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Mining pollution triggered a regime shift in the cladoceran community of Lake Kirkkojärvi, southern Finland

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## **Abstract**

Mining is one of the key industries in the world and mine water pollution is a serious threat to aquatic ecosystems. Historical monitoring data on the pollution history and impacts in aquatic ecosystems, however, are rarely available, so paleolimnological methods are required to explore the consequences of past pollution. We studied the history of cladoceran community dynamics in Lake Kirkkojärvi, southern Finland, including the periods before, during and after mining. We analyzed the geochemical composition and cladoceran subfossil remains in a  $^{210}\text{Pb}$ -dated sediment core to evaluate the magnitude, rate, and direction of cladoceran community changes through time. The cladoceran community was altered significantly by mining activity that occurred during the mid-20<sup>th</sup> century. During more recent times, however, eutrophication effects have overridden the impacts of mining. After mining ceased, the cladoceran community underwent an abrupt regime shift towards taxa that reflect more eutrophic conditions. This change was caused by intensive farming activity and fertilizer use over the past few decades. The recent history of Lake Kirkkojärvi is a textbook example of a regime shift triggered by multiple human-caused stressors. Our findings also highlight the utility of cladocerans as bio-indicators in pollution research and illustrate the sensitivity of aquatic ecosystems to anthropogenic modification.

## **Keywords:**

Minewater, Mining, Pollution, Cladocera, Paleolimnology, Finland

## Introduction

Industrial pollution is a major cause of degradation of freshwater ecosystems throughout the world. Fortunately, the information gained from past pollution cases can be used to better prepare for the future and improve environmental management practices. If long-term monitoring data are scarce or absent, however, the ecological impacts of past pollution are difficult to assess. Unfortunately, this is often the case, making it difficult to distinguish pollution impacts from other environmental changes. Paleolimnological methods, however, can be used to infer past environmental conditions, and provide information about the timing, direction, and magnitude of pollution effects. Cladocerans (water-fleas), an order of microscopic Crustacea, are widely used paleobioindicators (Jeppesen et al. 2001; Korhola and Rautio 2001). In addition to their utility as environmental bioindicators, the group is vital to the function of aquatic ecosystems, as they are a bridge for energy flow from primary producers to higher-level consumers in aquatic food webs (Sterner 2009).

Mining is one of the key industries in the world and can have strong negative impacts on the environment. Mining dam failures are a serious threat to ecosystems and human health, with recent examples in Brazil (Escobar 2015), Romania (Soldán et al. 2001), Spain (Feasby et al. 1999) and Italy (Alexander 1985). Mine tailings have also been shown to have negative impacts on lake ecosystems (Coard et al. 1983; Bozelli 1996; Garrido et al. 2003; Vandysh 2004). Even abandoned mines can cause environmental problems, decades after the termination of mining operations. For instance, acid mine drainage (AMD) is a serious problem related to abandoned mines (Kelly 1988; Johnson 2002).

Lake Kirkkojärvi, located in Viljakkala, southern Finland, has received contaminated effluent that originated from tailings produced by the Haveri copper and gold (Cu-Au) mine, which operated between 1938 and 1960. Previous paleolimnological studies identified historical changes in sediment geochemistry, protists, and diatom communities after the shutdown of the Haveri mine (Kihlman and Kauppila 2010). The delay in peak concentrations of metals in sediments was explained by the slow and gradual oxidation of mine tailings (Parviainen et al. 2012). The relatively low ecological impact of AMD (high concentrations of Cu, Zn, Ni, As) was attributed to the limited bioavailability of metals in the lake water. Whereas ecological changes coincided with high metal concentrations, the post-mining aquatic communities were influenced more by elevated nutrient concentrations than metal pollution (Kihlman and Kauppila 2010).

Because of their position in the food web, zooplankters are vulnerable to shifts in food availability and predation. In particular, cladocerans have been used extensively in stress research (Suhett et al. 2015) and ecotoxicology studies (Sarma and Nandini 2006). Generally, cladocerans are sensitive to metal pollution (Brix et al. 2001; Von Der Ohe and Liess 2004). Shifts in the cladoceran community, however, may also reflect changes in predation (Nykänen et al. 2006), lake trophic status (Nevalainen and Luoto 2013), and water level (Korhola et al. 2000), which make cladocerans excellent bioindicators of the environmental history of lakes. Their preservation in sediments, rapid reproduction, short life cycles, small body size, and critical role in pelagic food webs make them the preferred zooplankton group in studies of ecological stress, particularly in the context of ongoing natural and human-induced changes in aquatic ecosystems.

