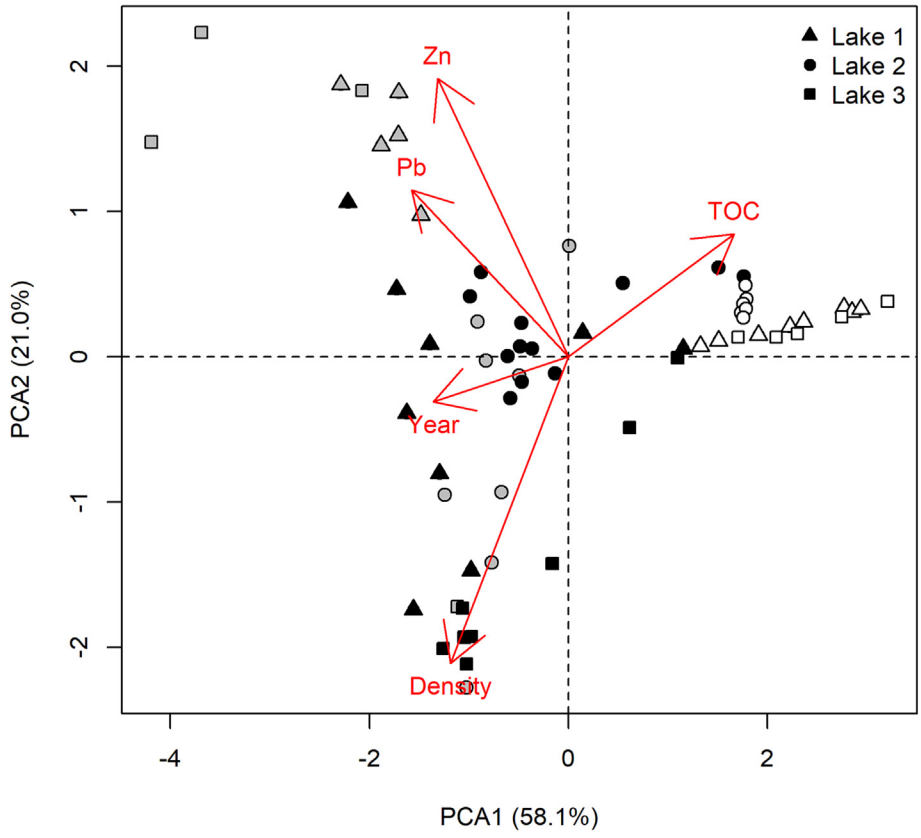

Fig. 4. Principal component analysis (PCA) for each different sediment layer in Lake 1 (triangles), Lake 2 (circles) and Lake 3 (squares). Open symbols correspond to pre-mining conditions (<1943), black symbols correspond to the time of active mining (1943–2001) and grey symbols correspond to post-mining conditions (>2001) and arrows represent all variables included in the PCA.

mining, sediment density was a stronger driver in Lake 3, indicated by negative loadings on PC2. Lake 2, however, showed little differences in geochemical properties during and after mining cessation. We acknowledge that post-sedimentation processes, such as diffusion, migration pore water advection, and mixing of the surface sediment layers (induced by waves, sediment-dwelling organisms or plants) could affect metal accumulation in lake sediments and justify, at least in part, some of the observed variations. However, Pb and Zn are regarded as stable elements, and only minor changes are expected to occur after sediment layers are buried at depths deeper than 5 cm (Percival and Outridge, 2013). Our assumption that changes in metal concentrations reflect variations in mining activities in the area is thus reasonable considering also that distinct metal concentration changes in sediments have been observed without any strong indication of diffusion even several hundred years after deposition (Bindler and Rydberg, 2016).

