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Waste water discharge from a large Ni-Zn open cast mine degrades benthic integrity of Lake Nuasjärvi (Finland)

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Abstract

The Talvivaara/Terrafame multi-metal mining company is Europe's largest nickel open cast mine, it is also known for the largest wastewater leakage in the Finnish mining history and a series of other accidents. In this paleolimnological study, influences of a recently constructed treated waste water discharge pipeline into Lake Nuasjärvi were investigated by analyzing past (pre-disturbance) and present community compositions of key aquatic organism groups, including diatoms, Cladocera and Chironomidae, along spatial (distance, water depth) gradients. In addition to defining ecological changes and impacts of saline mine waters in the lake, chironomids were used to quantitatively reconstruct bottom water oxygen conditions before and after the pipe installation (in 2015). The diatom and cladoceran communities, which reflect more the open-water habitat, showed only relatively minor changes throughout the lake, but a general decrease in diversity was observed within both groups. Chironomids, which live on substrates, showed more significant changes, including complete faunal turnovers and deteriorated benthic quality, especially at the sites close to the pipe outlet, where also chironomid diversity was almost completely lost. Furthermore, the reconstructed hypolimnetic oxygen values indicated a major oxygen decline and even anoxia at the sites near the pipe outlet. The limnoecological influence of the pipe decreased at sites located counter-flow or behind underwater barriers suggesting that the waste waters currently have location-specific impacts. Our study clearly demonstrates that whereas the upper water layers appear to have generally maintained their previous state, the deep-water layers close to the pipe outlet have lost their ecological integrity. Furthermore, the current hypolimnetic anoxia close to the pipe indicates enhanced lake stratification caused by the salinated mine waters. This study clearly exhibits the need to investigate different water bodies at several trophic levels in a spatiotemporal context to be able to reliably assess limnoecological impacts of mining.

Keywords: Chironomidae; Cladocera; diatoms; mine impact; paleolimnology

Capsule: Stronger stratification due to salinated mine waters has caused diminished water column mixing, enhanced oxygen deficiency and increased deterioration of benthic communities.

1. Introduction

Environmental impacts of mining include erosion, contamination of soil, surface water and groundwater and ecosystem degradation. Water contamination has a serious influence on limnology of the affected lakes and their aquatic communities including micro-organisms, such as diatom (Bacillariophyta) algae, and macro-organisms, such as cladoceran (Crustacea: Cladocera) zooplankton and chironomid (Diptera: Chironomidae) macrobenthos (Doig et al., 2015; Thienpont et al., 2016). The sedimentary remains of these organisms also preserve well as (sub)fossils in lake bottom deposits enabling their use in tracking catchment originated pollution histories through stratigraphic paleolimnological analyses (Cohen, 2003; Smol, 2008). In particular, diatom assemblages (primary producers) tend to reflect the epilimnetic water quality (pH and nutrients) (Weckström et al., 2003; Tammelin et al., 2017) and cladocerans (secondary producers) the general limnoecological status (trophic state, pollution) of the water column (Jeppesen et al., 2011; Leppänen, 2018). In turn, the bottom-dwelling aquatic larval stages of chironomids have direct species-specific metabolic responses to the bottom water (hypolimnetic) dissolved oxygen concentration (Quinlan et al., 1998; Luoto et al., 2017) with some particular species having hemoglobin (red larvae known as "bloodworms") to survive under periods of anoxia (Brodersen et al., 2008).

Epilimnetic contamination in lakes is dependent on the characteristics of pollution, residence time and on whether the pollution is persistent (Abel, 2014). The same situation as in the surface waters prevails in deeper water parts, with the exception that the residence time is typically much longer. Often associated with pollution, lakes tend to become stratified isolating the hypolimnetic water body and trapping contaminants to the bottom water and sediments. For example, mining related salinization due to high amounts of dissolved ions, such as sulphate, in wastewater is known to cause artificial stratification in lakes (Williams, 2001). Therefore, sedimentdwelling organisms at the lake bottom, such as most chironomids, are ecotoxically highly susceptible to mining impacts (Kansanen et al., 1985). Moreover, dissolved oxygen is a fundamental descriptor of ecosystem status, particularly in the benthic environment. Longer periods of oxygen depletion, i.e. anoxia, promote phosphorus release from the sediments (Krogerus and Ekholm, 2003) and increases the chemical cycling, which has serious ecological consequences including community reorganizations (Rogora et al., 2018). Therefore, benthic ecological integrity, which refers to the ability of deep water ecosystems to support and maintain biotic and abiotic processes and diverse communities (Carignan and Villard, 2002; Poikane et al., 2016), plays a key role when describing the status of aquatic ecosystems.

In Finland, the most prominent individual environmental case during the past years has been related to the pollution impacts of the Talvivaara/Terrafame mine, of which Ni-Zn-Cu-Co ore deposit holds the largest sulfidic Ni resource under exploitation in western Europe (Kontinen and Hanski, 2015). The enormous open cast Ni mine in Sotkamo (Kainuu region) was established in 2008, but has suffered from repeated leaks of treated and untreated mine waters to the environment. The nearby lakes have been exposed to sodium sulfate (Na_2SO_4) and metal contamination due to waste waters originating from the enrichment procedure (Kauppi et al., 2013) including larger lakes Kivijärvi, Jormasjärvi and Nuasjärvi (Mäkinen et al., 2010; Leppänen et al., 2017). Although Nuasjärvi is further downstream from the mine compared to Jormasjärvi, it has recently become of significant interest due to the construction of a treated waste water pipeline in 2015 directly from the mining district to the lake basin.

The aim of this study is to investigate the limnoecological conditions in Lake Nuasjärvi using paleolimnological methods and the top-bottom approach, where the surface sediment samples ("top") represent the present and the downcore samples ("mid" and "bottom") the past (e.g. Michelutti et al., 2001; Weckström et al., 2003). We analyze diatom, cladoceran and chironomid assemblages to build a holistic understanding of aquatic ecosystem changes including epilimnetic and hypolimnetic water bodies at the autotrophic and heterotrophic productivity levels. A special focus of the research is on the potential hypolimnetic degradation, and hence we also utilize chironomid assemblages in reconstructing long-term hypolimnetic oxygen conditions in the lake using two approaches; the Benthic Quality Index (BQI) (Jyväsjärvi et al., 2010) and the quantitative transfer function method (Little and Smol, 2001). With these analyses, we aim to provide novel knowledge on present and pre-impact ecological and limnological conditions of Lake Nuasjärvi that can be useful for local and regional environmental management, but can also provide a greater general understanding on how aquatic ecosystems respond to mine pollution at different habitats and productivity levels.

2. Material and methods

2.1. Study site

Nuasjärvi (64º10'N, 28º03'E; 138 m a.s.l.) is a mildly acidic (~6.4), large (surface area 96 km^3, shoreline length 171 km) and deep (maximum depth 44 m, average depth 8.5 m) lake located in the municipality of Sotkamo (population of ~10,000) in the Kainuu region, Finland. The village of Vuokatti, which is in a characteristic forested hill range, borders the lake on its eastern side. The city of Kajaani (population of ~37,000) is located on the western shore of the Rehja basin, which is separated from the Nuasjärvi basin by the strait Rimpilänsalmi (Fig. 1). The Rehja-Nuasjärvi lake complex (better known as Nuasjärvi) is part of a larger chain of lakes that continues through Lake Oulujärvi to the Gulf of Bothnia. Of the total of 187,888 lakes (>0.05 ha) in Finland (Raatikainen and Kuusisto, 1990), Rehja-Nuasjärvi is the 46^{th} largest. Its main inlets are located in the northeast and south of the Nuasjärvi basin and the main outlet is in the western side of the Rehja basin flowing through the city of Kajaani. The mean water flow from the east to the west is 88 m^3s^{-1} (23-

230 m^3s^{-1}) (Finnish Environment Institute). The mean annual air temperature (climate normal 1981-2010) in the area (Kajaani) is 1.8 °C and precipitation is 598 mm (Finnish Meteorological Institute, https://en.ilmatieteenlaitos.fi/climate). The lake is typically frozen from December to April.

Land use-related erosion and transport of particulate matter into Lake Nuasjärvi started as early as in the seventeenth century (Mäkinen et al., 2010), but there has been no significant increase in nutrient levels during the last 30 years (Fig. 2). The catchment bedrock consists in major part of black shale, which is rich in sulfides and metals (Loukola-Ruskeeniemi, 1996). Owing to the bedrock composition, the sulfur and metal (e.g. Ni) concentrations are locally elevated (Geological survey of Finland, https://hakku.gtk.fi/en). In the late 1960s, a talc mine (Lahnaslampi) began to operate on the southern side of Lake Nuasjärvi and continued until 2010. A discharge pipe from the now closed mine was used to direct treated waste water to the nearby southern bay of Nuasjärvi until 2010, after which there has not been waste water load from the Lahnaslampi mining district. With the exception of elemental (e.g. S, Ni) mobility and sedimentation, the geochemical impacts of the Lahnaslampi mine on Lake Nuasjärvi have been relatively minor (Mäkinen et al., 2010).