We applied paleolimnological methods to a  $^{210}\text{Pb}$ -dated sediment core from Lake Kirkkojärvi, Finland, to study the ecological impacts of the abandoned Haveri mine on the cladoceran community. We also evaluated whether recent shifts in the cladoceran community structure were a response to eutrophication or perhaps a shift at a higher trophic level than reported in a study by Kihlman and Kauppila (2010). Finally, we assessed the recovery dynamics of the cladoceran population after intense environmental stress. Our research contributes to the understanding of the ecological impacts of mining, and of abandoned mines, which are abundant worldwide, e.g. half a million in the U.S. alone (Fields 2003), and present serious threats to human health and the environment.

## Study site

Lake Kirkkojärvi is a small (75 ha), relatively shallow ( $z_{\text{max}} = 8.5$  m,  $z_{\text{mean}} = 2.3$  m) lake embayment of the Lake Kyrösjärvi system, located in Ylöjärvi, southern Finland (61.714837° N, 23.267732° E WGS84), at 83 m above sea level (a.s.l.) (Fig. 1). Mean annual temperature in the area is  $\sim 4$  °C, and annual precipitation is approximately 600 mm (Finnish Meteorological Institute 2015). The catchment area of Lake Kirkkojärvi consists mainly of agricultural fields and forests. Lake Kirkkojärvi is a turbid-water lake embayment, with nearly neutral pH (6.6-7.4) and mesotrophic nutrient status (Kihlman and Kauppila 2010). The different embayments of the Lake Kyrösjärvi system differ in their limnological characteristics. Lake Viljakkalanselkä (520 ha) is less nutrient-rich (Kihlman and Kauppila 2010) and the water is more transparent than in Lake Kirkkojärvi. The water in the main body of Lake Kyrösjärvi (9000 ha), which is located west of Lake Kirkkojärvi, is

characterized by its yellow-brown color, a consequence of higher concentrations of humic substances (Fig. 1). The fish community of the lake system is relatively rich, hosting roach *Rutilus rutilus*, perch *Perca fluviatilis*, pike *Esox lucius*, bream *Abrahamis brama*, asp *Aspius aspius*, burbot *Lota lota*, crusian carp *Carassius carassius*, blue bream *Abramis ballerus*, vendace *Coregonus albula*, and European smelt *Osmerus eperlanus*. The lake system has been stocked with whitefish *Coregonus lavaretus* and pike perch *Sander lucioperca* (Pitkänen 2007) over the past three decades (Keskitalo 2014).

### Environmental history and mining pollution

The first major human-induced changes in the recorded history of Lake Kirkkojärvi were deliberate reductions in lake levels during the 19<sup>th</sup> century (Vänni 1928), in an attempt to produce more arable land and minimize flood impacts. The previous shoreline, when the lake level was approximately 86 m a.s.l., three meters higher than today, can be seen in old maps. In the early 19<sup>th</sup> century, Peltosaari and Inkula were islands (Calonius 1805; Fig. 1). Following the water-level modifications during the late 19<sup>th</sup> century, however, Inkula became attached to the mainland, and during the 1950s, waste from the Haveri mine connected Peltosaari Island to the mainland. The population in Viljakkala reached its peak (~3000) in the early 1950s (Kaskimies and Sinisalo 1973), which corresponds well to the peak years of the agricultural sector in Finland. The era of the “green revolution” after the Second World War was characterized by use of heavy agricultural machinery and industrial fertilizers. Problems associated with eutrophication are widespread in agricultural areas in Finland, and especially affect small and medium-sized lakes (European Environment Agency 2010). In Lake Kirkkojärvi and neighboring Lake Viljakkalanselkä, phosphorus concentrations have increased slightly over the past few decades (Kihlman and Kauppila 2010).

