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TECHNICAL ARTICLE

Properties and Stability of Mining-Induced Meromixis in Two Small Boreal Lakes in Eastern Finland

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Abstract
Mine waters are a significant point source stressor for aquatic environments, not only due to their acidity and high metal concentrations, but also because of their high electrolyte concentrations. Ion-rich mine waters can disturb the seasonal mixing of lake waters, even leading to permanent stratification, i.e. meromixis. In this study, we investigated two small natural lakes receiving waters from closed Ni-Cu mines. To characterize the present chemical and physical conditions of these two boreal lakes, we collected water samples and in-situ water column measurements seasonally in 2017 and 2018. We modelled the stability of meromixis in the lakes under varying physico-chemical and meteorological conditions with the MATLAB-based open-source model code, MyLake. Chemical analyses and water column measurements show that both lakes are currently meromictic with a chemocline separating the circulating, well-oxygenated upper water from the non-circulating, hypoxic bottom water. The main anion was SO_4 in both lakes, while the main cations were Ca, Mg, Na, and K. Elevated concentrations of conservative elements flowing from the mine areas are crucial in maintaining the meromixis. Modelling scenarios suggest that the meromixis would be sustained for several decades even if the external load ceased completely. Lake morphology and sheltered surroundings also seem to contribute to maintaining the meromixis in these lakes. Consequently, our results indicate that small headwaters are sensitive to persistent meromixis even when external loading is mild.

Keywords Limnology · Mine environment · Modelling · Stratification · Sulfate

Introduction

Sulfidic mine waters have long been recognized as a major ecological threat to nearby waterbodies (Lottermoser 2007). This is not only due to acidity and the high concentrations of metals, but also because of the water's high electrolyte concentrations (i.e. salinity). For instance, in the currently active Talvivaara/Terrafame polymetal mine area in Finland, an accidental release of high volumes of saline mine water has disrupted the circulation regime of a nearby lake and significantly affected important zooplankton and phytoplankton groups in the lake (Leppänen et al. 2017). Another lake further downstream in the chain of lakes has also changed ecologically because of the saline drainage (Leppänen et al. 2019).

In addition to active mines, closed or abandoned mine sites can continue to affect the environment. The Ministry of the Environment of Finland has identified 37 closed or abandoned mines in the country that could cause serious negative environmental effects due to acidic or metalliferous mine drainage waters or potentially acid-producing wastes (Räisänen et al. 2013; Tornivaara et al. 2018, 2020). It can be assumed that waters emanating from these wastes also have higher electrolyte concentrations than baseline levels. The Finnish landscape, and the whole Fennoscandian Shield, is dotted with small, shallow lakes, which means that most of these mine areas are likely affecting natural lakes. These effects can sometimes be due to several centuries of mining and can persist for several decades after the mines have been abandoned (e.g. Ek and Renberg 2001; Salonen et al.

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2006). However, even relatively short-term mining activity can generate harmful wastes (Fischer et al. 2020).

Saline mine water drainage can disturb the circulation pattern of a lake. The increase in density acts as a stabilizing force and the water column circulates only to a depth where wind-induced mixing is more effective than the stabilizing forces. A chemocline forms between the circulating mixolimnion and stagnant monimolimnion at the depth where equilibrium is reached. This type of a mixing regime is known as meromixis. The density difference between the mixolimnion and monimolimnion needs to be sustained for a lake to remain meromictic. The salinity of natural waters is often caused by NaCl but in mine environments, other salts may dominate and contribute to the density of water. Thus, in this study, the term salinity includes all ions that contribute to the density of the lake water.

We investigated two natural lakes near closed nickel-copper mines in eastern Finland to answer questions regarding the type of meromixis in the lakes and the characteristics that maintain the meromixis. Water samples and water column measurements were collected seasonally to characterize the lakes. In both cases, the mines were active for only a few decades and, although the waste waters from the mines have been processed according to the environmental permits of the time (Räisänen et al. 2015), the mixing regimes in the lakes seem to have changed. We hypothesise that the continuing saline drainage from the mine waste areas is the main agent maintaining the permanent stratification in these lakes. To test this hypothesis, we modelled conditions that would overturn the current stratification in the lakes and tested the stability of the meromixis under varying meteorological and geochemical conditions.