Further south, the Talvivaara mine (bioheapleaching plant) began to operate in 2008 (Riekkola-Vanhanen, 2013), but has experienced consecutive serious leaks of both treated and untreated mine water and polluted the nearby lakes with contaminants derived from the black schist ore and enrichment procedure, such as highly soluble Na_2SO_4, Ca, Mg, Fe, Mn, Ni and Zn. The treated waste water amounts allowed by the environmental permit have been clearly exceeded over several years. Treated waste waters have been directed through a direct pipeline from the mining district to Nuasjärvi since November 2015. In 2016, an annual amount of 13,600 t of sulfate from the treated mine waste water was directed through the pipeline, whereas additional 2,000 t of sulfate was transported through Lake Jormasjärvi into Lake Nuasjärvi. Consequently, the lake water sulfate concentrations increased between 2015 and 2016 from 7 to 15 (up to 200) mg l^{-1} on average

(Mäkinen, 2017). The Talvivaara (nowadays Terrafame) case has been described as one of the most significant environmental disasters in Finland being under major political debates due to its economic and environmental impacts (e.g. Sairinen et al., 2017). During the recent years, the surface water contamination has been controlled by filtration water cleaning operations and hydrological rearrangements. The present water quality of Lake Nuasjärvi is classified as “good” by the Finnish Environment Center. Although there has been no observed increase in lake productivity (total phosphorus), a clear increase in electric conductivity in Lake Nuasjärvi has been monitored after the installation of the mine waste water discharge pipeline (Fig. 2.). Despite the fresh surface waters deriving from the Sotkamo water route (eastern inlet in Fig. 1) and the general flow direction from the east to the west, according to measurements, in-lake bottom currents carry the waste waters also in other directions causing elevated electric conductivity and sulfate concentrations in all deep basins near the pipe outlet (Mäkinen, 2017).

2.2. Sediment samples

Eight short sediment profiles (sites N4, N7, N8, N10, N11, N13 and N16) were retrieved in August 2017 from different parts of Lake Nuasjärvi using a HTH kayak corer (Renberg and Hansson, 2008). Six of the profiles originate from the main Nuasjärvi basin and one (N4) from the Rimpilänsalmi strait located between the Nuasjärvi and Rehja basins (Fig. 1). The water depth of the sampling sites varies between 14 and 29 m (Fig. 1). From each sediment core three samples were extracted covering the “early-impact” conditions (10-11 cm), pre-waste water pipeline conditions (3-4 cm) and the present (0-0.25 cm). According to known sedimentation rates of 0.1-0.2 cm/y (Mäkinen and Kauppila, 2006), these time periods correspond tentatively to the early 20th century (10-11 cm), the turn of the Millennium (3-4 cm) and 2016-2017 (0-0.25 cm). Since the lowermost reference samples do not extend as far into the past to represent a period without any human influence, these samples do not represent true natural reference conditions, but they do represent conditions prior to mining operations in the catchment (including the Lahnaslampi mine) and are discussed herein as "reference" samples. It should also be noted that since the reference samples are not chronologically established, there may be significant variation in their actual ages.

2.3. Diatom analysis

Samples for diatom analysis were prepared using H_2O_2 digestion and HCl (37%) -treatment with repeated washing in distilled water to clean the material of organic matter and carbonates (Battarbee, 1986). The cleaned diatoms were mounted in Naphrax. 300 diatom valves were set as the minimum count from each sample. Diatoms were counted along random transects and identified at 1000x magnification based mainly on Krammer and Lange-Bertalot (1986, 1988, 1991a, 1991b).

2.4. Cladocera analysis

Cladocera samples were prepared following the procedure described in Korhola and Rautio (2001). Subsequently, the sediment samples were treated with hot KOH (10%) and sieved through a 50 µm mesh. The residue was stained using safranin and permanent slides were prepared using a gelatin glycerol jelly. Identification of the specimens under a light microscope (100-400x magnification) was based on Szeroczyñska and Sarmaja-Korjonen (2007). The minimum count size of individuals was set to 100 per sample.

2.5. Chironomid analysis

Standard methods were applied in fossil chironomid analysis (Brooks et al., 2007). The wet sediment was sieved through a mesh (100-µm) and the residue was examined under a stereomicroscope (25x magnification) for larval head capsule extraction using a target counting sum of 50 per sample. The head capsules were mounted permanently with Euparal on microscope slides for taxonomic identification (according to Brooks et al., 2007) under a light microscope (400x magnification).

2.6. Numerical techniques

Similarity analysis using the Bray-Curtis dissimilarity measure was done on relative taxon abundances to indicate divergence between the intervals. In the analysis, the value of 1 indicates a fully similar assemblage, whereas the value of 0 indicates a fully dissimilar assemblage. As a diversity measure, Shannon index (H´) and the rarefaction technique were applied on counted absolute abundances. The similarity and diversity analyses were performed using the program Past3 (Hammer et al., 2001).

Minimum hypolimnetic dissolved oxygen (DO) was reconstructed using a 30-lake chironomid-based calibration model for Finland (Luoto and Nevalainen, 2011; Luoto and Salonen, 2010; Luoto et al., 2019). The calibration sites range from anoxic (O_2 = 0.5 mg l^{-1}) to hypersaturated sites (O_2 = 18.1 mg l^{-1}). The weighted averaging partial least squares (WA-PLS) model has a coefficient of determination (leave-one-out cross-validation) of 0.74 and a root mean squared error of prediction of 2.3 mg l^{-1}. For detection of impact, also the Benthic Quality Index (BQI) was applied. The BQI uses the varying tolerance of different species of chironomids to hypolimnetic oxygen levels (Wiederholm, 1980; Johnson, 1998). The BQI varies between 0 (no indicator taxa present = anoxic) and 5 (high hypolimnetic oxygen availability), and here the biological indicator metrics by Johnson (1998) were used. In the context of lake trophic state, BQI 0 indicates hypertrophy, BQI 1-2 eutrophy, BQI 3 mesotrophy, BQI 4 oligotrophy and BQI 5 ultraoligotrophy (Luoto and Raunio, 2011). In this study, we consider the BQI to reflect hypolimnetic oxygen conditions, for which chironomids have a direct metabolic response (Brodersen et al., 2008). The BQI is suitable to be used in the current study, since it is particularly designed for deep boreal lakes (Wiederholm, 1980). To examine the relationship between inferred oxygen and BQI, we used the Pearson product-moment correlation (R), correlation coefficient (R^2) and level of statistical significance (p).

3. Results

The most abundant diatoms in the Lake Nuasjärvi samples included planktonic *Aulacoseira* species *A. ambigua* (2.4-33.4%), *A. islandica* (0.6-19.7%) and *A. subarctica* (6.0-26.0%), together with *Tabellaria flocculosa* (4.5-14.8%). The *Aulacoseira* species were less abundant at sampling point N4 (Fig. 3) taken from the shallower strait (Fig. 1). *A. ambigua* and *A. islandica* showed generally increasing abundance towards the top samples, whereas *A. subarctica* did not display a distinct coherent directional shift. *T. flocculosa* showed a slight but consistent decrease in abundance. Rarefied richness of diatoms varied from 24 to 59 and the Shannon index from 2.1 to 3.4. At most sampling sites, diatom diversity was highest in the lowermost “reference” samples (Fig. 3). Similarity analysis revealed that the most dissimilar surface (0-0.25 cm) diatom sample compared to the “reference” sample (10-11 cm) was at sampling point N11 (Bray-Curtis 0.47), whereas the most similar samples were at N7 and N8 (Bray-Curtis 0.60-0.75) (Fig. 4).

All Cladocera samples were dominated by planktonic *Eubosmina* spp. (68.7-83.3%). Less abundant, but regularly occurring cladocerans included *Bosmina longirostris* (0.5-5.7%), *Chydorus sphaericus* (1.2-12.0%) and *Alonella nana* (0.7-6.7%). The cladoceran assemblages did not exhibit a clear spatiotemporal development, but in general, diversity was lower in the surface samples with the exception of sampling point N11 (Fig. 5). Rarefied cladoceran richness varied between 7.6 and 13.4 and the Shannon index between 0.6 and 1.3. In all, the cladoceran surface and "reference" samples were very similar (Bray-Curtis >0.8), but sampling points N11 and N4 showed nonetheless some minor changes in the community composition (Fig. 6).