Industrial-scale copper (Cu) and gold (Au) mining began in Haveri in 1940. During the 1950s, annual production volumes reached 120,000 tonnes for total ore and 300 kg for Au (Kaskimies and Sinisalo 1973). The Haveri ore-processing facility used water to flush tailings downhill into the waste area located on the shore of Lake Kirkkojärvi (Hannu Uotila pers. commun.). In aerial photograph from 1950–54 (Pöyry 2015), mine waste can be seen entering the lake. This photographic evidence shows that the waste area was not yet dammed in 1950–1954, and the dam that exists today was constructed later. Despite the presence of the dam, multiple overflow incidents occurred (Hannu Uotila pers. commun.) and during the late 1950s, tailings were dumped

directly into the lake, outside the dam (Parviainen et al. 2012). The mine was closed in 1957, but low-volume mining continued until the end of 1960 (Kaskimies and Sinisalo 1973). Maintenance of the tailings area was terminated after the mine closure, and during the early years of the 1960s the north side of the tailings dam failed and was never repaired (Hannu Uotila pers. commun.).

According to a previous paleolimnological study, highest metal concentrations in the sediment and the clearest changes in the diatom and protist communities occurred during the post-mining period (1970s), caused by AMD, and later, by nutrient enrichment (Kihlman and Kauppila 2010). In addition, according to the OIVA database (Finnish Environmental Institute), lakewater iron (Fe) and manganese (Mn) concentrations increased from the 1960s to the 1970s. The tailings dump is still leaking acid and metal-contaminated leachate into the bayhead of Lake Kirkkojärvi, but the current toxic impact is practically non-existent (Leppänen et al. 2017).

## Materials and methods

### Coring

We retrieved a 44.5-cm-long, 6-cm-diameter sediment core from the central basin of Lake Kirkkojärvi, at a water depth of 8.5 meters, using an Uwitec corer (UWITEC, Mondsee, Austria; <http://www.uwitec.at/html/frame.html>; sediment), in May 2016 (Fig. 1). The sediment core was subsampled at 0.5-cm intervals, and samples were stored in plastic bags at 4 °C, within 3 hours of core retrieval.

### Sediment dating, geochemistry, and correlation

Sediment dating was conducted in the Liverpool University Environmental Radioactivity Laboratory by radiometric ( $^{137}\text{Cs}$ ,  $^{210}\text{Pb}$ ) measurement of freeze-dried subsamples, according to the methods described in Appleby et al. (1986, 1992). The age-depth model and sedimentation rate were determined using the corrected Constant Rate of Supply (CRS) Model, for which the 1986 (Chernobyl)  $^{137}\text{Cs}$  peak is used as a reference depth (Appleby et al. 2001). The  $^{210}\text{Pb}$  activity was determined via its gamma emissions, and  $^{226}\text{Ra}$  by the  $\gamma$ -rays emitted by its daughter isotope  $^{214}\text{Pb}$ , following three weeks storage in sealed containers to allow radioactive equilibration. Sediment

water content was calculated by weight loss after freeze-drying. The acid-soluble concentration of Zn was analyzed in 21 samples at the Metropolilab Helsinki, which is an accredited (FINAS T058) testing laboratory. We also used previously analyzed geochemical data to assist in our analysis of the impacts of metal pollution (arsenic, cadmium, cobalt, chromium, copper, iron, nickel, lead, zinc, sulfur and molybdenum) and nutrient enrichment (diatom-inferred total phosphorus [DI-TP]) on Lake Kirkkojärvi. DI-TP data was digitized from Kihlman and Kauppila (2010) whereas for the metals data (Parviainen et al. 2012), the original values were used. We correlated our sediment core with the sediment cores recovered a decade ago by Kihlman and Kauppila (2010) and Parviainen et al. (2012), using the  $^{137}\text{Cs}$  and Zn stratigraphies.

### Cladoceran analysis

Cladoceran sample preparation and analysis was conducted according to the guidelines in Korhola and Rautio (2001) and Kurek et al. (2010), whereas species identification and nomenclature were based mostly on Szeroczyńska and Sarmaja-Korjonen (2007). At least 144 individuals were identified in each sample. Number of individuals was based on the most numerous subfossil component. Relative abundances of each taxon were calculated.