Study Sites

The natural lakes Valkeinen and Sortavalanjärvi are situated in eastern Finland ≈ 110 km apart. Both are relatively small headwaters and receive waters from the closed Kotalahti and Laukunkangas Ni-Cu mines, respectively. The region belongs to the boreal zone, where vegetation is dominated by coniferous forests and the climate is characterized by snowy winters and mild summers.

Lake Valkeinen in the municipality of Leppävirta has a catchment area of 0.93 km^2, one outlet, and three inlets, of which one drains from the mine tailings area (Finnish Environment Institute 2014a, Fig. 1, Table 1). Lake Sortavalanjärvi is in the municipalities of Enonkoski and Savonlinna and has a catchment area of 3.1 km^2 (Finnish Environment Institute 2014a, Fig. 2, Table 1). The lake has four inlets, of which one drains from the mined area east of the lake, and one outlet at the western end of the lake (Fig. 2).

The Kotalahti Mine

The Kotalahti mine is situated to the north of Lake Valkeinen. The mine was active from 1957 to 1987 (Puustinen 2003). It had three underground mines and two open pit mines. The open pit mines were filled with tailings waste and water after active mining ceased (Räisänen et al. 2015). The main ore minerals were pyrrhotite, pentlandite, chalcopyrite, and pyrite (Papunen and Koskinen 1985). The ore deposit had average mill feed concentrations of 0.66% Ni, 0.26% Cu, and 3.8% S (Puustinen 2003). Approximately 14 Mt of rock was mined, of which 12.5 Mt was ore (Puustinen 2003). The mine waste area includes 9.4 Mt of tailings with a surface area of 0.75 km^2 (Räisänen et al. 2013, 2015).

Waters from the tailings area flow primarily north, i.e. away from Lake Valkeinen to wetland basins, and then to a treatment plant, from which the waters are piped to Lake Oravilahti (ISAVI 2014). Kauppila and Räisänen (2015) sampled the tailings seepage waters in 2009 in the northeast side of the tailings area and classified them as near-neutral (pH 6.5), high-metal waters with a sum of dissolved trace metals (Zn, Cu, Cd, Pb, Co, Ni) of ≈ 2000 µg L^{-1}, which includes ≈ 1600 µg L^{-1} Ni. In addition, the seepage waters contained ≈ 3200 mg L^{-1} of SO_4.

The Laukunkangas Mine

The Laukunkangas mine, east of Lake Sortavalanjärvi, was active from 1984 to 1994. The main ore minerals were pyrrhotite, pentlandite, and chalcopyrite (Grundström 1985). The average mill feed concentrations were 0.76% Ni and 0.22% Cu (Puustinen 2003). Approximately 8.4 Mt of rock was mined, of which 6.7 Mt was enriched ore (Puustinen 2003). The mine waste area includes 6.60 Mt of tailings south-east of the mine on an area of ≈ 0.6 km^2 (Räisänen et al. 2013, 2015). The tailings area also includes 0.22 Mt of tailings from two nearby satellite mines (Räisänen et al. 2013).

Two settling basins were constructed in the northern part of the waste area during the active mining period and were used as settling and storage ponds for recirculated process waters and dewatering waters (Pöyry and Isomäki 1996). After mining ceased in the 1990s, the waste areas were covered with a 30–50 cm thick till layer and the two settling basins were converted to wetlands by damming and flooding (Räisänen 2015; Räisänen et al. 2015). According to sampling conducted in 2002 and 2005 by the Geological Survey of Finland, the sediments of the wetland basins retained 60% of the S and Fe and 75–90% of other sulfidic metals of the waste area seepage waters.