None of the chironomid taxa encountered in Lake Nuasjärvi occurred in every sample. The most common taxa, which tolerate low oxygen conditions, were *Sergentia coracina*-type (mean abundance 10.7%), *Procladius* (9.9%) and *Chironomus plumosus*-type (8.8%). Especially *Procladius* had the tendency of increasing in the surface samples (Fig. 7). The most abundant taxa requiring high oxygen concentrations were *Psectrocladius sordidellus*-type (9.7%) and *Tanytarsus lugens*-type (8.7%), which generally decreased in the surface samples. In addition to chironomid midges, mandibles of the phantom midge *Chaoborus flavicans* occurred at all the sites except N4, though with low abundance. Rarefied richness of chironomids varied from 3.9 to 24.0 and the Shannon index from 0.9 to 3.0. Chironomid diversity distinctly decreased in the surface samples of sites N7, N8, N10 and N16 (Fig. 7). Unlike diatoms and especially cladocerans, chironomids showed major changes based on the similarity analysis (Fig. 8). Sampling point N8 was completely different (Bray-Curtis 0.07) in its composition and also N7 and N16 (Bray-Curtis <0.21) were distinctly dissimilar. Even at furthest sampling point N4, similarity remained low (Bray-Curtis 0.39).

All chironomid taxa found from the Lake Nuasjärvi samples also occurred in the chironomid-oxygen calibration set. The reconstructed hypolimnetic oxygen conditions showed variation from anoxia in the surface sample of site N8 (0.3 mg l^{-1}) to hypersaturation in the lowermost sample of site N13 (13.5 mg l^{-1}). These same samples also had the lowest (0) and highest (3.7) BQI suggesting permanent anoxia and good water quality, respectively. With the exception of site N11, all the sites had their lowest reconstructed oxygen concentration in their surface samples (Fig. 9). Lowest modern oxygen conditions (anoxia to poor oxygenation) were reconstructed for sites located close to the waste water discharge pipe. Considering the reference samples, all sites had moderate to well-oxygenated "reference" conditions (O^2 >4 mg l^{-1}). Similar to the oxygen reconstructions, the BQI was lower in the surface samples of most of the sites, except N4, N11 and N16. The lowest BQI of 0 was at N8. The BQI generally suggested moderate "baseline" status (2.2-3.3), but at site N13 the BQI of the lowest sample was high (3.7). The reconstructed hypolimnetic oxygen values showed statistically significant correlation with the BQI having R of 0.72, R^2 of 0.51 and $p<0.001$.

4. Discussion

4.1. Planktonic communities

The largest community shifts between the surface and "reference" samples in phytoplankton and zooplankton, i.e. diatoms and cladocerans, occurred at sampling site N11 (Figs 4, 6). This sampling site is located closest to the waste water discharge pipe. The ecological shift at N11 is reflected in diatoms with replacement of taxa such as *Lindavia rossii*, a taxon found to prefer oligotrophic and low conductivity lakes (Håkansson, 1990; Cremer et al., 2001), with other planktonic taxa such as *A. ambigua*, *A. islandica* and *A. subarctica* (Fig. 3). These trends were also present at several other sampling sites, together with a general decrease in *T. flocculosa*, which has a low pH optimum in northern Finland (Rantala et al., 2017). In cladocerans, the predominant planktonic taxon *Eubosmina* spp. slightly decreased at N11, whereas *C. sphaericus* increased (Fig. 5). *C. sphaericus* is a resilient taxon (Koff et al., 2016) and has been often found to increase under environmental perturbation, such as eutrophication (Nevalainen and Luoto, 2013) and mining pollution (Leppänen et al., 2018). Although usually considered as a littoral cladoceran, *C. sphaericus* can thrive in the plankton of eutrophic lakes as well (Manca et al., 2007). In shallow boreal lakes, *C. sphaericus* is assigned to the eutrophic/bad limnoecological integrity group with significant relationship with total phosphorus, whereas *Eubosmina* spp. is in the oligotrophic/good group and associated mainly with elevated water depth (Luoto et al., 2013). The diatom and cladoceran evidence may hence suggest an increase in nutrient conditions of the surface waters, although these changes are not overwhelmingly large. Although increased lake trophic conditions in Nuasjärvi were also suggested by a previous study (Mäkinen et al., 2010), the water quality monitoring data nonetheless shows no increase in total phosphorus (Fig. 2), evidencing that the epilimnetic trophic status has not changed. Instead of actual nutrient response, these phyto- and zooplankton community changes may represent an eutrophication-like response to climate change (similar community response) that has been observed from other large and stratified European lakes (Visconti et al., 2008; Noges et al., 2011). Moreover, the relatively minor compositional shifts in cladocerans throughout the basin are not directionally uniform (Fig. 5), since *Eubosmina* spp. increase and *C. sphaericus* decrease at several sampling sites, such as N7 and N8, which are close and downstream (considering surface flow) from the discharge pipe.

While the change in phytoplankton and zooplankton communities remained rather muted in Lake Nuasjärvi, there was a trend of decreasing diversity in both groups (Figs 3, 5). It has been shown that metal contamination leads to low diatom diversity (Tolotti et al., 2019) and a similar response to metal mining inputs has been shown for cladocerans as well (Winegardner et al., 2017). Hence, it may be that also in Lake Nuasjärvi the planktonic communities responded to the mine waste waters through decreasing diversity, whilst the community changes remained hidden due to the high dominance of *Aulacoseira* species and *Eubosmina* spp.

4.2. Benthic communities

Although the bottom-dwelling chironomids (Hofmann, 1998) showed the largest community changes of the examined organism groups in general, they did not exhibit a major change at site N11 (Figs 7, 8) as did diatoms and cladocerans. The greatest changes in chironomids occurred at sites N7 and N8, which are located close to the discharge pipe. At both these sites, *Procladius* showed a significant increase in the surface sample. *Procladius* is a resilient taxon (Cao et al., 2016), which typically increases under environmental perturbations, such as heavy metal pollution (Ilyashuk et al., 2003) and eutrophication (McKeown and Potito, 2016). As a free-living predatory taxon typically occurring in the profundal sediment-water interface, *Procladius* is also able to avoid oxygen deficit conditions when necessary and is sometimes considered as the most anoxia-tolerant chironomid taxon (Larocque-Tobler and Pla-Rabès, 2015). Another taxon that tolerates low-oxygen at the hypolimnium is *S. coracina*-type, which however disappears from the surface samples of sites N7 and N8. It has been shown that although *S. coracina*-type has a low hypolimnetic oxygen optimum (Quinlan and Smol, 2001), it is the most sensitive indicator for benthic collapse under hypoxia (Belle et al., 2017). This could suggest that a hypolimnetic oxygen threshold has been crossed at sites N7 and N8. In addition to *S. coracina*-type, *C. plumosus*-type possesses extracellular hemoglobin, and hence, tolerates very low oxygen concentrations and periods of anoxia (Quinlan et al., 1998; Nath, 2018). In Lake Nuasjärvi, *C. plumosus*-type, which is also an indicator of toxic metal impact having a very high tolerance (Ilyashuk et al., 2003; Brooks et al., 2005), shows a significant increase at site N7, but disappears from the adjacent site N8 further suggesting that the environmental conditions have become close to a threshold. Deterioration of environmental conditions is also reflected by a decrease in *P. sordidellus*-type, which is sensitive to human impacts (Chique et al., 2018) and in the present record disappears from the modern samples of sites N7 and N8.

The reason why sites N7 and N8 have had the largest change in chironomid composition is partly explained by the fact that these are also the deepest sites in the present dataset, hence being most vulnerable to hypolimnetic oxygen deterioration (Müller et al., 2012). A significant change has also occurred at site N16, which is located behind an aquatic sill when observed from the waste water discharge pipe (Fig. 8). At N16, *S. coracina*-type and *C. plumosus*-type again disappeared/decreased, while the non-sedentary *Procladius* markedly increased (Fig. 7). Concurrent with these shifts, *Glyptotendipes pallens*-type and *Dicrotendipes nervosus*-type clearly increased suggesting higher nutrient availability based on their ecological optima in Finnish lakes (Raunio et al., 2010; Luoto, 2011). Higher near-bottom nutrient availability could be due to low oxygen conditions that promote phosphorus release from sediments, but this process is complex and difficult to pinpoint in case studies (Hupfer and Lewandowski, 2008).