### Numerical analysis

We used principal components analysis (PCA) to assess patterns in the cladoceran assemblages through time, and to present changes in sedimentary metal concentrations. Skewed community data were square root transformed, and the metal concentration data from Parviainen et al. (2012) were log transformed prior to analysis. For each sample, we calculated the Shannon diversity index ( $H'$ ) and species richness using the rarefaction procedure (Birks and Line 1992). We used the constrained optimal sum of squares with untransformed percentage data (Birks and Gordon 1985) and associated broken-stick model (Bennett 1996) to identify statistically distinct zones in the cladoceran biostratigraphy. PCA, Shannon  $H'$ , and rarefaction were conducted using PAST statistics 3.06 (Hammer 2001), and zoning was done using the program ZONE 1.2 (Lotter and Juggins 1991). Locally weighted smoothing (LOESS) was applied to both the metal (Parviainen et al. 2012) and DI-TP data (Kihlman and Kauppila 2010).



## Results

### Sediment geochemistry

Total  $^{210}\text{Pb}$  activity decreased gradually with increasing depth in the sediment until 20 cm, followed by a rapid decline to 25 cm, where  $^{210}\text{Pb}$  reached background levels. Sediments below 25 cm appear to have a different mineralogical composition, as indicated by lower  $^{226}\text{Ra}$  activity than sediments above 25 cm. Sedimentation rate increased from a mean value of  $0.53\text{ cm yr}^{-1}$  during the 1980s to  $0.73\text{ cm yr}^{-1}$  in post-2000 samples. The highest  $^{137}\text{Cs}$  peak occurs at 18.0–18.5 cm. The water content data indicated the presence of a dense sediment layer at 21.5–26.0 cm, and a pronounced Zn peak was observed at a depth of 21.5 cm (Fig. 2).

### Cladoceran communities

Cladoceran remains were numerous and relatively well preserved throughout the core. A total of 32 taxa were identified from 32 samples. The average species richness was 14.8 (SD 2.3) and the average diversity was 1.37 (SD 0.3). The most abundant taxon was *Eubosmina longispina* (Leydig 1860) (64.2 %, SD 11.4), followed by *Bosmina longirostris* (O.F. Müller 1785) (11.3 %, SD 10.9), *Chydorus sphaericus* (O.F. Müller 1785) (5.3 %, SD 2.6), *Alona affinis* (Baird 1843) (2.6 %, SD 1.1), and *Daphnia* spp. (O.F. Müller 1785) (2.0 %, SD 2.8). The majority of the most abundant species exhibit changes in the record, however some species, e.g. *Alonella nana* (Baird 1843) and *Leptodora kindtii* (Focke 1844), do not show clear stratigraphic changes. Results are summarized in Figs. 3 and 4, and detailed species data are presented in Electronic Supplementary Material [ESM] (Table 1). Zonation analysis divided the cladoceran stratigraphy into three statistically significant zones (ZIII, ZII and ZI; Fig. 3), and the grouping is also clearly visible in the PCA biplot (Fig. 4). The highest loading in PCA axis 1 was detected for *B. longirostris*.

### ZIII 44.5-32.5 cm (late 19<sup>th</sup>-early 20<sup>th</sup> century)

Samples in ZIII (Fig. 3) are grouped in the top left quadrant of the PCA biplot (Fig. 4). The planktonic community is dominated by relatively large species such as *E. longispina*, *Daphnia* spp. and *Eubosmina coregoni* (Baird 1857). The species composition is relatively stable, but exhibits decreasing relative abundance of *E. coregoni* towards the top of the zone. *Monospilus dispar* (Sars 1862), *Limnosida frontosa* (Sars 1862) and *Rhyncotalona falcata* (Sars 1903) are the most abundant littoral species. Average species richness and diversity were 14.1 and 1.41, respectively. The proportion of planktonic taxa increases slightly in ZIII (Fig. 3).

### ZII 30.5-23.5 cm (early 20<sup>th</sup> century to early 1970s)

Samples in ZII (Fig. 3) exhibit the lowest levels of diversity and species richness, and the proportion of planktonic taxa declines at the top of ZII. Samples plotted in the bottom left quadrant of the PCA biplot (Fig. 4). Remains of planktonic *E. longispina* are dominant. *Chydorus sphaericus* exhibits increasing relative abundance in ZII (Fig. 3) and the predatory *Bythotrephes longimanus* (Leydig 1860) is present only in ZII (ESM Table 1). In contrast, many other species are present only rarely, or even disappear from the record (e.g. *Polyphemus pediculus* (Linnaeus 1761), *E. coregoni*, *Daphnia* spp., *Alonella excisa* (Fischer 1854), *Alona quadranqularis* (O.F. Müller 1785), *Alonopsis elongata* (Sars 1861) and *Disparalona rostrata* (Koch 1841)) (ESM Table 1). Average species richness was 12.7 and diversity was 0.92 (Fig. 3).