As expected, the lowest Pb and Zn concentrations were found in pre-mining sediment layers in all cores (mean concentrations in Lake 1, 2 and 3, respectively: 264 mg kg^{-1} Pb and 303 mg kg^{-1} Zn, 340 mg kg^{-1} Pb and 417 mg kg^{-1} Zn, 314 mg kg^{-1} Pb and 189 mg kg^{-1} Zn). But mean values varied up to 4 (for Pb) and 3 times (for Zn) between cores (Fig. 3C), which evidences the high spatial variability in metal accumulation in the lakes even before mine exploitation. As an example, the lowest Pb and Zn pre-mining concentrations were 88 mg kg^{-1} Pb and 239 mg kg^{-1} Zn in Lake 1, 294 mg kg^{-1} Pb and 313 mg kg^{-1} Zn in Lake 2 and 162 mg kg^{-1} Pb and 139 mg kg^{-1} Zn in Lake 3. Lake 2 also showed slight dissimilarities with Lakes 1 and 3 during the pre-mining period (Fig. 4), which highlights the heterogeneity and difficulties to evaluate background levels even on a relatively small spatial scale. In terms of temporal reconstruction of the mining impact, the onset of increasing metal concentrations also differed between cores and metals. During the early mining period (1943–1970), when TOC content decreased drastically (Fig. 3A), a rapid increase in Pb concentration occurred in all lakes, which is in line with previous studies that showed a negative correlation between TOC and metal content (Grayson and Plater, 2009; Morra et al., 2015). However, metal enrichment differed between lakes, finding a 13-fold increase in Lake 1, while more modest increases where observed in Lake 2 (7-fold increase) and Lake 3 (3-fold increase) (Fig. 3C). Similar results were found for Zn, with concentrations that increased in all lakes during the early mining period (1948–1970), but with a moderate enrichment compared to Pb (2-fold increase for all lakes, Fig. 3C).

The different spatial and temporal patterns of Pb and Zn found in the lakes can be related to the development of mine tailings and the post treatment efforts. Lakes 1 and 2 are located in the catchment area of the early established tailing ponds, and the initial increase in Pb (approximately 1940–1950) roughly corresponds to the construction of tailing pond A around 1952 and B around 1965. Pb accumulation peaked in the late 1980s in both Lake 1 (9520 mg kg^{-1}; 36-fold increase) and 2 (5258 mg kg^{-1}; 16-fold increase), a few years after tailing A and B were last in use (Fig. 3C). Lake 3, acting as clearing pond for tailing H between 1997 and 2001, instead showed a drastic increase in Pb accumulation during the 2000s with a peak in early 2010 (13741 mg kg^{-1}; 44-fold increase), corresponding to the time after tailing H was last in use. We do not rule out the possibility that the recent increase of Pb in Lake 3 was caused by increased leaching during post-treatment activities or due to altered biogeochemical processes. However, due to the magnitude of metal release from tailings, it is likely that differences in Pb accumulation in Lake 3 were mainly controlled by mining operations ascribing differences in Pb accumulation patters between lakes to differences in their historical use during mining operations. Even though atmospheric deposition may have affected the three lakes, especially during the early development of the mine (Csavina et al., 2012; Schelle et al., 2008; Sucharová and Suchara, 2004), run-off and leakage are the major sources of metals in mining impacted lakes. In these sites, most metals are transported in particulate-associated form

through stream water coming from the tailings (Macklin et al., 2006). As such, the two clearing ponds (Lakes 1 and 3) receiving direct discharge from mine tailings and run-off from contaminated surfaces had twice as high accumulation of Pb than Lake 2, which received no direct discharge from the tailings (Fig. 3C). Zn accumulation peaked later than Pb, with maximum concentrations from the late 1990s (Lake 2) to late 2010 (Lake 1 and 3). Zn peaks were, again, more pronounced in the two clearing ponds (Lakes 1 and 3), with a maximum concentration of 5282 and 4808 mg kg^{-1}, respectively (increase by a factor of 17 and 25 in relation to pre-mining conditions) compared to a maximum of 2181 mg kg^{-1} Zn in Lake 2 (increase by a factor of 6 in relation to pre-mining conditions). Based on our records, we cannot rule out that lake characteristics (e.g. size, depth, bathymetry) and catchment properties (e.g. size, relief, surface) might also have contributed to the accumulation patterns of the studied metals. However, overall sedimentation rates have been dominated by the event layer, and estimates of natural sedimentation rates during non-impacted periods were similar and low among all three study lakes. This indicates that our lakes show similar sedimentation drivers and processes despite their differences in topological and morphological characteristics.

Post-mining conditions also show diverging patterns in the more recent years, mainly due to different patterns in metal accumulation (Fig. 4). Geochemical properties were more similar during and after mining in Lake 2. In contrast, Lakes 1 and 3 were more strongly affected by metal accumulation in post-mining sediment layers, especially in Lake 3 (Fig. 4). This is confirmed by the results from the PCA, which showed that sediment layers accumulated during active mining in Lake 3 had negative loadings on PC2, while layers accumulated after mining ceased had positive loadings on PC2. Although Pb concentrations in top sediments decreased after mining ceased in Lake 1 and 3 (from a peak of 9520 to 4857 mg kg^{-1} and from 13,741 to 6348 mg kg^{-1}, respectively), Pb concentrations were still elevated compared to pre-mining conditions in recent times (enrichment factor 15–22, Fig. 3C). Zn concentrations, on the other hand, remained elevated in the uppermost 3 cm for the two clearing ponds (Lake 1 and 3). In contrast, Zn concentrations in Lake 2 were reduced by almost half in surface sediments. Although Pb concentrations initially decreased after the peak in the late 1990s, Pb concentrations increased in the upper sediment layers (from 1385 to 3119 mg kg^{-1}, Fig. 3C). This indicates differences in the recovery patterns between lake 2 and the settling ponds, which could be attributed to differences in direct (Lake 1 and 3) vs. indirect (Lake 2) discharge. As such, even between lakes within the same catchment, extrapolation of results should be made with caution. As Lake 2 never was used as a settling pond, this further supports a significant runoff from the surrounding catchment, also after mining has ended (Bauerek et al., 2013; El Khalil et al., 2008).