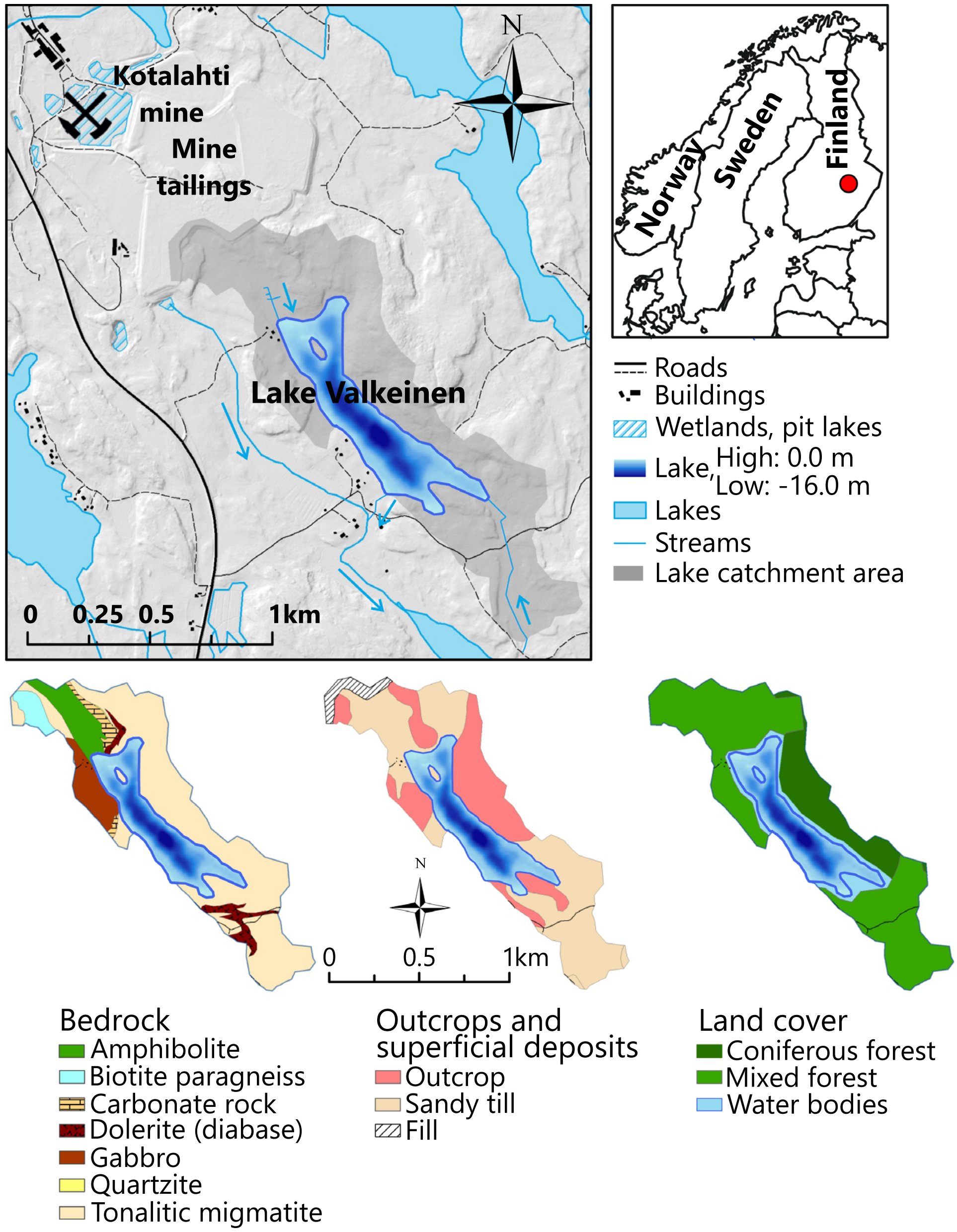


Fig. 1 Lake Valkeinen and Kotalahti mine. Maps modified from Finnish Environment Institute (2014a; 2014b), and Geological Survey of Finland (2015; 2016)

The waters draining into Lake Sortavalanjärvi contained on average 0.34 mg L^{-1} Ni and < 0.5 mg L^{-1} Fe and Mn (Räisänen et al. 2015). Kauppila and Räisänen (2015) sampled the tailings seepage waters in 2009 and classified them as high-acid (pH 2.5–3) and high-metal with a sum of dissolved trace metals (Zn, Cu, Cd, Pb, Co, Ni) of 2000 µg L^{-1}. The concentrations were ≈ 1900 µg L^{-1} for Ni, ≈ 600 mg L^{-1} for Fe, and ≈ 5500 mg L^{-1} for SO_4 (Kauppila and Räisänen 2015).