In the case of all organism groups, site N13 had no significant change in their aquatic community, this site being the shallowest of the sites and close to the shore, suggesting that the ecological changes are emphasized in open water areas and especially at deep lake bottoms. Similar to diversity of diatoms and cladocerans, chironomid diversity decreased in the surface samples (Fig. 7). Especially deep-water sites became dominated by only a few taxa, while others disappeared (Fig. 7), indicating a detrimental loss of benthic food web functions (Kivilä et al., 2019). It has been shown that chironomid diversity typically decreases under heavy metal contamination (Mousavi et al. 2003), but also under oxygen depletion (Brodersen and Quinlan, 2006). Moreover, chironomid richness closely corresponds to the ecological quality of lakes (Tolonen et al., 2014). In Lake Nuasjärvi, the sites with collapsed benthic biodiversity were characterized by the disappearance of collector-filterers (such as the Tanytarsini taxa), which collect fine particulate organic matter from the water column using a variety of filters, and subsequent dominance of few taxa representing feeding guilds of predators (e.g. *Procladius*), shredders (e.g. *G. pallens*-type) and/or collector-gatherers (e.g. *D. nervosus*-type and *Nanocladius rectinervis*-type) (Merritt and Cummins, 1996; Mandaville, 2002). This change in benthic ecological functions in Lake Nuasjärvi is in line with a previous study on disturbance (eutrophication) driven functional change from southern Finland, where collector-filterers were the main describer of a functionally diverse community, whereas predators, shredders and collector-gatherers were characteristic of driving functional degradation (Luoto & Ojala, 2014). Since chironomids have vital functions in lakes through operating crucial biogeochemical cycles by taking part in processes related to detrital decomposition, nutrient release and transfer, prey control and food supply (Palmer, 1997; Covich et al., 1999), the loss in taxonomic and functional diversity of the benthos should be of great concern (Luoto and Ojala, 2018).

4.3. Ecological impacts of salinated mine water

Based on monitoring data, electric conductivity, i.e. salinity (Williams and Sherwood, 1994), has significantly increased in Lake Nuasjärvi following the construction of the treated waste water discharge pipeline in late 2015 (Fig. 2). In a previous study (Leppänen et al., 2019), the ecological harmfulness of saline mine water from the Talvivaara/Terrafame mine to the aquatic life (including diatoms, cladocerans and chironomids) was demonstrated in Lake Jormasjärvi, located between Lake Nuasjärvi and the mining district. In Jormasjärvi, the impact of salinated mine water was the strongest close to the inlet stream which carries the effluents through smaller basins, but the water quality improved moving away from the inlet and towards Lake Nuasjärvi (Leppänen et al., 2019). From another site, Lake Kivijärvi, a smaller basin located only 7 km from the mine, it was shown that saline mine water had turned the lake meromictic and the sediment was found heavily contaminated (Leppänen et al., 2017). The ecological change in Lake Kivijärvi was reflected through major turnovers in phytoplankton and zooplankton communities through time (Leppänen et al., 2017).

In Lake Nuasjärvi, the effects of salinity are not as visible despite the fact that there has been a distinct increase in salinity of the lake, particularly after the installation of the mine waste water discharge pipe (Fig. 2). This is not surprising though, since the pipeline was not built until the end of 2015 and hence the time for the ecosystem to respond has remained rather short. Similar to Lake Jormasjärvi, the decrease in the diatom *L. rossii* in almost all of the surface samples (Fig. 3) can nonetheless be a reflection of salinity change since it prefers low-conductivity lakes (Håkansson, 1990; Camburn and Charles, 2000). In cladocerans, *B. longirostris* is more resilient to increased salinity compared to many other taxa (Adamczuk, 2016; Zawisza et al., 2016), but in the current record no straightforward pattern was observed in its distribution (Fig. 5). However, the chironomids *Procladius* and *N. rectinervis*-type, which show distinct increases in the surface samples (Fig. 7) are known to have high abundances in lakes with elevated conductivity and total dissolved solids (TDS) (Eggermont et al., 2006; Zhang et al., 2007; Dickson et al., 2012) suggesting increased salinity in all samples, especially at sampling site N8. Stratifying effects of salinity can be considered to be most emphasized at deepest parts of basins (Novotny et al., 2008), and in the Lake Nuasjärvi dataset, N8 represents the deepest sampling site and has elevated measured electric conductivity (Mäkinen, 2017), giving further strength to the interpretation, and highlights the connections between saline mine water inputs, enhancing stratification and decreasing hypolimnetic oxygen. Furthermore, it may not be surprising that the effects of salinity are most visible in the macrobenthic community, since the highest electric conductivity values in Lake Nuasjärvi are measured from deep water layers (Fig. 2). Above these poorly mixed saline layers, the fresh surface waters deriving from the Sotkamo route (eastern inlet in Fig. 1) maintain a faster flow rate in epilimnetic waters (Mäkinen, 2017), keeping them more unpolluted. In addition to chemical stratification, salinity can enhance thermal stratification in lakes (Novotny and Stefan, 2012). Although not investigated in this study, the studied organism groups all have a metabolic response to water temperature (Rühland et al., 2008; Zawiska et al., 2017; Nevalainen et al., 2013) that may act as an additional factor behind their within-lake distribution patterns.

It has been demonstrated that intralake distribution of “modern” fossils of diatoms, cladocerans and chironomids in lake surface sediment samples well represent the living communities of those particular habitats (Laird et al., 2010; Luoto, 2010; Nevalainen et al., 2018), i.e. the fossils are found close to the species’ living environments. However, it has also been shown that motile animals, such as Cladocera, are able to avoid or escape polluted conditions (Lopes et al. 2009) that could partly explain their muted response in Lake Nuasjärvi (Fig. 5). For example, while the planktonic *Eubosmina* may be absent from the bottom waters of the polluted sites, it can thrive at the same time in the above fresh surface waters, enabling the accumulation of its fossil into the sediments at these sites. In the case of sessile benthic invertebrates, such as many chironomids, a similar phenomenon is not possible.

4.4. Reconstructed hypolimnetic oxygen and Benthic Quality Index

Comparison between the chironomid-inferred hypolimnetic oxygen values and the BQI indicated a statistically significant relationship ($p<0.001$) implying that they both reflect the same environmental control. The highest “reference” bottom water oxygen conditions were reconstructed for sample N13 (Fig. 9), which is logical, since it is the shallowest close-to-shore sampling site, and hence, more prone to vertical mixing (Rogora et al., 2018). The supersaturated oxygen conditions of the sample were confirmed by the BQI, which showed value close to 4 and included high-oxygen BQI indicator taxa (Johnson, 1998), such as *Heterotanytarsus, Heterotrissocladius marcidus*(-type) and *H. grimshawi*(-type). At other sites, the bottom sample oxygen conditions (~3-8 mg l^{-1}) reflected moderate oxygen availability.

The oxygen development showed spatial variability in Lake Nuasjärvi, although in none of the samples oxygen conditions improved from the bottom to the top (Fig. 9). At sampling site N13 hypersaturated oxygen conditions decreased to a moderate level (~6 mg l^{-1}), whereas in

N4, N11 and N16, the values remained at a moderate level, showing only a minor decrease from the "reference" samples to the surface samples. Considering the results in terms of the potential impact of the mine waste water discharge pipeline built in 2015, it should be noted that the sample N13 is located close to the shore and counter-flow according to waste water discharge modeling (Lehtinen et al., 2017). Also the sample N11 is located counter-flow, whereas N16 is behind an aquatic sill in relation to the pipe end and N4 is located further away at the Rimpilänsalmi strait explaining the minor limnological responses at these sites. In the samples from sites N4, N11 and N16, there was also no decrease in the BQI values confirming the minor benthic limnoecological effects of the pipe construction in directions of counter-surface flow and further away (~6 km) from the waste water discharge point.

When looking at sampling sites located closest to the waste water discharge pipe, i.e. sites N7, N8 and N10, it can be seen that all have experienced a clear decrease in the hypolimnetic oxygen levels. In an assessment of contemporary waste water dispersal in Lake Nuasjärvi, these particular areas were found to suffer from the lack of springtime mixing and subsequent oxygen deficiency (Mäkinen, 2017) fitting well with our results. The "reference" oxygen concentrations of 3-6 mg l^{-1} have decreased in the surface samples to 0-2.8 mg l^{-1} with anoxic conditions at N8 and close to anoxic at N10. In the waste water dispersal assessment (Mäkinen, 2017), elevated electric conductivity values indicate that salinated mine waters are transported by the bottom currents towards north-west, and hence, specifically towards our sampling sites N8 and N10.

Also the BQI decreases at the close-to-pipe sites, but only a minor change is observed at sites N7 and N10. At N7, the reason why the BQI does not reflect the clear oxygen decrease owes to the presence of *Tanytarsus* taxa (including Tanytarsini and *T. pallidicornis*-type). In the BQI, *Tanytarsus* is assigned with a relatively high value of 3 (Johnson, 1998), although the genus includes both oxyphobic and oxybiotic taxa (Little and Smol, 2001). At sample N10, the relatively minor decrease in BQI compared to the oxygen decline is due to high abundance of *S. coracina*-

type in the surface sample. Although *S. coracina*-type possesses hemoglobin (Lindegaard, 1995) and has low hypolimnetic oxygen optimum in the applied calibration set (Luoto and Salonen, 2010), similar to *Tanytarsus*, it has been assigned a relatively high BQI value of 3 (Johnson, 1998). It should be clear though that *S. coracina*-type is an indicator of low-oxygen conditions, since hemoglobin is ecologically costly (Panis et al., 1995), and hence, the taxon would not have this attribute without a reason. In summary, the results clearly show decreased hypolimnetic oxygen conditions in Lake Nuasjärvi following the construction of the mine waste water discharge pipeline. The impacts are emphasized in the deep water sites adjacent to the pipe and they are most likely dictated by enhanced stratification caused by the mine water-originated salinization of the lake water. It should be noted that all these changes have occurred within less than two years after the construction of the pipeline.