### ZI 22-0 cm (early 1970s to 2016)

The samples in ZI (Fig. 3) are situated on the right side of the PCA biplot (Fig. 4). This zone is characterized by a pronounced increase of relative abundance of *B. longirostris* and simultaneous decrease of *E. longispina*. Also, *E. coreogni* appears again in Lake Kivijärvi, and some littoral species such as *D. rostrata*, *Acroperus harpae* (Baird 1835), and *Pleuroxus unicatus* (Baird 1850) increase in relative abundance (Fig. 3; ESM Table 1). Average species richness was 14.9, and diversity was 1.53 (Fig. 3).

## Discussion

### Sediment geochemistry

The largest  $^{137}\text{Cs}$  peak (1971 Bq/kg) at 18-18.5 cm indicates the 1986 Chernobyl reactor accident (Appleby et al. 1991) as the lake sediments in the region are characterized by very high  $^{137}\text{Cs}$  concentrations at 1986 (Ilus et al. 1993). The secondary peak at 8 cm, driven by one data point only, is considered insignificant. Such features are common and may be caused by numerous processes in the catchment (Ilus and Saxén 2005). Because the  $^{210}\text{Pb}$  activity reaches background at 24 cm, the dates below this depth are unreliable. We do not know whether the sediments below 25 cm were deposited in the early 19<sup>th</sup> century, prior to the first water level manipulation. Low water content at depths between 21.5 and 26.0 cm could be related to the presence of mined materials that have different water retention properties. The Zn peak at the top of the dense sediment layer (21.5 cm) and the  $^{137}\text{Cs}$  record are in good agreement with the results of Kihlman and Kauppila (2010) and Parviainen et al. (2012), suggesting similar geochemical stratigraphies. In our sediment core, highest metal concentrations are at about 20-22 cm depth and mark the timing of the high-magnitude AMD. Absence of  $^{210}\text{Pb}$  and  $^{137}\text{Cs}$  in the dense sediment section (21.5-26.0) suggests presence of tailings material, which originated from the mined rock and was not exposed to atmospheric radionuclide fallout. Similar dilution phenomena have been detected before in lake sediments impacted by mining (Couillard et al. 2004; McDonald and Urban 2007). The mined material was probably transported to the lake before the dam was constructed (1940–1954). It may, however, have originated from direct dumping (1950–1960) or the dam failure ca. 1960. The rate of sedimentation for the dense section, however, is unknown. Diagenetic redistribution of elements (Outridge and Wang 2016) and elemental transport via pore water (Boudreau et al. 2013) cannot be ruled out. Corresponding, distinct metal peaks, however, were also detected in adjacent Lake Viljakkalanselkä (Parviainen et al. 2012), where the dense section was absent.

## Cladoceran communities

### ZIII 44.5-32.5 cm (late 19<sup>th</sup>-early 20<sup>th</sup> century)

The species present in Lake Kirkkojärvi are common taxa, typically found in Finnish lakes (Nevalainen et al. 2013) and water bodies throughout Europe (Bjerring et al. 2009). In the samples of ZIII, the low variation in cladoceran diversity and species richness suggest relatively stable conditions throughout this period. The clearest directional change is the declining relative abundance of *E. coregoni*. This may reflect the lake level manipulation, because *E. coregoni* is the only strictly pelagic species in the sediment samples (Walseng et al. 2006). It is possible that loss of the southern connection between Lake Kyrösjärvi and Lake Kirkkojärvi during the 19<sup>th</sup> century restricted migration from the substantially larger body of water (Lake Kyrösjärvi). Changes in waterways are known to cause large changes in cladoceran communities (Kerfoot et al. 1999). The proportion of planktonic taxa, however, increases in ZIII, which contradicts the hypothesis of water level decline (Korhola et al. 2000; Nevalainen et al. 2011). Another possible explanation could be changes in predation dynamics. Because other large-bodied species (*Daphnia* spp., *L. frontosa*, *P. pediculus*, *L. kindtii*) do not exhibit distinct shifts in proportional abundance, a change in predation dynamics is unlikely.

