



Spatio-temporal impact of salinated mine water on Lake Jormasjärvi, Finland[☆]

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ABSTRACT

The salinization of freshwater environments is a global concern, and one of the largest sources of salinated water is the mining industry. An increasing number of modern mines are working with low grade sulfide ores, resulting in increased volumes of potentially harmful saline drainage. We used water monitoring data, together with data on sedimentary fossil remains (cladoceran, diatom and chironomid), to analyze the spatio-temporal (5 sampling locations and 3 sediment depths) impact of salinated mine water originating from the Talvivaara/Terrafame open cast mine on multiple components of the aquatic ecosystem of Lake Jormasjärvi, Finland. Lake Jormasjärvi is the fourth and largest lake in a chain of lakes along the path of the mine water. Despite the location and large water volume, the mine water has changed the chemistry of Lake Jormasjärvi, reflected in increased electrical conductivity values since 2010. The ecological impact is significant around the inflow region of the lake, as all biological indicator groups show a rapid and directional shift towards new species composition. There is a clear trend in improved water quality as one moves further from the point of inflow, and as one looks back in time. Our results show that salinated mine water may induce rapid and large scale changes, even far downstream along a chain of several sinking basins. This is of special importance in cases where large amounts of waste water are processed in the vicinity of protected habitats.

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1. Introduction

Mining is one of the key industries in the world, but mine water has the potential to inflict serious environmental damage downstream (e.g. Lin et al., 2005; Daniel et al., 2014). The pollution problems associated with mine water have been acknowledged for centuries (Hoover and Hoover, 1950) and despite of the recent achievements in mine water remediation (Younger et al., 2002), new risks too are related to advances in technology. New enrichment and extraction technologies have opened up new opportunities to access low-grade deposits. Subsequently, the volume of mine wastes generated is becoming increasingly large (Hudson-

Edwards and Dold, 2015). Large mines have to manage enormous quantities of water and, subsequently, in some cases the mine water has the potential to affect large waterbodies downstream.

Many metals are associated with massive sulfide deposits, and sulfate (SO₄) is a typical constituent of mine waters generated by sulfide ore mining. Poor grade metal sulfide mines are regarded as the future of modern mining, and numerous new mines will be activated in the near future (Watling, 2015). SO₄ pollution may induce multiple environmental impacts. SO₄ accelerates mercury (Hg) methylation, which results in elevated concentrations of the most toxic form of Hg in the food web (e.g. Gilmour et al., 1992), and subsequently poses a risk to human health (Hong et al., 2012). Moreover, in sedimentary pore water, SO₄ has toxic effects on aquatic plants (Myrbo et al., 2017). Elevated salinity induces osmotic stress for fresh water biota, and SO₄ is acutely toxic in large concentrations (Meays and Nordin, 2013; Wang et al., 2016). In addition, SO₄ pollution may cause a lake to become meromictic,

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which may eventually destroy the deep habitat, thus totally altering the ecosystem (Leppänen et al., 2017). Salinization has been acknowledged as a global threat (Cañedo-Argüelles et al., 2013; Kaushal et al., 2018) to freshwater ecosystems. However, despite of some studies on the impact of salinization on freshwater lake ecosystems (e.g. Hoffman et al., 1981) and river ecosystems (e.g. Kaushal et al., 2005; Corsi et al., 2010; Dugan et al., 2017) there is still a growing need to assess the impact of salinization on especially freshwater lakes in colder regions.

Mining companies in many countries are obliged by environmental legislation (e.g. Directive, 2014/52/EU in European Union) to establish monitoring programs which generate water chemistry datasets, usually spanning many years. These datasets can be used to assess the impacts of mine effluents on the chemistry of the receiving waters. Even though biological sampling is usually less comprehensive than chemical sampling, paleolimnological methods can be applied to study historical populations (Smol, 2008). Such thorough and detailed pollution histories, combined with available water chemistry data, enables one to establish a spatio-temporal framework which integrates pollution characteristics and corresponding impacts on biological indicators in time and space. An extensive range of paleolimnological proxies exists to study the impacts of environmental pollution in lake systems (Smol, 2008). In boreal lakes, suitable paleobioindicators are e.g. diatoms (Rimet, 2012; Debenest et al., 2013), cladocerans (Jeppesen et al., 2001; Eggermont and Martens, 2011), and chironomid larvae (Luoto and Ojala, 2014; Thienpont et al., 2016).

The objective of this study is to evaluate the impact of salinated mine water pollution on the aquatic ecosystem of a boreal lake along a spatio-temporal gradient, by using three paleobiological indicator groups and water chemistry data. The main hypothesis is that the salinated mine water has changed the aquatic ecosystem over both space and time, with increasing impacts trending towards the present and the pollution source.

2. Materials and methods

2.1. Study area

Lake Jormasjärvi is a dimictic, slightly acidic (pH ~6.3), oligomesotrophic (total P ~13 µg/L, total N 400 µg/L) and brown colored (88.1 mg Pt/L) lake (OIVA database, 2017). It is situated in the region of Kainuu, central Finland (N 64° 4' 29.347" E 28° 10' 6.468" WGS84), at an altitude of 145 m.a.s.l. The regional average for annual temperature is ~2 °C, and for precipitation ~600 mm (Kersalo and Pirinen, 2009). The lake has a surface area of 21 km² and a mean depth and maximum depth of approximately 6 m and 28 m, respectively, while the catchment area covers 300 km² (Fig. 1). The bedrock of the catchment area consists mainly of mica schists and black shale, which are easily eroded rocks containing high concentration of sulfides and metals. Thus, the background levels of metal and sulfur concentrations in Lake Jormasjärvi are relatively high (Kumpulainen, 2001). The vegetation of the catchment area consists mainly of coniferous forest (85%) and smaller water formations (total area 8.5%). The population level of the catchment area is low, with only ~200 residents, and the proportion of agricultural fields is negligible (1%). Nearly the entire area of the catchment has been ditched for forestry, with the total length of ditching exceeding 2800 km (>9 km/km²). The most intense ditching has been conducted at the western part of the catchment area (Supplementary Fig. 1). The catchment area also underwent intensive foresting activities (cutting, ditching) during the 1960s and 1970s, which is reflected in the elevated mineral concentrations in the sediment layers and higher metals concentrations in the sediments, as sampled in the central-western part of the lake

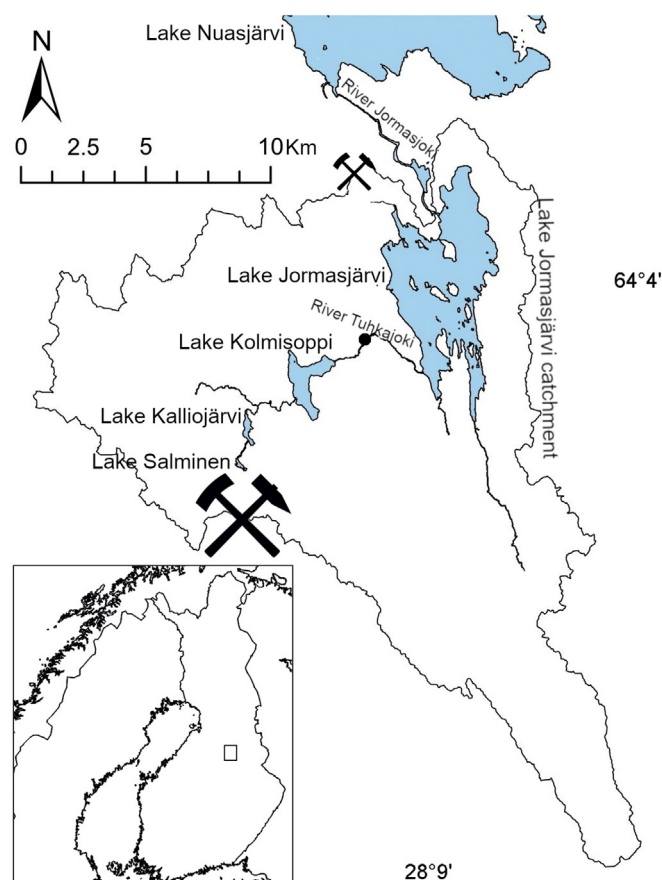


Fig. 1. Study area. The mine symbol denotes the location of the Talvivaara/Terrafame mine. By default, the effluents flow northwards from the mine. The smaller mine symbol represents the closed talc mine of Lahnaslampi. Circle indicates water chemistry sampling site for Tuhkajoki River.