5. Conclusions

The results of this top-bottom paleolimnological study from Lake Nuasjärvi suggest minor changes in diatom communities, negligible effects on cladocerans and major changes in chironomid communities between the "reference" and modern samples. The results hence show emphasized ecological impacts of the construction of the treated mine waste water discharge pipe in 2015 on deepwater macrobenthic communities, whereas the phytoplankton and zooplankton communities of upper water layers have been less affected. Shifts in macrobenthic communities have occurred throughout the basin, but the largest changes in ecological integrity were found from the deepest sampling sites located close to the waste water pipe outlet with complete faunal turnovers and loss in diversity.

In addition to decreased benthic quality (BQI), hypolimnetic dissolved oxygen conditions, inferred using chironomids, have decreased at the sites near the waste water discharge pipe outlet, whereas lesser changes have occurred further away from the main basin and counter-flow from the pipe. Since the current data or the water quality monitoring data do not show signs of epilimnetic nutrient enrichment, the decreased and partly anoxic conditions found in the deep basins of Lake Nuasjärvi are most likely owing to increased salinity derived from the mine waste waters that have caused enhanced limnological stratification. Subsequently, the stronger stratification causes diminished water column mixing, enhances the oxygen decrease and increases the deterioration of benthic communities.

This study demonstrates the harmfulness of treated mine waste waters to bottom waters even in large and deep lakes. It also highlights the sensitivity of biological communities towards mine waste water discharge as clear and directional changes can already be seen in less than two years after the construction of the waste water discharge pipeline. While this study cannot disentangle the direct ecotoxicological effects of the mine waste water inputs on aquatic life, it clearly shows the serious limnoecological impacts through salinization and subsequent stratification that cause fatal hypolimnetic oxygen deficiency and further lead to the almost total degradation of benthic ecological integrity near the pipe outlet. This study also suggests that investigating solely surface water bodies may not be enough to detect the impacts on the hypolimnion. It is therefore recommended that when performing baseline and impact assessments of lakes, several trophic levels representing different within-lake habitat environments should be used. In addition, as shown in this study, the spatiotemporal perspective is invaluable since the applied paleoecological proxies do not provide an integrated whole-lake signal, but rather represent sampling site-specific evidence.

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8. Figure captions

Fig. 1. Location of the study site in Finland. Major inlets and outlets are marked with arrows. The eastern inlet is the Sotkamo route, which brings fresh unpolluted water to the epilimnion of Lake Nuasjärvi. The city of Kajaani is located on the western shore of the Rehja basin (western basin) and the municipality of Vuokatti on the eastern shore of the Nuasjärvi basin (blue color). The mining district begins 10 km south of Nuasjärvi. The mine is marked with a hammer symbol and the constructed waste water pipeline is marked with a dashed line.

Fig. 2. Monitored electric conductivity (EC) and total phosphorus (totP) in Lake Nuasjärvi since the 1980s (Finnish Environment Institute). The sampling frequency at 5 different depths (~1m, 5m, 10m, 15m, 20m) for EC is 1-6 times per year and for totP once a year (August).

Fig. 3. Diatom assemblages (relative proportions of taxa with abundance >3%) in the surface (0.25 cm = red) and "reference" (3-4 cm = green and 10-11 cm = blue) samples in Lake Nuasjärvi. Also given are rarefied taxon richness and the Shannon diversity index (H).

Fig. 4. Spatiotemporal dissimilarity (Bray-Curtis) between the "reference" (10-11 cm) and surface samples (0-0.25 cm) of diatom assemblages at the sampling points N4, N7, N8, N10, N11, N13 and N16. The outlet of the waste water discharge pipe is marked with a star and the water sampling station with a triangle.

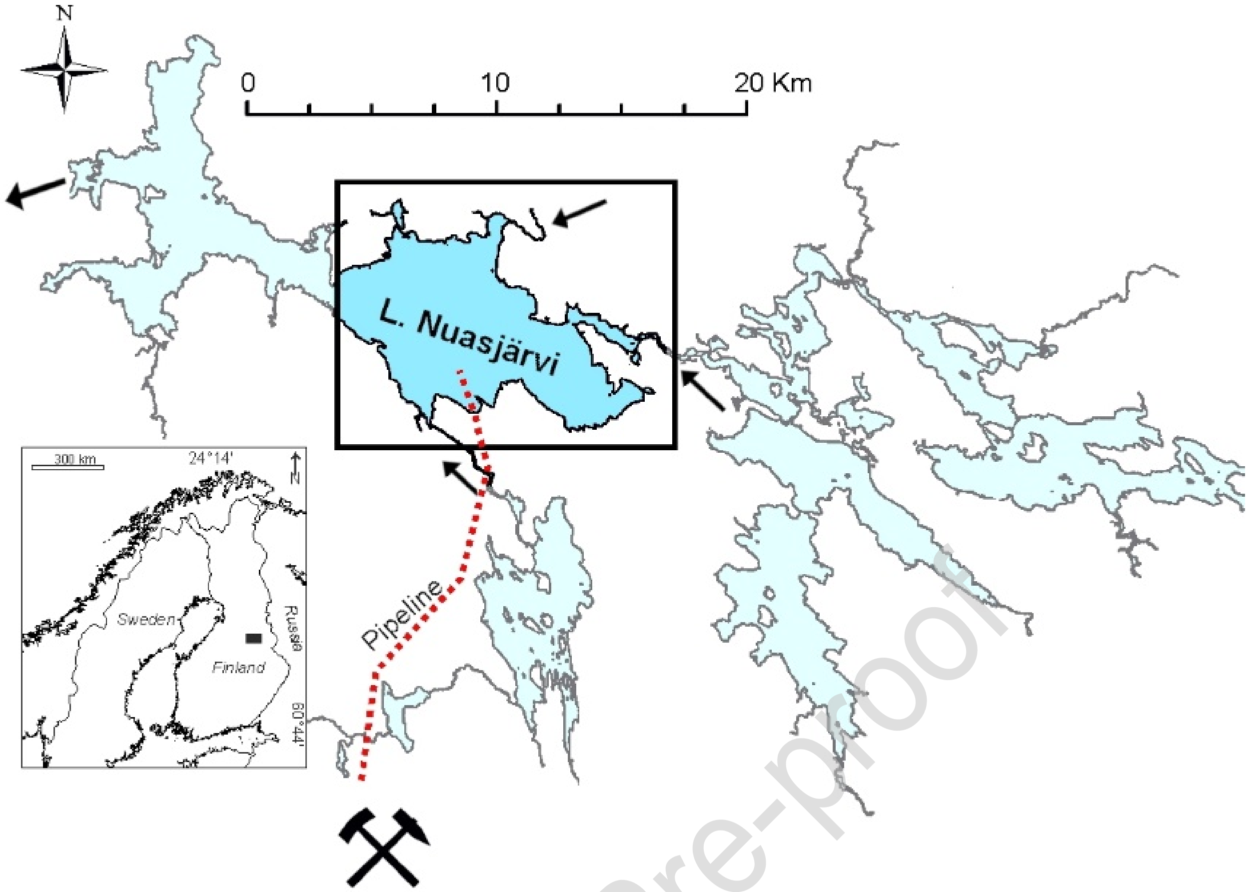
Fig. 5. Cladocera assemblages (relative proportions of taxa with abundance >2%) in the surface (0.25 cm = red) and "reference" (3-4 cm = green and 10-11 cm = blue) samples in Lake Nuasjärvi. Also given are rarefied taxon richness and the Shannon diversity index (H).

Fig. 6. Spatiotemporal dissimilarity (Bray-Curtis) between the "reference" (10-11 cm) and surface samples (0-0.25 cm) of Cladocera assemblages at sampling points N4, N7, N8, N10, N11, N13 and N16. The outlet of the waste water discharge pipe is marked with a star and the water sampling station with a triangle.

Fig. 7. Chironomid assemblages (relative proportions of taxa with abundance >10%) in the surface (0.25 cm = red) and "reference" (3-4 cm = green and 10-11 cm = blue) samples in Lake Nuasjärvi. The taxa are arranged according to their hypolimnetic oxygen optima in the applied calibration set with taxa tolerating oxygen deficiency on the left side of the upper graph and taxa demanding higher oxygen concentrations on the right side of the lower graph. Also given are rarefied taxon richness and the Shannon diversity index (H).

Fig. 8. Spatiotemporal dissimilarity (Bray-Curtis) between the "reference" (10-11 cm) and surface samples (0-0.25 cm) of chironomid assemblages at sampling points N4, N7, N8, N10, N11, N13 and N16. The outlet of the waste water discharge pipe is marked with a star and the water sampling station with a triangle.