Materials and Methods

Water Column Measurements

Continuous water column measurement profiles were taken at the deepest part of the lakes (≈ 16 m). At Lake Valkeinen, measurements were made once or twice a month, excluding July, from late February to late October

Table 1 Location, meteorological, and morphometrical characteristics of the study lakes

Lake name	Valkeinen	Sortavalanjärvi
Location	62° 33' N 27° 38' E	62° 2' N 28° 45 E
Meteorology		
Annual mean air T°	+ 3.7 °C[1]	+ 4.3 °C[2]
Annual mean precipitation	611 mm[1]	604 mm[2]
Mean air T° coldest month (Feb)	− 8.2 °C[1]	− 7.5 °C[2]
Mean air T° warmest month (July)	+ 17.2 °C[1]	+ 17.6 °C[2]
Lake morphometry[3]		
Surface area	0.19 km^2	0.24 km^2
Maximum depth	16 m	16 m
Average depth	5.3 m	5.3 m
Relative depth[4]	3.5%	2.9%
Volume	1.01 million m^3	1.25 million m^3
Water residence time	> 1000 days	379 days

[1]Kuopio Maaninka station, Finnish Meteorological Institute (FMI), AD 1989–2018

[2]Savonlinna Punkaharju station, FMI, AD 1989–2018

[3]Operational VEMALA nutrient loading model for Finnish Watersheds (Huttunen et al. (2016))

[4]Calculated according to Wetzel (2001)

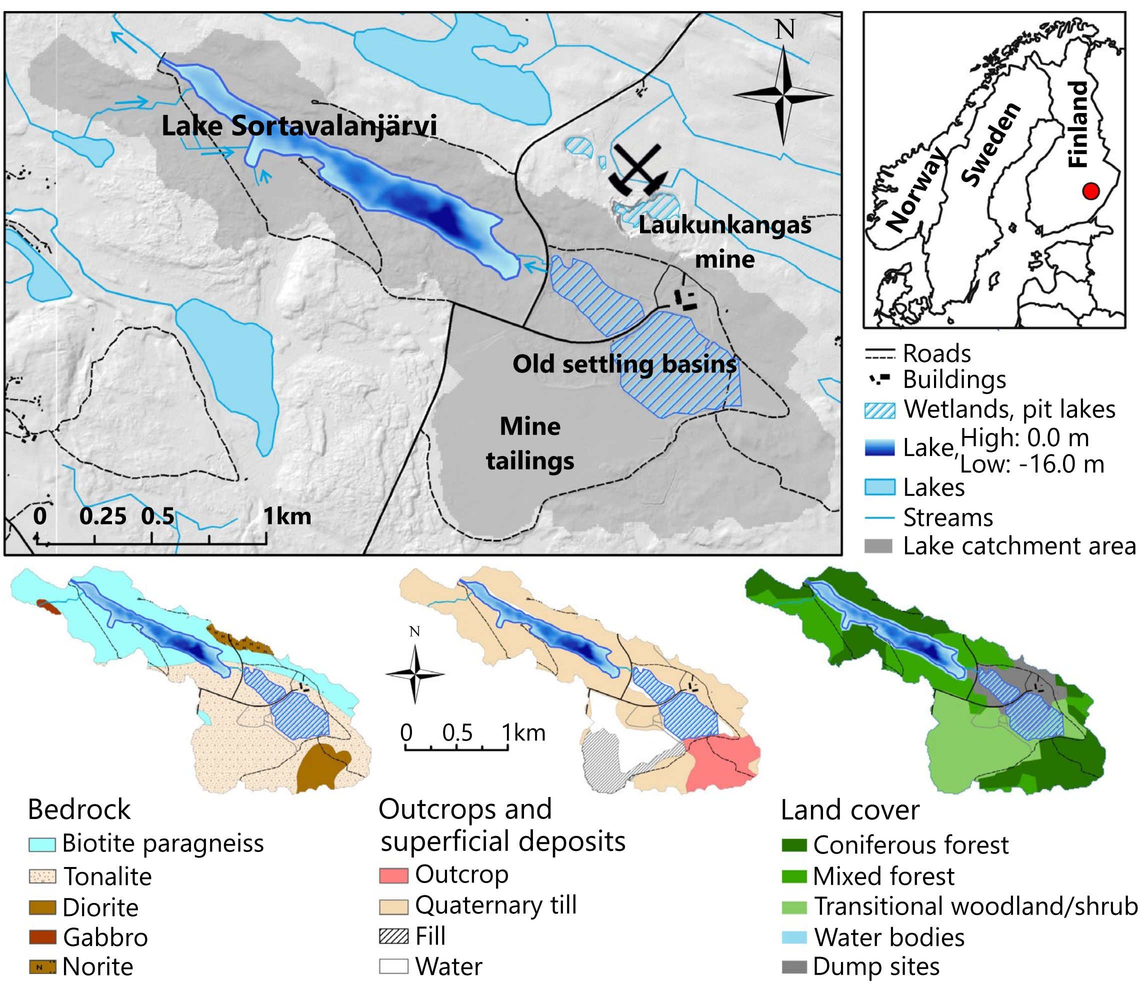


Fig. 2 Lake Sortavalanjärvi and Laukunkangas mine. Maps modified from Finnish Environment Institute (2014a; 2014b), and Geological Survey of Finland (2010; 2016)