(Mäkinen and Kauppila, 2006). In addition, one of the ponds of the closed Lahnaslampi talc mine is located in the catchment area of Lake Jormasjärvi, which may also have an impact on the water quality of the lake, particularly in the central western area. However, no clear chemical changes in the lake were detected for the decades previous to the opening of the mine (Mäkinen and Kauppila, 2006).

The Talvivaara mine, located at the southern end of Lake Jormasjärvi, has altered Lake Jormasjärvi's water chemistry over the past decade. The Talvivaara nickel-sulfide deposit is one of the largest in Europe (Perez, 2016). The mine has struggled with water management issues since operations began in 2008. Numerous leaks of untreated and treated mine water have been reported, and multiple nearby lakes are currently polluted by metals and Na₂SO₄. The mine waters drain to two directions, southern and northern route. The most important mine-derived components in the effluent are Na₂SO₄, Ca, Mg and metals (Fe, Mn, Ni, Zn), due to the black schist ore and chemicals originating from the enrichment procedure (Kauppi et al., 2013). Metals are removed and acidity is treated during the water purification process, but the treated mine water is characterized by elevated Na₂SO₄ and is thus classified as saline drainage (Opitz and Timms, 2016). The environmental permit (since 2013) limits the SO₄ concentration of treated waste water to 6000 mg/L, but in earlier years the concentrations have been considerably higher (e.g. average 15 000 mg/L SO₄ in 2010, 13 000 mg/L SO₄ in 2011; Hietala et al., 2012). Since 2015, the nearby waterbodies have been bypassed by a pipeline, which channels the mine water directly into Lake Nuasjärvi (Fig. 1)

(Hakala, 2017). However, a fraction of the mine water is still being released into the lake chain from the north side of the mine (e.g. 1.5 million m³ in 2016; Hakala, 2017, 5.1 million m³ in 2015; Marttila and Hakala, 2016). The chain of lakes receiving the waste waters of the mine along the northern route is comprised of four lakes (L. Salminen, L. Kalliojärvi, L. Kolmisoppi, and L. Jormasjärvi) and roughly 16 km of connecting rivers, before draining into the considerably larger Lake Nuasjärvi (Fig. 1). Lake Jormasjärvi is the fourth lake in this lake chain. The three smaller lakes upstream of Lake Jormasjärvi act as settling basins, considerably reducing the amount of pollutants entering the lake (Fig. 1). The average deep water SO₄ concentrations are ~10 000 mg/L in Lake Salminen, ~4000 mg/L in Lake Kalliojärvi, and ~300 mg/L in Lake Kolmisoppi (Sivula et al., 2018). Despite this process, significant amounts of mine water still enters Lake Jormasjärvi, which can clearly be seen in the elevated SO₄ and manganese (Mn) concentrations close to the inflow area in the southern part of the lake (ELY, 2017).

Due to the Talvivaara/Terrafame mine water release, Lake Jormasjärvi and the upstream waterbodies have been characterized by elevated salinity concentrations since 2010 (ELY, 2017). In addition to the contemporary controlled releases of salinated mine water into the upper reaches of the waterway, a major dam accident also occurred in 2012. However, according to the Finnish Environmental Institute (Kauppi et al., 2013), Lake Jormasjärvi was not impacted by this accidental dam leak. The current state of Lake Jormasjärvi is thus mainly the result of a controlled mine water discharge. According to the national authorities, the primary pollutants in Lake Jormasjärvi are Mn and SO₄. However, very high Mn concentrations were also reported in Lake Jormasjärvi before the mining activities began (over 2000 µg/L; ELY, 2017), and thus Mn is not an ideal surrogate for mine water assessment in this study. In contrast, SO₄ and sodium (Na) concentrations have exhibited ten-fold increases in the surface water during the post mining era. Moreover, in the inflowing Tuhkajoki River (Fig. 1), the average concentrations of SO₄ and calcium (Ca) have increased by hundred- and ten-fold, respectively, since 2007 (Table 1). Thus, the level of electric conductivity (EC) can act as a potential surrogate parameter to demonstrate the impact of mine water discharge.

2.2. Sampling

A total of five short (<25 cm) sediment cores were retrieved

Table 1

General mean surface water chemistry data of Lake Jormasjärvi (15 sampling locations) and the Tuhkajoki River. *denoted data is sampled from the Tuhkajoki River (Fig. 1). N represents the number of measured samples, and SD the standard deviation. EC denotes electrical conductivity. Concentrations are total unfiltered concentrations. pH is presented as median values and min-max ranges (in SD column).

| | Pre-2008 | N | SD | Post-2008 | N | SD |
|-----------------------|----------|----|---------|-----------|-----|---------|
| Total P µg/L | 15.6 | 60 | 8.6 | 11.7 | 167 | 4.0 |
| Total N µg/L | 447 | 78 | 81.3 | 419 | 171 | 115 |
| pH | 6.3 | 78 | 5.5–6.8 | 6.3 | 169 | 4.8–6.9 |
| Ca mg/L | 2.8 | 9 | 0.5 | 12.5 | 73 | 5.5 |
| Mg mg/L | 0.96 | 5 | 0.05 | 2.9 | 73 | 1.0 |
| SO ₄ mg/L | 5.1 | 5 | 1.5 | 49 | 1 | |
| Na mg/L | 1.2 | 4 | 0.05 | 12.3 | 111 | 6.1 |
| EC mS/m | 3.1 | 51 | 0.4 | 14.0 | 169 | 7.7 |
| Zn µg/L | 16.3 | 9 | 10.4 | 37.3 | 64 | 18.1 |
| Ni µg/L | 8.9 | 9 | 6.3 | 12.2 | 99 | 6.0 |
| Total P µg/L* | 21.2 | 22 | 8.6 | 13.3 | 87 | 5.6 |
| Total N µg/L* | 528 | 22 | 161 | 569 | 88 | 139 |
| Fe µg/L* | 1343 | 3 | 432 | 837 | 176 | 348 |
| Mn µg/L* | 116 | 3 | 41.6 | 640 | 138 | 463 |
| Ca mg/L* | 3.0 | 3 | 0.6 | 36.1 | 83 | 17 |
| Mg mg/L* | 0.9 | 1 | | 8.4 | 74 | 5.0 |
| SO ₄ mg/L* | 1.9 | 2 | 1.3 | 201 | 106 | 110 |