The offset between Zn and Pb accumulation could not be explained by any particular activity or event in the mining area, but as the result of different minerogenic composition of the mined ore (Saintilan et al., 2016). This different accumulation pattern may also be attributed to different mobility of these elements in soil and sediment (Huang et al., 2012; Wang et al., 2019), increased leakage of Zn from the mine tailings (Kossoff et al., 2011), remobilization within the sediment (Bordas and Bourg, 2001; Duran et al., 2012), passive upward diffusion through the sediment column (McDonald et al., 2010), or bioaccumulation of Zn in the upper sediment by living plants (Cambrollé et al., 2008) as Zn, but not Pb, is an essential element for organisms (Eisler, 1988; Eisler, 1993).

Leaching of Zn from old mine tailings is commonly occurring, which often results in exceeded quality guidelines in aquatic environments in Sweden (Erixson et al., 2012). The high accumulation of Pb in surface sediment suggests that Pb bound to soils and tailings still could leach into the lakes after mining has ended (Carvalho et al., 2014; Wang et al., 2019), which further provide support for the long retention time of Pb in sediments (Knox et al., 2006; Xu et al., 2019). Post-treatment of the mine tailings, including liming, might have prevented some transport of metals from the tailings. However, diffusive input of metals from contaminated soils is difficult to prevent (Blake et al., 2007), as proven by elevated Pb concentrations in soils surrounding the study lakes (Lidman and Berglund, 2022). Tailings contain large amount of metals, and even after post-treatment efforts such as coverage and planting of vegetation, erosion events and leaching may occur (Merrington and Alloway, 1994). As such, full recovery of mining impacted lakes can be delayed by 50–200 years (Pelletier et al., 2020; Schillereff et al., 2016).

3.3. Sediment toxicity

The variability in metal accumulation between lakes is evident and indicates that lakes are affected heterogeneously by mining, even within a small spatial scale (Cooke et al., 2017; Gonzalez-Miqueo et al., 2010). This is also reflected by the results of the PEC-Q. PEC-Q for all lakes exceeded the level of possible negative effects on the ecosystem (PEC-Q > 0.5) in all sediment layers. For the pre-mining compartment, levels of probable detrimental effects (PEC-Q > 2) were occasionally found, especially for Lake 3 (Fig. 3D). After 1940s, PEC-Q exceeded the value of 2 in all lakes and the ratio further increased during the event layer, reaching maximum values from 21 to 58 (Fig. 3D) that exceed similar risk assessments in rural environments (Rose et al., 2018). Based on the PEC-Q results, the highest risk for toxic effects in the study lakes occurred at the end of the mining period in Lake 1 and 3, when metal concentrations of both Pb and Zn were high, but sediment deposition rates started to decrease. In Lake 2, however, the toxicity risk remained constant during most of the mining period. Overall, and in line with the results from the metal accumulation, the toxicity assessment based on PEC-Q indicated more negative ecological impact in Lake 1 and 3 than in Lake 2. Of the two metals, Pb contributed most to the toxic risk due to its higher concentrations and lower PEC_M compared to Zn (MacDonald et al., 2000).

Using PEC-Q to estimate the toxic impact in sediments assumes an additive contribution of toxicity of metals (Long et al., 2006). Therefore, if co-occurring Pb and Zn have antagonistic or synergistic toxic effects, the risk assessment might be over- or underestimated. For example, Pb could replace Zn in enzymes, disrupting their regular action (Skerfving and Bergdahl, 2015). It has also been shown that Zn availability may further affect the toxicity of Pb (Wani et al., 2019). Also, the mechanisms of Zn toxicity are less understood than those for Pb, and Zn deficiency is generally more common than Zn toxicity, at least in vertebrates (Sandstead, 2015). However, Zn toxicity has occurred in aquatic insects (Wesner et al., 2014) and Zn may also be more toxic than Pb for invertebrates (Lidman et al., 2020). Thus, the potential interaction effects of Pb and Zn should be studied further. Another limitation of using PEC-Q as a toxic assessment approach is that PEC-Q is based on total metal concentration, without considering the bioavailable fraction of the metal of interest. The bioavailability of metals, in turn, is strongly affected by organic matter, pH, speciation, and availability of competing cations (Simpson and Batley, 2007), factors that might also change during mining activity (Belzile et al., 2004; McDonald et al., 2010), and need to be considered when assessing metal toxicity.