Fig. 9. Minimum hypolimnetic oxygen conditions inferred using the Finnish chironomid-based calibration model (Luoto and Salonen, 2010) and the Benthic Quality Index (Johnson, 1998) in the modern (0.25 cm) and "reference" (3-4 and 10-11 cm) samples from Lake Nuasjärvi. A brief description of each sampling point in regard to the mine waste water discharge pipe is also given (counter-flow refers to surface waters).



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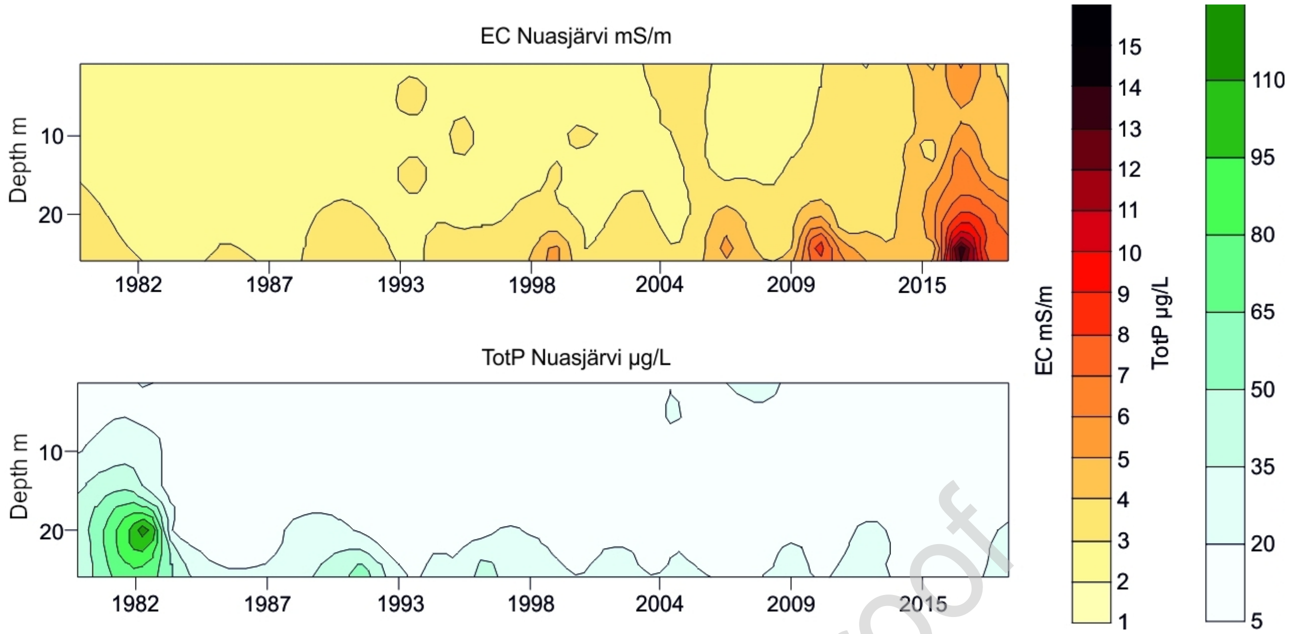

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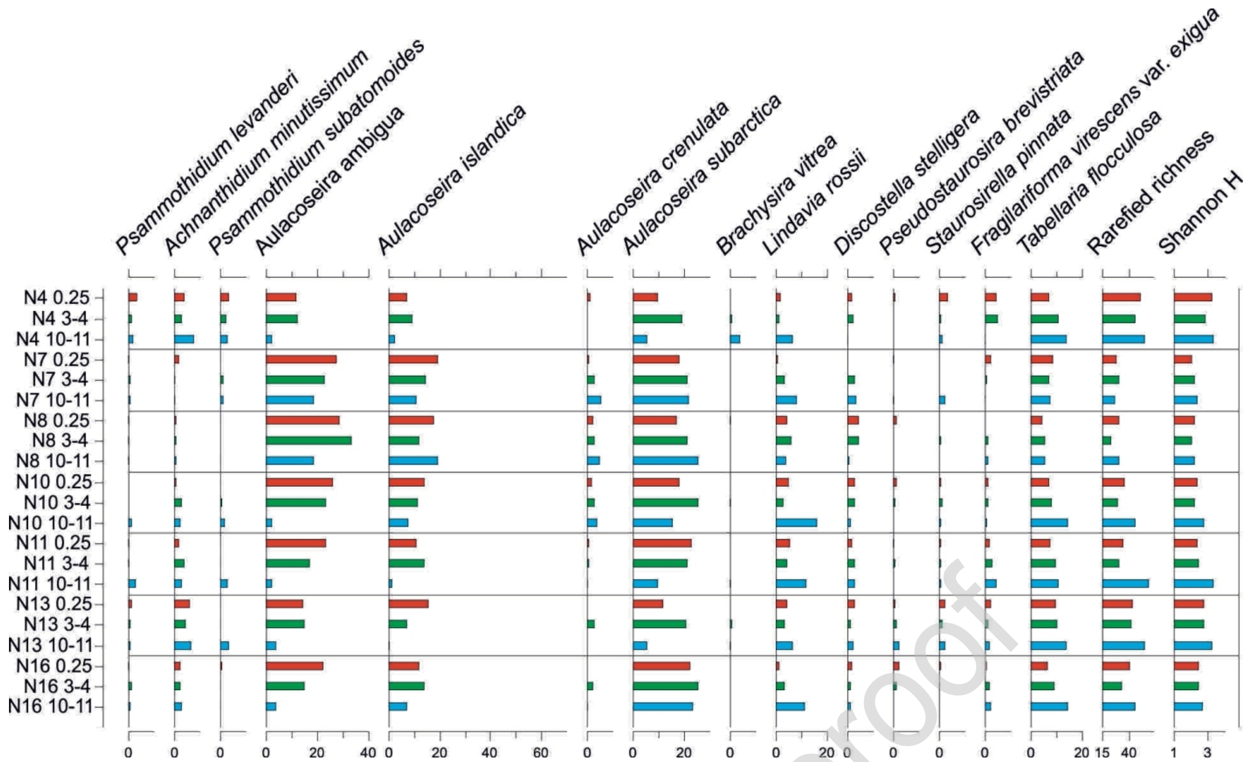

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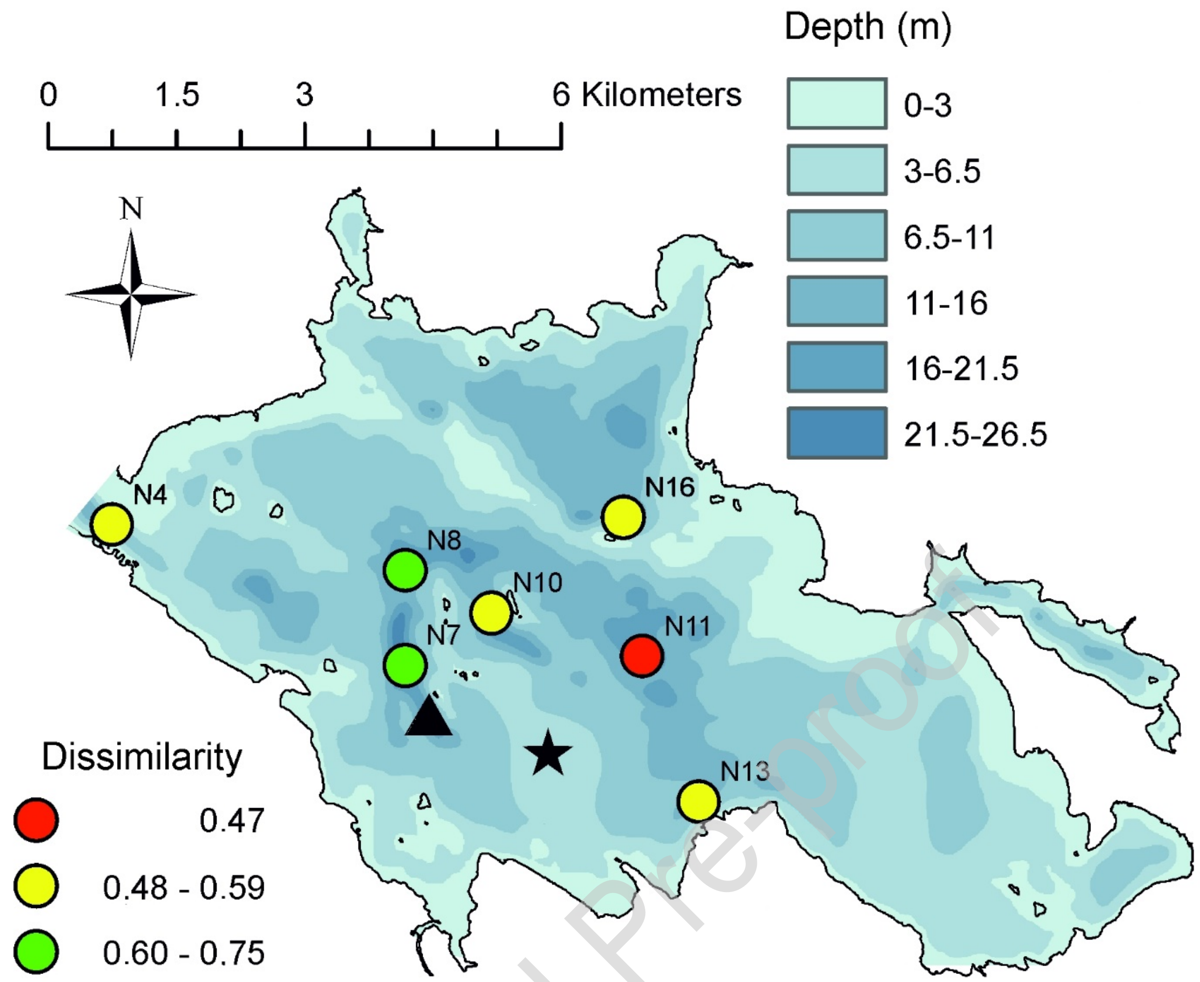