using a HTH kayak corer (Renberg and Hansson, 2008) in August 2017, from five sampling sites (Fig. 2A) representing a spatial and temporal gradient. The spatial gradient covers a transect from the Tuhkajoki River inlet (site 1) in the south to the outlet of the Jormasjärvi River (site 5) in the north, whereas the sediment cores represent a temporal gradient covering the current time (0–0.25 cm), the pre- or early -mining period (3–4 cm), and the background/reference period (10–11 cm). The depths of the sub-sampled sediments are based on known sedimentation rates (Mäkinen and Kauppila, 2006), and should correspond roughly to the times of 2017 (0–0.25 cm), 2010 (3–4 cm), and the 1980s (10–11 cm). The subsamples were stored in plastic Ziploc bags and kept in cold storage prior to their analysis. The sediment samples were analyzed for diatoms, cladocerans, and chironomids in order to evaluate the mine water's impact throughout multiple trophic levels. We used measured water chemistry monitoring data to identify the most useful surrogate parameter to demonstrate the effects of the mine water discharge, and to assess the water quality changes in Lake Jormasjärvi and the Tuhkajoki River. The water chemistry data consists of sampling results from different sampling sites and sampling depths. The water chemistry data was obtained from the national databank, maintained by the Finnish environmental authority (OIVA database, 2017).

2.3. Diatom analysis

Diatoms (Bacillariophyceae) are unicellular primary producers occurring in virtually any kind of aquatic environment (Round et al., 1990). As many diatom species have narrow optima along different environmental gradients (Dixit et al., 1992), they are probably the most commonly used group for paleolimnological analyses (Moser et al., 1996). Diatoms are also extensively used as paleo-bioindicators in ecotoxicological research (e.g. review by Rimet, 2012; Debenest et al., 2013 and therein).

The diatoms were prepared using H₂O₂ digestion and HCl (37%) -treatment, with repeated washing in distilled water to clean the material of organic matter and carbonates, and the cleaned diatoms were mounted in Naphrax (Battarbee, 1986). A minimum of 300 diatom valves from each sample were identified and counted along random transects at 1000x magnification. Diatom identification was based mainly on Krammer and Lange-Bertalot (1986, 1988, 1991a, 1991b).

2.4. Cladoceran analysis

Cladocera (Branchiopoda) are microscopic crustacean zooplankton, and are regarded as excellent indicators of environmental change (Jeppesen et al., 2001). The group is used extensively in ecotoxicological tests (Sarma and Nandini, 2006; Lampert, 2011; Suhett et al., 2015), and cladoceran assemblages have been studied in many mining pollution cases in the past (e.g. Bradbury and Megarad, 1972; Melville, 1995; Kerfoot et al., 1999; Belyaeva and Deneke, 2007; Ferrari et al., 2015; Winegardner et al., 2017). In addition, cladocerans have been used in paleolimnological studies for decades (Korhola and Rautio, 2001). The community is comprised of littoral and pelagic species, with a wide range of environmental preferences. The community change thus reflects the changes in environmental conditions (Eggermont and Martens, 2011). In addition, cladocera are on the second level in the food web hierarchy, as they graze on phytoplankton and are preyed upon by invertebrate predators and juvenile fish, making the group important in terms of energy flows in the ecosystem (Eggermont and Martens, 2011).

The cladoceran analysis was conducted following the procedure presented in Korhola and Rautio (2001). In short, the sediment

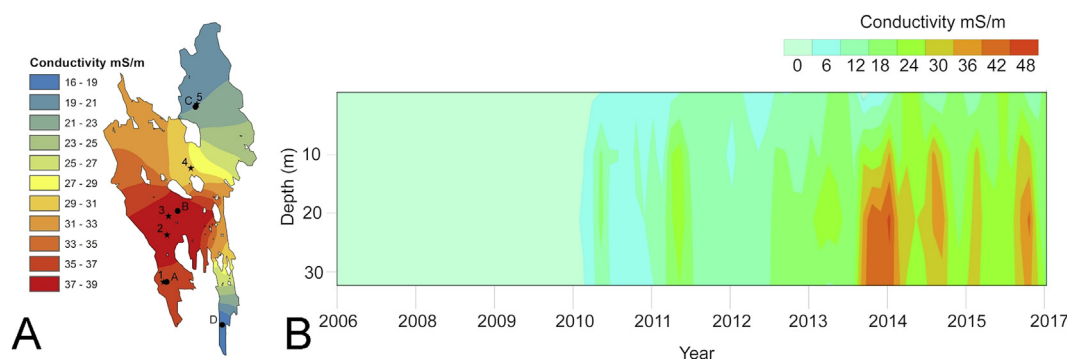


Fig. 2. Interpolated conductivity (Spline –method in ArcMap) values for the post 2012 bottom water averages during the spring (March–May) stratification. Water sampling points = circles, sediment sampling points = stars ($n = 12$). Water depths for the water sampling points are A = 18.5 m, B = 26.5 m, C = 11 m, D = 17 m, and for the sediment sampling points 1 = 16.5 m, 2 = 15 m, 3 = 19 m, 4 = 21.5 m and 5 = 10.5 m (A) Conductivity values representing the whole lake at different depths and different sites between 2006 and 2017 interpolated using kriging –method ($n = 625$) (B).

samples were treated in hot 10% KOH and sieved through a 50 μ m mesh. The residue was stained using safranin, and permanent slides were prepared using a gelatin glycerol jelly. Slides were examined using light microscopy at 100–400x magnification. The minimum number of individuals counted from each sample was set to 100 (Kurek et al., 2010). Identification and nomenclature is based on Szeroczyńska and Sarmaja-Korjonen (2007).

2.5. Chironomidae analysis

Chironomidae (Insecta: Diptera) larvae take part in benthic processes that are essential for lake ecosystem functions (Hölker et al., 2015). Their community compositions typically respond to climate conditions on the regional scale (Brooks, 2006) and limnological factors, such as hypolimnetic oxygen conditions, on the local scale (Brodersen and Quinlan, 2006). Having strict ecological preferences, chironomids have been extensively used in contemporary lake monitoring (Raunio et al., 2011) and paleolimnological assessments of lake reference status (Luoto and Ojala, 2014). Moreover, chironomids have been shown to be very useful in tracking the long-term ecological and environmental consequences of mine pollution in lakes (Thienpont et al., 2016; Stewart et al., 2018).

Standard methods were utilized in the fossil chironomid analysis (Brooks et al., 2007). The wet sediment was sieved through a mesh (100- μ m) and the residue was examined under a stereomicroscope (25x magnification). Larval head capsules were extracted and mounted permanently with Euparal on microscope slides. Taxonomic identification following Brooks et al. (2007) was performed under a light microscope (400x magnification). The minimum chironomid head capsule number per sample was set to 50 (Larocque, 2001; Quinlan and Smol, 2001).

2.6. Numerical analysis

We used kriging interpolation (Surfer 11, Golden Software, LLC) to illustrate the temporal changes in electric conductivity at different sampling depths. In order to illustrate the changes of EC at different depths during the past 11 years, all available records from sampling sites A, B and C (Fig. 2A) since 2006 were used (625 measurements; OIVA database, 2017). Deep water EC data (during spring stratification) retrieved from the OIVA database (2017), was used to illustrate the spatial differences of EC. The spatial variation in EC was illustrated using the splines with the barriers –interpolation method, where the lake shoreline was used as a barrier (ArcMap, version 10.3.1 software, ESRI). Although the

splines and the kriging methods usually produce nearly identical results, the kriging method was used due to its slightly better accuracy (Dubruet, 1984; Laslett, 1994), whereas the splines with barriers-method was used in spatial analysis due to the possibility of including the lake shoreline in the calculation.