In compliance with elevated Pb and Zn concentrations in post-mining sediment, there was still a high risk for toxic effects (PEC-Q > 13) in the aquatic ecosystem after mining stopped (Fig. 3D). Slow recovery has also been found in other mining-impacted environments, even after remediation has been performed (Gutierrez, 2020). However, the mining operation highly affected the TOC content in sediments, with large differences between pre-, event- and post-mining layers (Fig. 4). Therefore, when PEC-Q were normalized to organic matter content, the toxic risk was reduced, albeit still evident in all three lakes, irrespective of their historical impact by the mine (Fig. 3D). Due to efficient binding of dissolved metals (Ankley et al., 1996; Di Toro et al., 2005), a higher organic matter content generally decrease the metal uptake by organisms (Besser et al., 2003; Zhang et al., 2014) and hence reduce the toxic risk. Thus, when there are large temporal or spatial variations in organic matter content, normalization of PEC-Q may be appropriate. The need for normalization in the present study was further required as PEC-Q, but not normalized PEC-Q, indicated a possible negative effect (PEC-Q > 0.5) during pre-mining conditions (Fig. 3D), which seems unreasonable.

As an effect of the low TOC content during mining, normalized PEC-Q indicated an even higher risk for toxic effects (a quotient between 30 and

69) than non-normalized PEC-Q in all lakes during mining operations. Although the toxic risk was highest at the late mining stage, the sediment layer with the highest risk, based on PEC-Q vs. normalized PEC-Q, did not always coincide. This was especially true for Lake 2, where Pb concentrations were lower and TOC content was more variable in the event layer compared to Lake 1 and 3. Although TOC content has not yet approached pre-mining conditions in post-mining sediment, normalizing PEC-Q by organic content reduced the toxic risk by 3–4 times in Lake 1 and 3, and as much as 10 times in Lake 2 in the uppermost sediment, compared to the non-normalized approach. Due to a high TOC content in the most recent sediment layer in Lake 2, the conditions in this lake indicate a possible, but not a probable risk for detrimental effects based on normalized PEC-Q, hence recovery is expected within a near future. Nonetheless, no matter whether using normalized or non-normalized PEC-Q, there is still a current risk of toxic effects in the area, especially in the settling ponds (Lakes 1 and 3). This has previously been confirmed by reduced aquatic insect emergence and survival in the studied lakes (Lidman et al., 2020). However, the community composition in mining impacted lakes may also adapt to the high metal exposure and sensitive species may be replaced by more tolerant ones (Thienpont et al., 2016).

4. Conclusions

This study evidences the very high spatial and temporal variability in pollution burden legacy in mining-impacted lakes, which constitutes a significant environmental hazard for aquatic ecosystems that should be adequately quantified. Although mining affected all three lakes, those receiving direct discharge from the mine (i.e., used as settling ponds) accumulated higher metal concentrations compared to lakes subjected to runoff only. However, there were also temporal differences in metal accumulation patterns between lakes that were used as settling ponds, especially regarding the recovery patterns and toxic risks after mining had ceased. This highlights a need to assess multiple lakes within the same catchment, when risk assessments of mining-impacted environments are conducted. Finally, we recommend using TOC normalized PEC-Q and considering interactive effects between contaminants in current environmental risk assessments to better assess toxic effects of contaminated sediments.

CRediT authorship contribution statement

Johan Lidman: Conceptualization, Methodology, Investigation, Data curation, Formal analysis, Visualization, Writing – original draft, Project administration. **Carolina Olid:** Methodology, Formal analysis, Validation, Writing – review & editing. **Christian Bigler:** Conceptualization, Methodology, Investigation, Supervision, Writing – review & editing. **Åsa M.M. Berglund:** Conceptualization, Methodology, Investigation, Validation, Supervision, Writing – review & editing, Project administration, Funding acquisition.

Data availability

Data is available through the Swedish National Data Service (https://doi.org/10.5878/a7yd-dy51).

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests: Johan Lidman reports financial support was provided by Boliden AB. Johan Lidman's salary is, due to his position at the industrial doctoral school at Umeå University, partly funded by Boliden AB.

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