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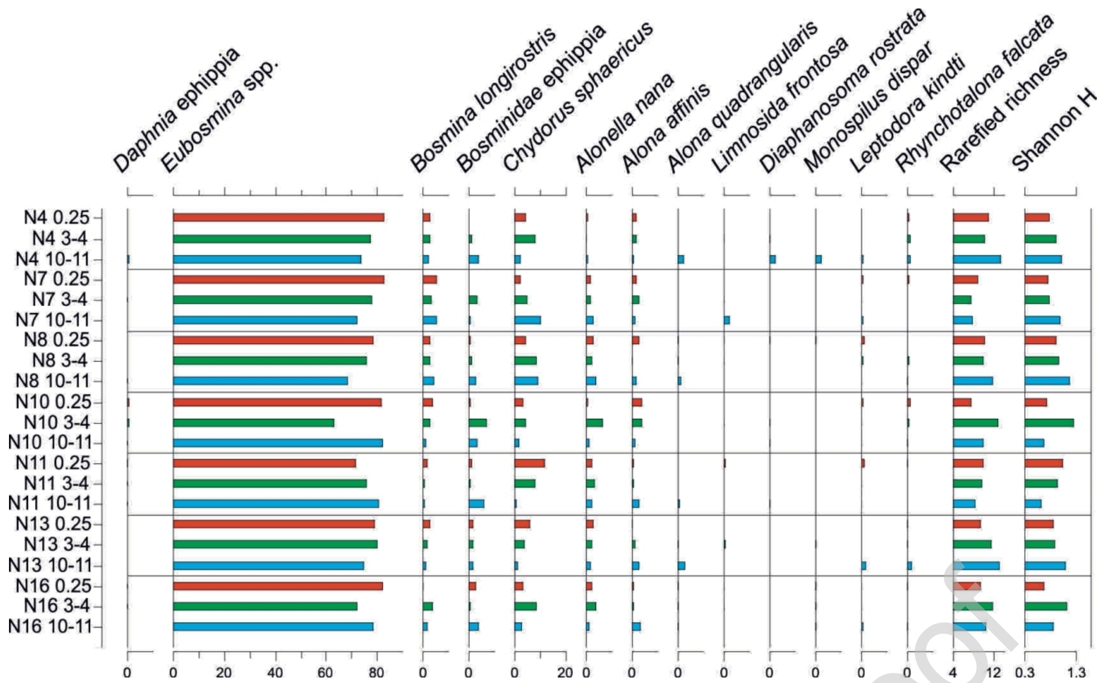

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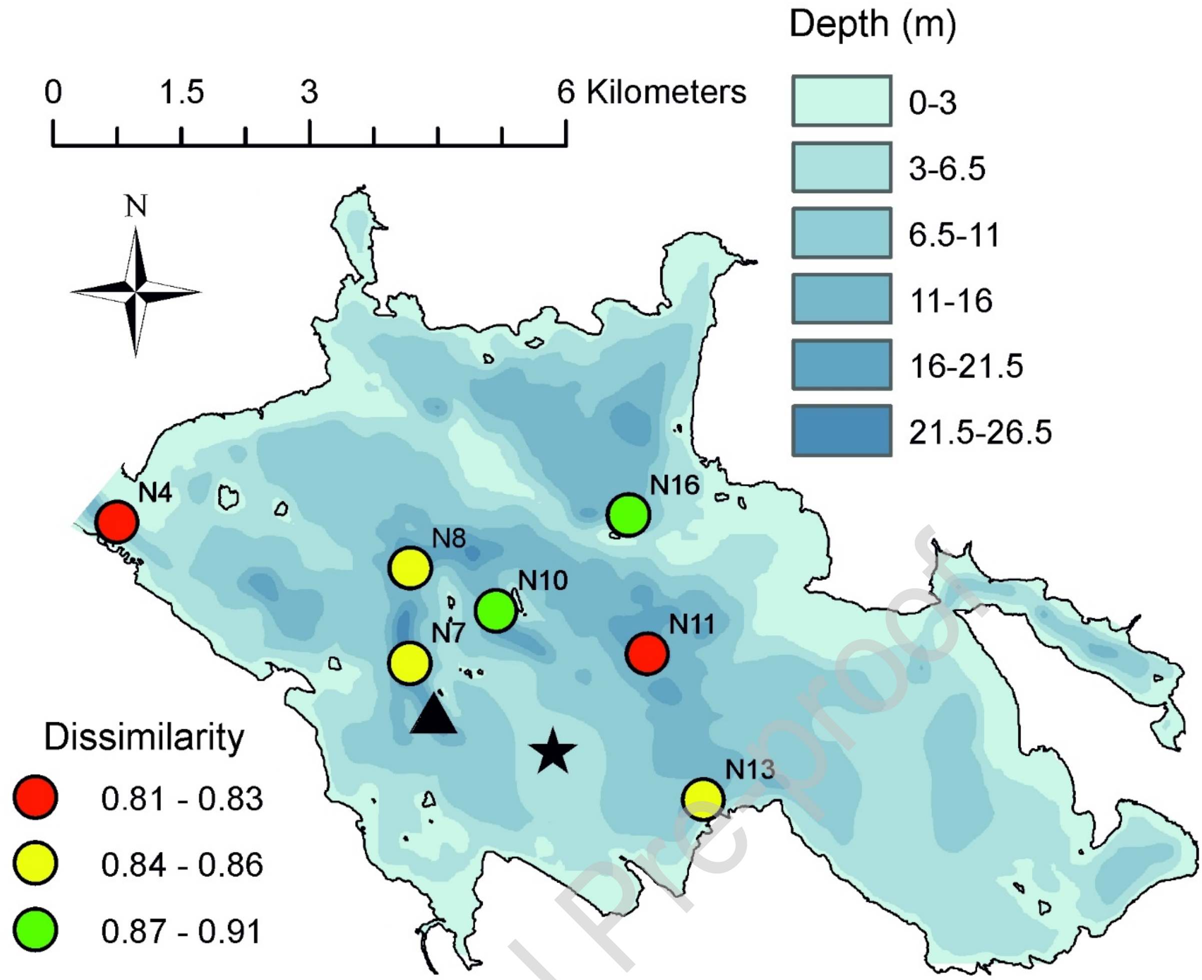

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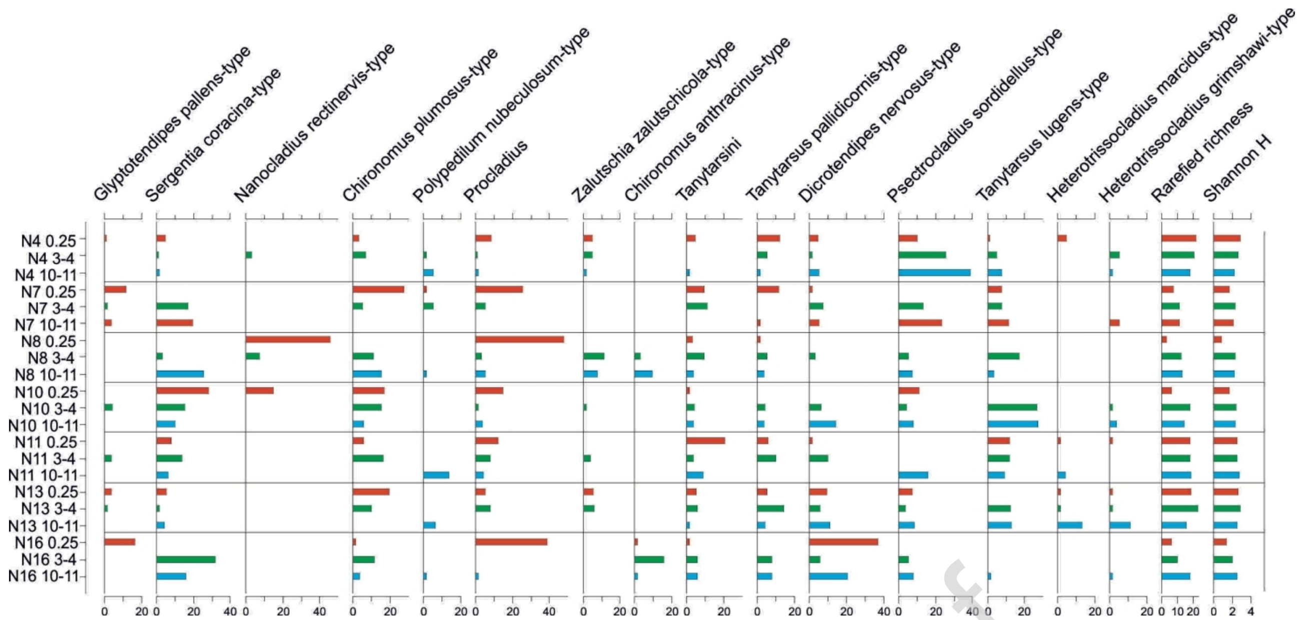