All biota counts were converted into percentages and plotted as a stratigraphical frequency diagrams, including only the most common taxa, using the program C2, version 1.7.2 (Juggins, 2007). Detrended correspondence analysis (DCA) and principal component analysis (PCA) were used in order to reveal the compositional changes in the biological data. DCA and PCA were performed using CANOCO for Windows, version 5.01 (ter Braak and Šmilauer, 2007–2012). Dissimilarity index (Bray–Curtis), diversity values (Shannon H'), and rarefied species richness analyses were performed in order to characterize the taxonomical differences between the spatio-temporal samples. Diversity index and species richness were calculated from counted values, whereas the Bray–Curtis similarity matrix was based on proportional data. To test the significance and the magnitude of the difference of biological communities between sample depths (between 1980's, 2010 and 2017), the samples were grouped according to sampling depth and analyzed with ANOSIM (Bray–Curtis dissimilarity index and 999 permutations). The ANOSIM analysis, dissimilarity, diversity and species richness analyses were performed using PAST statistics 3.06 (Hammer et al., 2001).

3. Results

3.1. Mine effluent

Nutrients and pH have remained stable in Lake Jormasjärvi and in the Tuhkajoki River over the study period. In contrast, EC, SO_4 and Na show higher concentrations in post-2008 samples, and illustrate the onset of the mine water input (Fig. 2, Table 1). As the concentration of SO_4 is a major contributor to changes in EC (Supplementary Fig 2), it can be used as a surrogate to assess the spatial and temporal distributions of mine water in Lake Jormasjärvi. The southern sampling points show higher EC levels compared to the northern part of the lake (Fig. 2A; Supplementary Fig 3), and elevated salinity is clearly visible in the deeper water samples (Fig. 2B). The heavy metal concentrations in Lake Jormasjärvi's surface water have remained relatively low, despite the input of the mine water. The average surface water concentrations for mine derived heavy metals, such as Ni and Zn, in the middle of the lake show a clear but relatively small increase, whereas the concentration of Mn exhibits a more pronounced increase. Specifically, the average surface water concentrations for Mn are higher at

the southern sampling site compared to the central and northern parts of the lake (Supplementary Fig. 4).

3.2. Diatoms

The diatoms were relatively fragmented, especially in the topmost samples. At the species level, the most striking feature is the increasing abundance of *Diatoma tenuis* in the topmost samples and the contemporary decrease of the planktonic taxa such as *Lindavia rossii* and *Discostella stelligera* (Fig. 3). This change is strongest in the southern sampling sites, close to the Tuhkajoki River inlet (Sites 1 and 2). For diatoms, the ANOSIM results indicate that the difference between the grouped samples of 0–0.25 cm and 10–11 cm was relatively strong ($r = 0.544$) and statistically significant ($p < 0.01$), and the difference between the samples of 0–0.25 cm and 3–4 cm was weak ($r = 0.12$) and not significant. The difference between the samples of 3–4 cm and 10–11 cm was strong ($r = 0.544$) and statistically significant ($p = 0.011$). The diatom diversity (Shannon H') varied between 2.44 and 3.17, and species richness between 40 and 58. Species richness and diversity show increasing values from the top samples to the bottom samples. They do not, however, show any clear spatial trend. The temporal and spatial variability in diatom assemblages can be seen in Fig. 4A and B and the samples 0–0.25 cm and 3–4 cm at site 1 exhibit the lowest similarity values compared to the other samples. The increasing similarity gradient trajectory from the southern end of the lake to the northern part is disrupted by site 4, with its decreased similarity value for sample of 0–0.25 cm. The same result is also visible in the PCA biplot (Fig. 4C), where the 0–0.25 cm

samples are plotted mainly to the right side of the PCA biplot, the 3–4 cm samples between the top and bottom samples, and the samples of 10–11 cm on the left of the PCA biplot (Fig. 4C).

3.3. Cladocera

Similar to the diatoms, the cladoceran remains were poorly preserved, especially in the surface samples (0.25 cm). In particular, the remains of *Daphnia* were badly degraded, and the counting of individuals was thus not possible. At the species level, the most striking feature is the increasing relative abundance of planktonic *Bosmina longirostris* in the topmost samples (Fig. 5) and the simultaneous decline of *Eubosmina longispina*. This change is strongest in the southern sampling sites, close to the Tuhkajoki River inlet (Sites 1 and 2). In addition, *Rhynchotalona falcata*, *Eurycerus* spp., *Limnospira frontosa*, *Alona rustica*, and *Latona setifera* are all absent in Site 1 surface samples. According to ANOSIM, the difference between the grouped 0–0.25 cm and 10–11 cm samples was strong ($r = 0.684$) and statistically significant ($p < 0.01$), whereas the difference between the 0–0.25 cm and 3–4 cm samples was slightly weaker ($r = 0.364$) but still statistically significant ($p < 0.015$). The difference between the samples of 3–4 cm and 10–11 cm was weak ($r = 0.24$) and not statistically significant. The cladoceran diversity (Shannon H') varied between 0.54 and 1.10, and species richness between 9.4 and 15.1. The temporal and spatial variability in cladoceran assemblages is shown in Fig. 6A and B, as the top samples (0–0.25 cm) at sites 1 and 2, in particular, exhibit the lowest similarity values compared to the other samples. The same result is clearly visible in the PCA biplot (Fig. 6C), where the