### ZII 30.5-23.5 cm (early 20<sup>th</sup> century to early 1970s)

In ZII, a change in the lake ecosystem is clearly visible in the Shannon index values and the species relative abundance data (Fig. 3), and is also observed in the PCA bi-plot as a pronounced community shift (Fig. 4). In addition, the relative abundance of the resilient *C. sphaericus* (Bradbury and Megard 1972; Belyaeva and Deneke 2007; Stankovic et al. 2011) increases at the top of the zone whereas the abundance of *E. longispina* declines. This further indicates deteriorating conditions. The decline in the proportion of planktonic species at a depth of 27 to 24 cm in the core is a result of the declining abundance of *E. longispina* and of the disappearance of *Daphnia* and *E. coregoni*. Mine water has been noted to decrease cladoceran productivity (Kerfoot et al. 1999; Doig et al. 2015), species diversity (Holopainen et al. 2008; Winegardner et al. 2017), and the stability of brood size and community density (Bozelli 1996). In contrast to some paleolimnological studies that reported a dramatic collapse of the cladoceran community simultaneously with distinct metal peaks in the sediment record (Thienpont et al. 2016), the impact in Lake Kirkkojärvi is probably

related to increased input of mineral matter more than a decade before the onset of polymetal AMD. The DI-TP exhibits minor nutrient enrichment, which may also have affected the cladoceran community, but it is not possible to differentiate this from the effects of tailings pollution.

The lowest diversity and richness values are in the depth interval 23.5–27.5 cm, which coincides with the lowest water content in the sediments. According to geochemical data (Parviainen et al. 2012), this sediment section is characterized by an elevated concentration of mineral matter and also minor, but evident, enrichment of some metals. This section most likely originated from mining activities, namely from the deposition of mine waste during the mining, which is also visible in aerial photographs but also from transport of the tailings which were dumped outside the dam. Deposits at 27.5 cm probably date to the 1940s or 1950s. Although the impact of the metals on the lake ecosystem was probably minor, the role of mineral matter may have been of greater importance. The harmful impacts of solids on cladoceran assemblages have been widely documented (EIFAC 1964; Bilotta and Brazier 2008), and the negative effects of turbidity from tailings have also been noted (Garrido et al. 2003). It is likely that high amounts of particles interfere with cladoceran feeding efficiency (Arruda et al. 1983; Kirk 1992). In Lake Kirkkojärvi, the tailings grain size is 1-850  $\mu\text{m}$  and the silt fraction being most abundant (Parviainen 2009). The decreasing relative abundance of *M. dispar* and *R. falcata*, which prefer, but are not restricted to sandy bottoms (Hoffman 1987), may be a result of habitat loss caused by siltation. Despite the fact that aerial photographs suggest an increased flush of mine waste into the lake, no numerical data regarding the amount of suspended solids or turbidity in Lake Kirkkojärvi during the 1940s or 1950s are available.

Another explanation for the deterioration of the cladoceran community can be related to changes in predation dynamics. Data regarding historical changes in the fish community, however, are not available, and the size structure of the cladoceran community does not indicate any clear shifts in predation pressure. Moreover, cladocerans have high dispersal potential (Louette and de Meester 2005; Frisch et al. 2012), and the cladoceran community of Lake Kirkkojärvi (as well as fish and predator invertebrate populations) probably received migrants from adjacent waters (Lake Viljakkalanselkä and Lake Kyrösjärvi), which may have damped the ecological changes.

ZI 22 cm to present (early 1970s to 2016)