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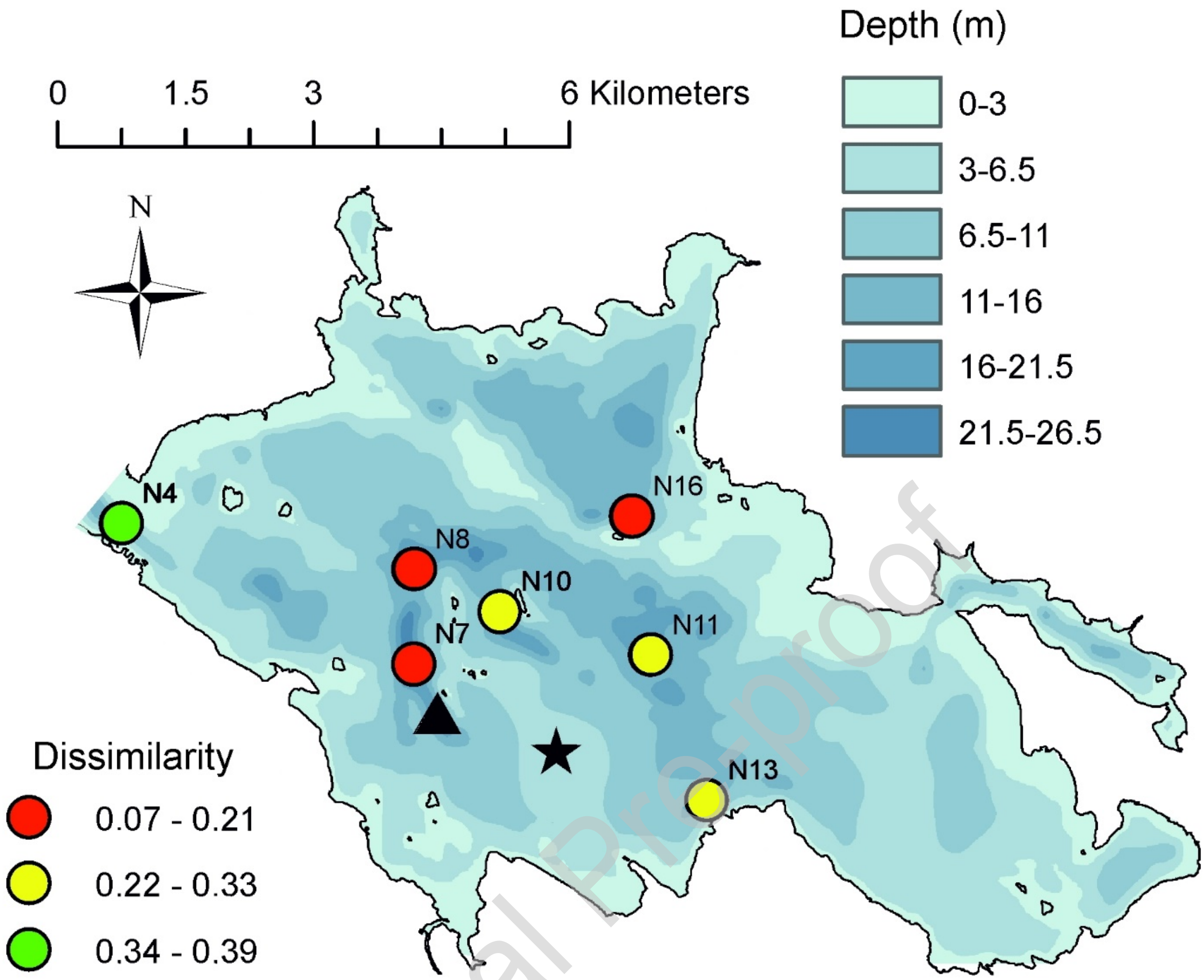

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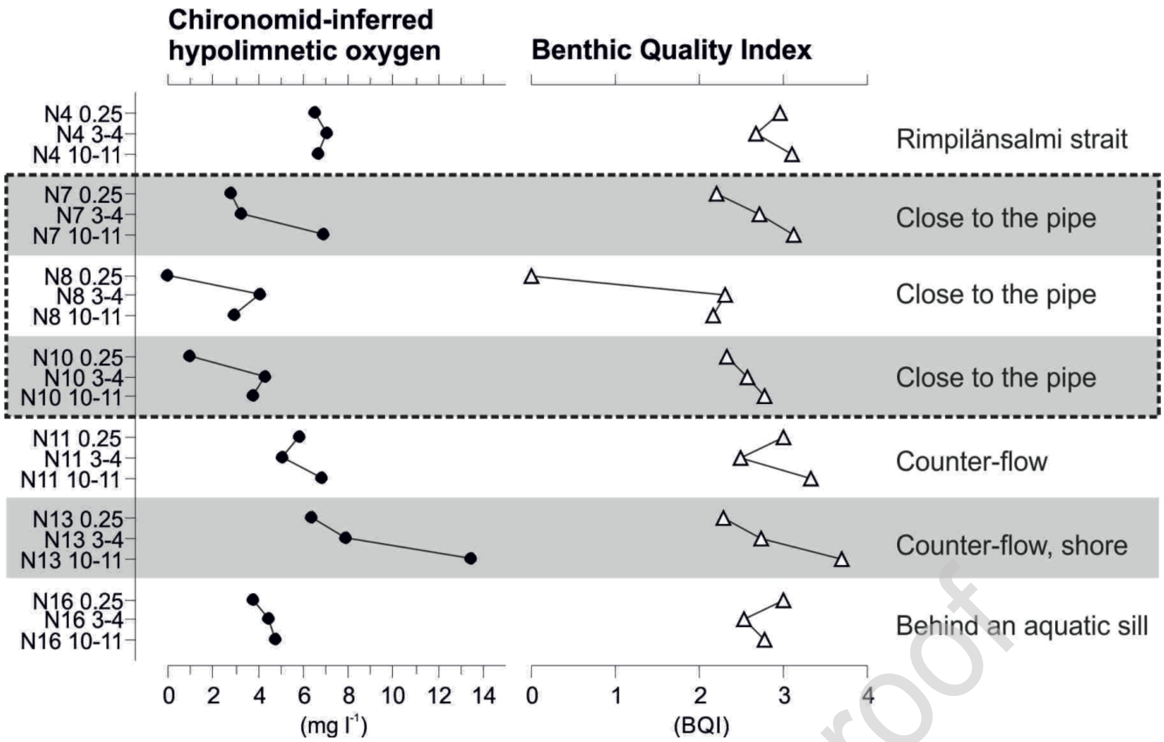

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Declaration of interests

☒ The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

☐The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Highlights

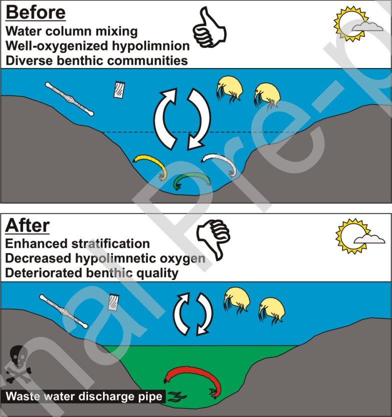
1. Impacts of a pipeline from an enormous nickel mine on Lake Nuasjärvi were examined.

2. A paleolimnological top-bottom approach (diatoms, Cladocera, chironomids) was used.

3. Diverse communities and water column mixing characterized pre-pipeline conditions.

4. The pipeline-based salinated mine waste waters enhanced stratification and anoxia.

5. Subsequently, benthic ecological integrity degraded and diversity decreased.


Before
Water column mixing
Well-oxygenized hypolimnion
Diverse benthic communities
After
Enhanced stratification
Decreased hypolimnetic oxygen
Deteriorated benthic quality
Waste water discharge pipe