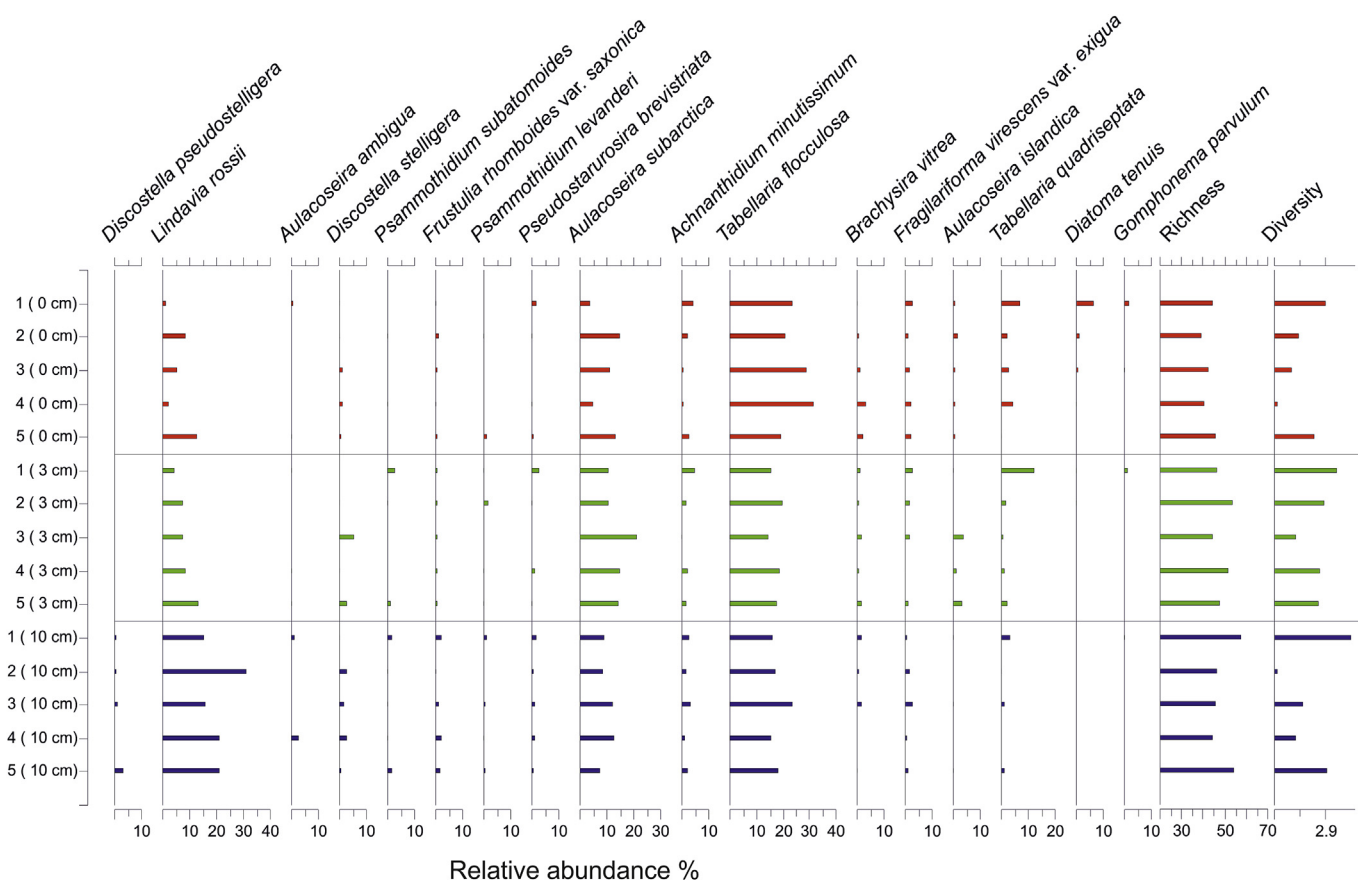


Fig. 3. Diatom stratigraphy. Samples are coded as sampling site number and depth. Red bars indicate samples of 0–0.25 cm, green bars samples of 3–4 cm, and blue bars samples of 10–11 cm. Richness indicates the species richness. Diversity indicates the Shannon H' value. Note the differences in the x-axis scale. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

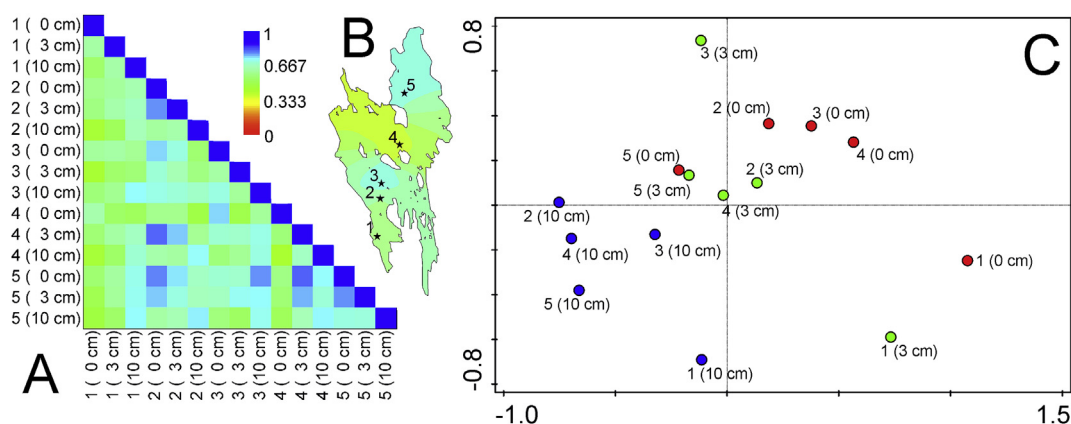


Fig. 4. Diatom similarity matrix between all spatio-temporal samples (A), a spatial presentation of the similarity between the samples 0–0.25 and 10–11 cm of the five sampling sites (B) and a DCA biplot, where the red dots represent the samples of 0–0.25 cm, green dots the samples of 3–4 cm, and the blue dots the samples of 10–11 cm (C). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

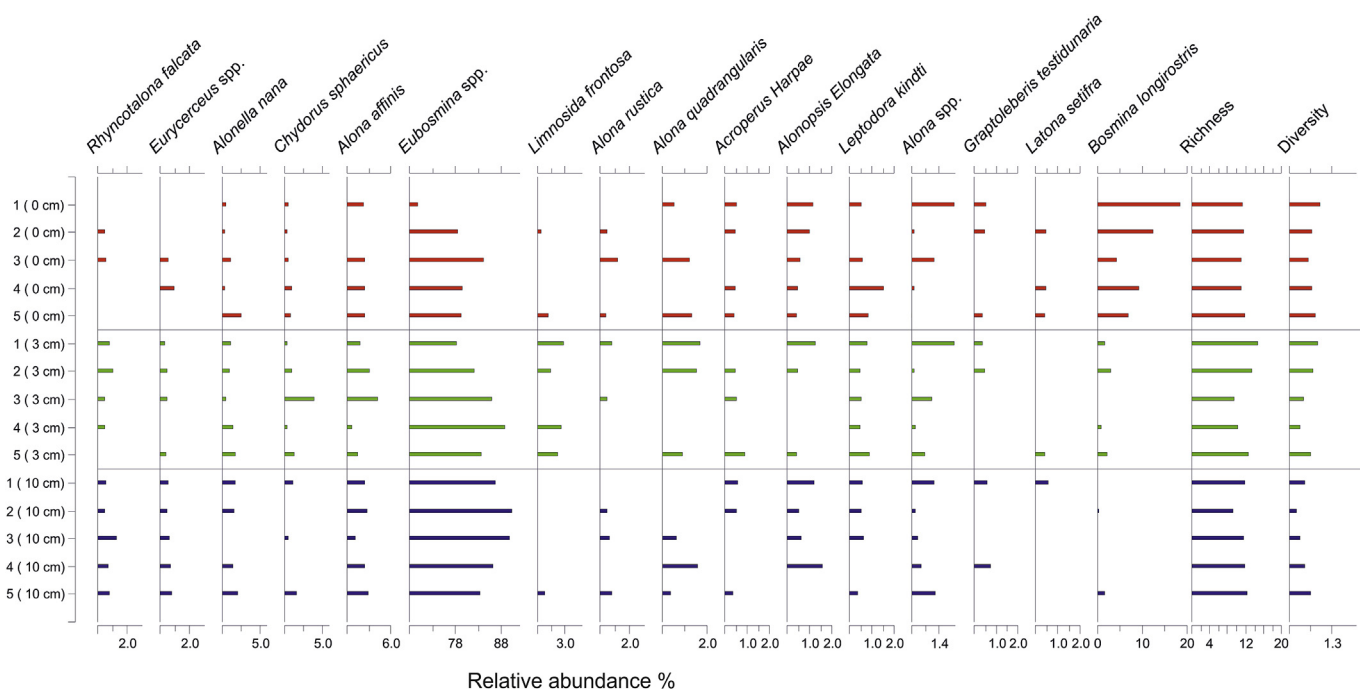


Fig. 5. Cladoceran stratigraphy. Samples are coded as sampling site number and depth. Red bars indicate samples of 0–0.25 cm, green bars indicate samples of 3–4 cm, and blue bars indicate samples of 10–11 cm. Richness indicates the species richness. Diversity indicates the Shannon H' value. Note the differences in the x-axis scale. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

0–0.25 cm samples are plotted to the right side of the PCA biplot, the 3–4 cm samples to the center of the biplot, and the samples of 10–11 cm to the left of the PCA biplot.