Even though mining-related metal pollution has been shown to inflict serious damage to cladoceran communities (Doig et al. 2015; Winegardner et al. 2017), the depth of contamination peaks at around 21.5 cm, is characterized by relatively rich and diverse community (Figs. 3 and 4), reflecting a less degraded ecosystem. According to Kihlman and Kauppila (2010), the metals may not have been in bio-available form during the sedimentation process, and the impact on biota may thus have been minor. Similar results regarding sediment contamination and unharmed zooplankton communities in the vicinity of abandoned mines have been noted elsewhere (Ciszewski et al. 2013). The cladoceran community in Lake Kirkkojärvi did not, however, return to its original composition, i.e. that of ZIII. A similar phenomenon has been reported with respect to earlier cases of industrial pollution (Valois et al. 2011; Dupuis et al. 2015). In Lake Kirkkojärvi, the contemporary cladoceran community is characterized by a high relative abundance of *B. longirostris* and *C. sphaericus*, which are regarded as indicators of nutrient-enriched aquatic ecosystems (Boucherle and Zullig 1983; Hoffman 1987; Nevalainen and Luoto 2013). The rapid proportional increase of *B. longirostris* is simultaneous to Zn peak but the species is regarded as sensitive to heavy metal pollution (Koivisto et al. 1992; Bossuyt and Janssen 2005) suggesting low bioavailability of toxic metals in Lake Kirkkojärvi. The fact that the proportional abundance of *B. longirostris* stays relatively unchanged despite the declining AMD suggests that the heavy metal peak is not the main driver of the *B. longirostris* abundance. According to Kihlman and Kauppila (2010) many planktonic diatom species exhibit elevated abundances in the post 1970s samples. This improved food availability for planktonic *B. longirostris* may partly explain the success of this species during the past decades.

*Chydorus sphaericus* is a resilient species (Zawisza et al. 2007; Sienkiewicz and Gasiorowski 2016) that is capable of utilizing many types of food resources (Ahlgren et al. 1990) and tolerates a wide range of environmental conditions, such as low pH (Belyaeva and Deneke 2007). Thus, it is not surprising that *C. sphaericus* is present in Lake Kirkkojärvi, even though it has been affected by inputs of mine tailings, AMD, and nutrient enrichment. In addition, the increasing dominance of littoral species (e.g. *A. harpae*, *D. rostrata*, and *P. unicatus*) may indicate increased availability of habitats created by greater macrophyte cover. Even though predators may induce changes in zooplankton communities, e.g. via size-selective predation (Brooks and Dodson 1965), the cladoceran community changes in Lake Kirkkojärvi cannot be attributed to predation alone, because the community changes are not similar across species with similar sizes or habitats.

Moreover, evidence from diatom and protist communities also indicates eutrophication in Lake Kirkkojärvi during recent decades. Namely, arcellacean *Cucurbitella tricuspis* and diatoms such as *Aulacoseira ambigua*, *Asterionella formosa*, and *Fragilaria crotonensis*, which are regarded as meso- to eutrophic species (Anderson et al. 1995; Saros et al. 2005; Manoylov et al. 2009), have increased in relative abundance in Lake Kirkkojärvi, and in Lake Viljakkalanselkä, during the past few decades (Kihlman and Kauppila 2010). Similar trends in the trophic status of both water bodies suggest that the underlying mechanism for eutrophication is related to regional changes such as intensified agriculture, rather than mining impact, especially as it is known that after ca. 1950, intensive use of artificial fertilizers caused widespread eutrophication in freshwaters (Räsänen et al. 2006).

#### The cladoceran community shift in Lake Kirkkojärvi

Fish have the potential to cause major changes in zooplankton communities (Brooks and Dodson 1965; Stenson 1976), but in Lake Kirkkojärvi, the cladoceran community change occurred a decade earlier than the reported fish introductions. In addition, fish were not introduced to the Kirkkojärvi embayment, but only to the Kyrösjärvi main basin.

According to Kihlman and Kauppila (2010), phosphorus concentrations in Lake Kirkkojärvi have increased during the past few decades, and the most pronounced change occurred during the mining period. The initial reason for the noticeable, but relatively minor diatom-inferred phosphorus enrichment (from ~16 to ~20  $\mu\text{g L}^{-1}$ ) is difficult to explain, but it was probably related to increased human impact in the catchment. The human population of Viljakkala was highest during the 1950s, corresponding to the time at which there was an increase in the use of artificial agricultural fertilizers and a concurrent impact in the trophic status of freshwater ecosystems in general (Räsänen et al. 2006). Minor phosphorus enrichment was also detected in neighboring Lake Viljakkalanselkä (Kihlman and Kauppila 2010), but the modern cladoceran community there resembles the community structure of ZIII (Leppänen et al. 2017).