3.4. Chironomidae

Similar to the diatoms and cladocerans, the chironomid head capsules were partly fragmented (split in half), yet mostly identifiable. Some of the Tanytarsini could not be identified to their species-type due to the absence of mandibles. Chironomid diversity varied between 1.6 and 2.7 H' and the rarefied taxon richness between 5.0 and 16.0 (Fig. 7). In general, the diversity indices were lower in the 0.25 cm samples. Of the free-living predatory chironomids, the *Ablabesmyia monilis*-type disappeared from the surface sediment samples, while *Procladius* increased. The *Chironomus*

plumosus-type is absent from the samples of 3–4 cm. For Chironomidae, the ANOSIM results indicate that the difference between grouped 0–0.25 cm and 10–11 cm samples was relatively strong ($r = 0.472$) and statistically significant ($p = 0.015$), and the difference between samples 0–0.25 cm and 3–4 cm was strong ($r = 0.532$) and statistically significant ($p < 0.01$) (Fig. 8). The difference between the samples of 3–4 cm and 10–11 cm was weak ($r = -0.06$) and not statistically significant.

4. Discussion

4.1. The impact of mine effluent on water chemistry

The concentration of heavy metals in Lake Jormasjärvi has slightly increased due to the mine waste water (Table 1). However,

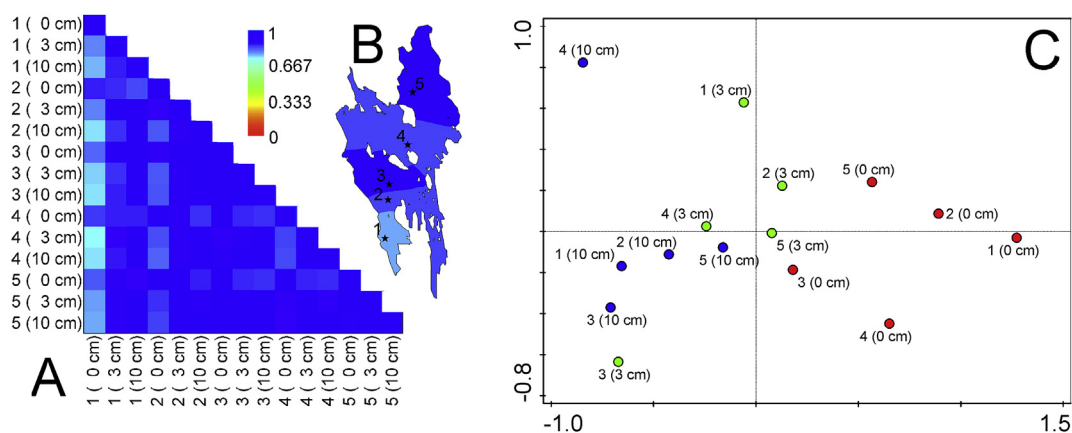


Fig. 6. Cladoceran similarity matrix between all spatio-temporal samples (A), a spatial presentation of the similarity between the samples 0–0.25 and 10–11 cm of the five sampling sites (B) and a PCA biplot, where the red dots represent the samples of 0–0.25 cm, green dots the samples of 3–4 cm, and the blue dots the samples of 10–11 cm (C). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

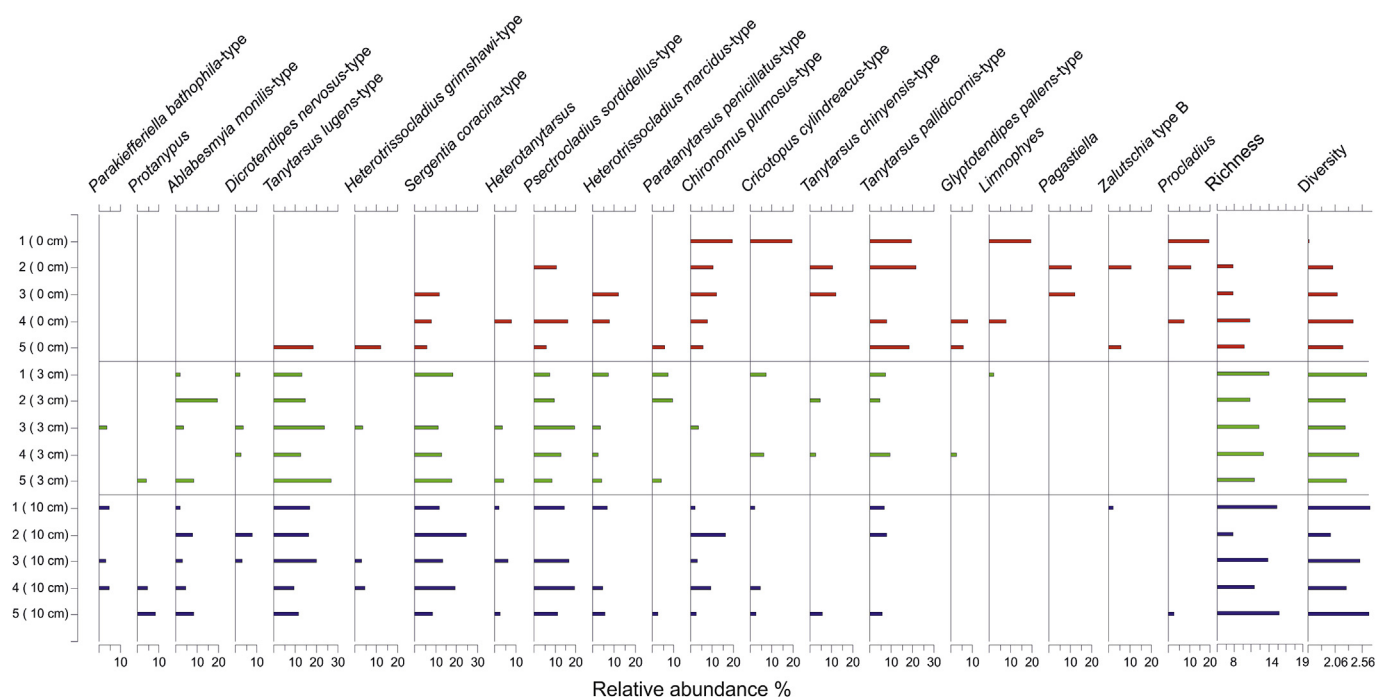


Fig. 7. Chironomid stratigraphy. Samples are coded as sampling site number and depth. Red bars indicate samples of 0–0.25 cm, green bars samples of 3–4 cm, and blue bars samples of 10–11 cm. Richness indicates the species richness. Diversity indicates the Shannon H' value. Note the differences in the x-axis scale. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

the national environmental quality index (EQI) values for Lake Jormasjärvi (22 $\mu\text{g/L}$ for Ni; [ELY, 2017](#)) have been exceeded only twice in the surface water of the mid-lake monitoring site (site B, [Fig. 2A](#)) (31 $\mu\text{g/L}$ and 23 $\mu\text{g/L}$) ([OIVA database, 2017](#)). The relatively minor heavy metal pollution is further demonstrated in the sedimentary studies by [Huuhko \(2016\)](#). In contrast, Mn concentration has increased in the lake water due to the mining activities ([Kauppi et al., 2013](#); [ELY, 2017](#)), and the guideline value for soft waters (73 $\mu\text{g/L}$; [Harford et al., 2015](#)) has been exceeded many times ([Supplementary Fig. 3](#)). However, high concentrations of Mn were already reported from Lake Jormasjärvi before the mine was established ([ELY, 2017](#)), suggesting high natural background concentrations. Average nutrient concentrations are nearly similar in pre- and post-2008 samples. In contrast to metals and nutrients, the elevated major ions concentration of the mine water (SO_4 , Na,

Mg, Ca; presented here as EC) has had a clear impact on the water chemistry of the lake. Thus, EC can be considered as an important surrogate to describe the spatial dispersion of mine water pollution in Lake Jormasjärvi. In addition, EC is regarded as a good overall indicator of toxicity in highly salinated mine waters ([Van Dam et al., 2014](#)). Currently (since 2012), the EC values in the southern part of Lake Jormasjärvi near bottom water are approximately 35–39 mS/m ([Fig. 2A](#)), which should correspond roughly to SO_4 concentrations of 145–175 mg/L ([Supplementary Fig. 2](#)).

4.2. The impact of mine waste waters on biological communities

4.2.1. Diatoms

The clearest impact of the Talvivaara/Terrafame mine on the diatom community can be seen in the topmost samples of sites 1–3.

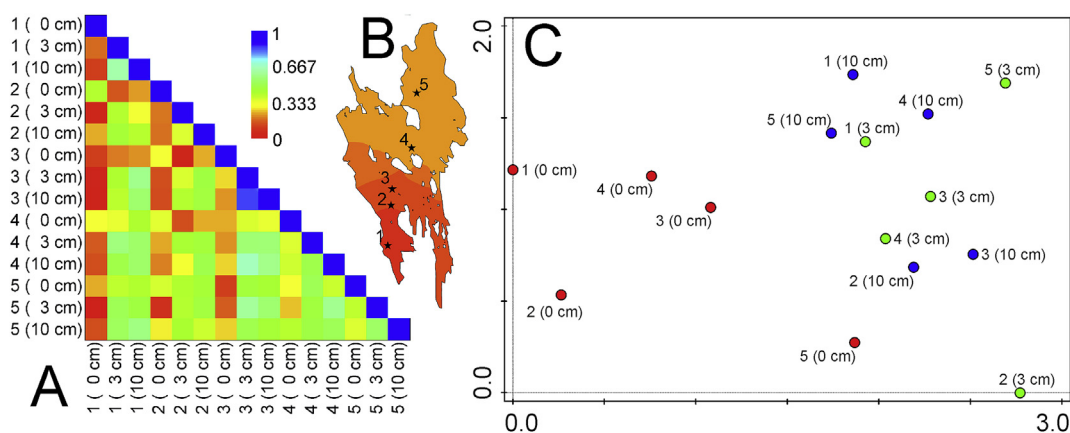


Fig. 8. Chironomid similarity matrix between all spatio-temporal samples (A), a spatial presentation of the similarity between the samples 0–0.25 and 10–11 cm of the five sampling sites (B) and a PCA biplot, where the red dots represent the samples of 0–0.25 cm, green dots the samples of 3–4 cm, and the blue dots the samples of 10–11 cm (C). (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

The striking appearance of *Diatoma tenuis* (relative abundance 7% at site 1; Fig. 3), known to thrive in slightly saline, high conductivity waters (Snoeijs, 1993), is a clear indicator of elevated salinity at sites 1–3. This species is totally absent in the deeper layer, and also at sites 4–5, where salinity seems to have already been diluted to lower levels (Fig. 2). A similar, but much clearer change (abundances of *Diatoma tenuis* up to 70% in the surface sediment samples) was observed in Lake Kivijärvi (located along the southern route of mine water), and was also impacted by the Talvivaara/Terrafame mine (Leppänen et al., 2017). Another striking feature is the decrease of planktonic species, especially *Lindavia rossii*, when moving from the bottommost samples towards the modern samples. *Cyclotella sensu lato* (includes the revised taxa *Discostella stelligera*, *Discostella pseudostelligera* and *Lindavia rossii*; Fig. 3) frequently thrive in low-productivity, oligo-to mesotrophic lakes (e.g. Willén, 1991). The decrease of these taxa might also be caused by a slight eutrophication, as suggested by Mäkinen and Kauppila (2006), and possibly by the cladocera and chironomid assemblages referenced in this study. However, the increase of the acidophilous/acidobiontic diatom *Tabellaria quadrisepata* (Renberg, 1990), and the general ecology of the most common diatom taxa in the most recent samples, indicates a slightly acid and oligotrophic environment - which is in line with the current measurements of pH (6.1), total phosphorus (11.6 µg/L), and total nitrogen (401 µg/L). The salinity gradient and diatom community change trajectory from sampling point 1 to 5 is broken by site 4, due to the mixing zone of the saline waters from the south with the runoff waters from the western catchment, impacted by the talc mine and forestry activities. In summary, based on the known ecological preferences of the diatom taxa encountered, the most significant changes in the diatom communities can be interpreted as to have been caused by the mine-derived salinity increase, and neither by the increase in toxic metal concentrations nor elevated nutrient concentrations.

4.2.2. Cladocera

The cladoceran community changes between 4 cm and 10 cm are relatively minor, with Bray-Curtis dissimilarity values varying between 0.86 (site 1) and 0.93 (site 5). The slight eutrophication, which was detected by Mäkinen and Kauppila (2006), seems to have had only a minor effect on cladoceran communities in samples from 10–11 cm to 3–4 cm, due to the nearly unnoticeable increase of the eutrophic species *B. longirostris* (Bjerring et al., 2009) in the samples of 3–4 cm. According to the Bray-Curtis similarity index, the most pronounced change between the depth of 3–4 cm and the