In Lake Kirkkojärvi, the reason for the observed pronounced shift in the cladoceran community may be related to reduced ecosystem resilience caused by the mining impact in ZII (23.5-30.5 cm). High species richness strengthens the zooplankton population stability (Downing et al. 2014) and facilitates ecosystem resilience (Downing and Leibold 2010), whereas reduced

resilience tends to increase the likelihood of regime shifts in aquatic ecosystems (Folke et al. 2004). Whereas the nutrient status of Lake Kirkkojärvi had been shifting towards a more eutrophic state for a long period of time, the mining impact during ZII greatly reduced the resilience, which led to the observed community change. Many post-mining communities reflect a trend toward a more nutrient-rich system, among them the protists, diatoms, and cladocerans in Lake Kirkkojärvi. This indicates that a regime shift propagated through multiple trophic levels. The Lake Kirkkojärvi regime change resembles a type of regime shift, defined by Randsalu-Wendrup et al. (2016), where an abrupt change in environmental conditions triggered a direct response in the ecosystem.

## **Conclusions**

In contrast to studies of AMD impact on primary producers (diatoms, protists), the cladoceran community of Lake Kirkkojärvi was clearly impacted by mining activity at the site. Subsequent changes in species composition, however, are in good agreement with the evidence from diatoms and protists that suggest post-mining eutrophication of the lake. We argue that the change in the cladoceran community was triggered by devastating, but relatively short-term impacts of mine pollution. These effects caused the cladoceran community to become less resilient, and thus more susceptible to pronounced change in species composition.

The need for minerals and metals is great in contemporary societies, and the mining industry is seeking environmentally friendly and economically viable techniques to meet these needs. Our results present important information regarding pollution impact assessment. In particular, our study highlights the utility of cladocerans as early warning indicators, and supports the importance of using multiple sediment variables in paleolimnological pollution research. Moreover, this study is an example of how anthropogenic pressures can transform a simple case of lake pollution into a multiple-stressor problem, via recent eutrophication.

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## Figure captions

**Fig. 1 A.** Location of the Haveri (inverted mine symbol) mine and the tailings area (gray), sampling site (red cross), and the approximate shoreline (dotted) of Lake Kirkkojärvi during the early 19<sup>th</sup> century, before mining and lowering of the water level, and the current shoreline. Notice the dam structures (thick black lines). **B.** Location of Lake Kirkkojärvi. The data for the map were downloaded from the National Land Survey of Finland open data databank (<http://www.maanmittauslaitos.fi/en/e-services/open-data-file-download-service>) under the open data CC 4.0 license (<https://creativecommons.org/licenses/by/4.0/>), and customized in ArcMap, Version 10.3.1 (<http://desktop.arcgis.com/en/arcmap/>) and in Corel Draw X8, version 18.0 (<http://www.coreldraw.com/en/product/graphic-design-software/>)

**Fig 2.** Water content, acid-soluble zinc (Zn), <sup>210</sup>Pb and <sup>137</sup>Cs activities, sedimentation rate, LOESS smoothed curves (span 0.7) for diatom-inferred total phosphorus (DI-TP) and axis 1 values from the metals dataset PCA. Horizontal lines indicate cladoceran zone boundaries. \* Data from Kihlman and Kauppila (2010), \*\* Data from Parviainen et al. (2012)

**Fig 3.** Cladoceran stratigraphy, proportion of pelagic taxa (*E. longispina*, *E. coregoni*, *Eubosmina* spp., *B. longirostris*, *Daphnia* spp., *P. pediculus*, *L. kindtii*, *L. frontosa*, *B. longimanus*), species richness and Shannon diversity. Y-axis represents sediment depth and corresponding date (C.E.), and X-axis represents relative abundance (%). Horizontal lines indicate zones ZI, ZII and ZIII. Only taxa that had relative abundances >2% in any of the samples are included. The species are ordered by increasing abundance, from lower left to top right. Complete species data are available in ESM Table 1

**Fig 4.** PCA plot. Filled circles indicate samples, accompanied by sample depth. Dotted line follows the record in stratigraphic order for clear interpretation of community change. Species arrows for 10 species that exhibited the largest loadings on PCA axis 1 and 2 are shown. Samples belonging to the same zones are enclosed in ellipses and identified by ZI, ZII, and ZIII. Variance explained: Axis 1, 53.5 % and eigenvalue of 6.4; Axis 2, 11.5 % and eigenvalue of 1.4