sediment surface sample was recorded at sampling point 1, followed by sampling points 4, 2, 3, and 5. The clearest change at the species level is the increasing abundance of *B. longirostris*, especially at sampling point 1, followed by sampling points 2, 4, 5, and 3. This result is not surprising, as sampling point 1 is closest to the inlet of the Tuhkajoki River, receiving the largest load of the mine effluent. Similar to the diatoms, the salinity gradient and cladoceran community change trajectory from sampling point 1 to 5 is broken by site 4, which exhibits lower similarity values than site 3. This difference is most probably related to the effluent originating from the closed Lahnaslampi mine (Fig. 1) and the intense forestry activities in the western part of the catchment. The location of this site is somewhat exceptional, as the runoff waters from the western catchment are mixed with the saline waters from the south. Cladocerans exhibit a wide variability in salinity tolerance (Aladin and Potts, 1995), and shifts in salinity induce changes in species assemblages (Aladin, 1991). Planktonic *E. longispina*, which has declined in surface samples, is usually present in dilute waters in Europe (Bjerring et al., 2009), and it dominates subarctic and low-conductivity lakes in Finland (Korhola, 1999). In contrast, *B. longirostris* is known to tolerate elevated salinity (e.g. Canton and Ward, 1981; El-Bassat and Taylor, 2007; Ferrari et al., 2009; Adamczuk, 2016; Zawisza et al., 2016). Because nutrient concentrations have not changed during the recent decade, the increasing abundance of *B. longirostris* in the surface samples is probably not related to nutrient status. Even though Mn concentrations in Lake Jormašjärvi have continuously exceeded the protection guideline values, *B. longirostris* is more sensitive towards metals pollution (e.g. copper; Koivisto et al., 1992; Koivisto and Ketola, 1995) than many other cladoceran species, and hence should not be able to increase in abundance if metal pollution was the ecologically most important component in the mine water. In addition, the chronic reproductive-inhibition IC(50) value for cladoceran *Ceriodaphnia dubia* for soft water is 3900 µg/L Mn (Lasier et al., 2000), and 1100 µg/L for *Moinodaphnia macleayi* (Harford et al., 2015). However, because toxicity tests regarding the Mn tolerance of *E. longispina* and *B. longirostris* have not yet been conducted, the role of Mn in this case is difficult to assess. A similar shift between *E. longispina* and *B. longirostris* has been detected in Lake Kivijärvi, also impacted by saline and heavy metal contaminated mine water from the Talvivaara/Terrafame mine (Leppänen et al., 2017). The impact of mine water discharge in Lake Jormašjärvi can also be seen in the littoral cladoceran community. For example, *R. falcata* and *L. frontosa*, which prefer oligotrophic and dilute waters (Bjerring et al., 2009), are absent in the surface sediment samples of the

southern sampling point. However, the SO_4 impact is probably not related to the toxicity of SO_4 , because the SO_4 concentrations in Lake Jormasjärvi are clearly below any levels where harmful effects have been reported (Mount et al., 1997). Moreover, SO_4 is considered to be an impermeant anion to the gills of freshwater animals (Griffith, 2017). Thus, the negative impacts of SO_4 is most probably connected to other salinity related interferences. For example, the SO_4 may affect the feeding efficiency of cladocerans (Stankovic et al., 2011; Soucek, 2009). Interestingly, Sivula et al. (2018) found that the Tuhkajoki River water was not acutely toxic for *Daphnia magna*, and questioned whether the acute *Daphnia* test is sensitive enough to evaluate the risks of mine water to aquatic organisms. Our results support this idea, since the species change is evident at the sampling site, which is located even further downstream.

4.2.3. Chironomidae

The most striking change in the chironomid assemblages is the disappearance of the *Ablabesmyia monilis*-type from the top samples, and the simultaneous increase in another predatory Tanytopodinae taxon, *Procladius*. In Finnish lakes, the *Ablabesmyia monilis*-type has an oligo-mesotrophic nutrient preference, whereas *Procladius* is most often encountered in eutrophic sites (Raunio et al., 2011; Luoto, 2011). However, since the data from diatoms and cladocerans do not suggest significant nutrient enrichment in Lake Jormasjärvi, the shift in Tanytopodinae taxa is likely related to other environmental forcing. In fact, these taxa also have distinct difference in their oxygen tolerances, as *A. monilis*-type demands higher oxygen availability, whereas *Procladius* is known to tolerate even temporary anoxia (Brodersen and Quinlan, 2006). In addition, *Procladius* has a higher salinity tolerance compared to other Tanytopodinae (Dickson et al., 2014), which might be part of the reason behind its increasing relative abundance in the samples from Lake Jormasjärvi. Although the species-specific ion- and osmoregulation of chironomid larvae at different salinities is not well-known, they clearly have differentiating tolerances for given salinity levels (Eggermont et al., 2006).

Another distinct feature of the chironomid stratigraphy is the absence of the *Chironomus plumosus*-type in all the 3–4 cm samples, except for sampling point 3. Similar to *Procladius*, the *C. plumosus*-type prefers oxygen-depleted lakes (Brooks et al., 2001), and these taxa are also known to be the most tolerant of metal contamination, including Mn, Ni, and Zn (Swansburg et al., 2002; Ilyashuk et al., 2003; Brooks et al., 2005), as well as increased salinity (Thienpont et al., 2015). Hence, in general, they thrive in lakes experiencing environmental perturbations. Although the decrease in the *C. plumosus*-type in the 3–4 cm samples is difficult to explain, it seems clear that the mine impacts are most clearly reflected by the *C. plumosus*-type, and especially *Procladius*, with a simultaneous decline in environmentally sensitive taxa such as the *A. monilis*-type and *Tanytarsus lugens*-type. These patterns are most clear in sampling points 1–4 (Fig. 7).

For chironomids, it appears that there are both significant temporal and spatial community differences (Fig. 8). The top samples clearly differ from the lower samples at all the sampling points, however, with a gradually decreasing community change from sampling point 1 to sampling point 5. This suggests that the benthic ecosystems in Lake Jormasjärvi have been as affected as the planktonic ecosystems, which are more reflected by the diatoms and cladocerans. Nonetheless, a similar breaking of the community change trajectory by sampling point 4, as discovered for the diatom and cladoceran data, is not found for chironomids.

4.3. Spatio-temporal trends

The planktonic habitat of Lake Jormasjärvi has undergone clear

changes during recent years, especially in the southern sites near the inlet of the Tuhkajoki River. This can be seen in a decline in the relative abundances of planktonic diatoms, and a shift in the planktonic cladoceran community. In addition, the most pronounced shift in species composition among the littoral diatom, cladoceran, and chironomid communities can also be seen in the southern sites. The continuous loading of salinated mine water has caused a general spatio-temporal pattern, which can be seen as a species shift from south to north and from past to present. The spatial trend, highlighted by the paleobioindicator groups described here, is in good agreement with modern measured EC values, whereas the temporal differences are clearly linked with the start and continued operation of the Talvivaara/Terrafame mine. These findings are important, considering that Lake Jormasjärvi is the fourth lake in the chain of lakes, thus receiving significantly lower amounts of mine water –contaminated waters than the upstream lakes. Despite this, there is almost a ten-fold increase of EC in the southern part of the lake near the river inflow and around four-fold increase in the northern part of the lake near the outflow (Supplementary Fig. 3). Even low salinity values and moderate increase in water salinity have been noted to have negative impacts on freshwater biota (Schallenberg et al., 2003). Our findings highlight the destructive potential of saline effluents in fresh water ecosystems (e.g. Cañedo-Argüelles et al., 2013; Dugan et al., 2017; Kaushal et al., 2018), and the magnitude of damage induced by salinated mine water discharges to watercourses (e.g. Zgórska et al., 2016).

5. Conclusion

Our spatio-temporal study has shown that the Talvivaara/Terrafame mine has had a pronounced impact on the aquatic ecosystem of Lake Jormasjärvi. Although the salinity and metals concentrations are reduced along the chain of lakes, a clear change can be seen in the biological communities of Lake Jormasjärvi, both in space and time. This change can be interpreted as to be mainly caused by the increased water salinity, being highest at the inflow site and lowest at the outflow site. The impact of the salinated mine water is similar among the different species groups, suggesting a food-web wide change in Lake Jormasjärvi. Our study highlights the strong impact of modern large and low-grade mines on the surrounding aquatic ecosystems. Clear changes in aquatic chemistry and in multiple biological communities can be observed even in the relatively large Lake Jormasjärvi.

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Appendix A. Supplementary data

Supplementary data related to this article can be found at <https://doi.org/10.1016/j.envpol.2019.01.111>.

Declarations of interest

None.

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