of 2017 and again in February 2018. At Lake Sortavalanjärvi, measurements were made in February and June of 2018. The measured variables included depth, temperature, pH, electrical conductivity (EC), reduction potential (ORP), and dissolved oxygen (DO). The measurements were taken with Yellow Springs Instruments XL600 and Professional Plus multiparameter sondes.

Water Samples

Water column samples were collected from different depths of the deepest part of the lake basin on Oct. 24th, 2017 (2, 9.5, 10, and 13 m of max depth 16 m), and Feb. 20th, 2018 (2, 9.5, 10, and 14 m) at Lake Valkeinen. Lake Sortavalanjärvi was sampled on Nov. 2nd, 2017 (2, 8.5, 9, and 12 m), and Feb. 21st, 2018 (2, 8, 8.5, 12, 14.5, and 15.5 m of max depth 16 m). Additionally, lake inlets and outlets were sampled at Lake Valkeinen in 2017 and 2018 and at Lake Sortavalanjärvi in 2017.

The water samples for cation analyses were filtered with 0.45 µm syringe filters into 100 mL bottles and preserved with nitric acid. Samples for anion analyses were taken into 500 mL bottles unfiltered. Alkalinity was measured from the samples in the field using a Hach titrator with both colour indicator and a pH meter. The element concentrations of the samples were analysed with inductively coupled plasma optical emission spectrometry/mass spectrometry (ICP-OES/MS) and anions with ion chromatography (IC) by the accredited (FINAS T025) testing laboratory Labtium Ltd. Laboratory duplicates (every 10th sample and one for each batch) and certified reference materials were used for quality control.

Stream flows were measured in 2019 using either v-weirs or graduated vessels and a stopwatch for both the incoming (from the mines, also from the south at Lake Valkeinen) and outflowing waters. Flows were measured twice for Lake Valkeinen and nine times for Lake Sortavalanjärvi. The water flow in the ditch from the tailings area to Lake Valkeinen was very slow and difficult to measure.

Continuous field measurements of water temperature were carried out at both lakes to support stratification modelling. Hobo field loggers were installed at the deepest part of both lakes in June 2019 at 1, 2, 8, 10, 14, and 15 m depths for Lake Valkeinen, and at 1, 3, 6, 9, 13.6, and 14.8 m depths for Lake Sortavalanjärvi. The loggers were removed in early December 2019 (Lake Valkeinen) and late October 2019 (Lake Sortavalanjärvi).

Modelling

The stratification in the lakes was modelled using the MATLAB-based open-source modelling software, MyLake (multi-year lake simulation model) originally developed by Saloranta and Andersen (2007). MyLake is a one-dimensional process-based model code for simulation of daily vertical distribution of lake water temperature and, thus, density stratification, evolution of seasonal lake ice and snow cover, sediment–water interactions, and phosphorus-phytoplankton dynamics. While being mainly a process-based model, some of its processes are phenomenological rather than strictly physical. The original version of MyLake assumed pure water, so a version was used that takes salinity-induced electrical conductivity into account when calculating density. An improved formulation for density (Moreira et al. 2016) was used:

$$\rho\left(T, \kappa_{25}\right) = \rho_W(T) + \kappa_{25}\left(\lambda_0 + \lambda_1(T - 25^oC)\right) \quad (1)$$

where ρ_W is the density of pure water and λ_0 and λ_1 are coefficients that reflect the temperature dependence of the density contribution of the solutes. MyLake requires cloud coverage, air temperature, relative humidity, air pressure, wind speed, precipitation, inflow discharge, and its temperature as a standard input, along with the new addition of electrical conductivity. Values for parameters λ_0 and λ_1 were estimated using the RHOMV program available online (Moreira 2014).

Internally, MyLake uses 1 m layer depths when performing calculations. To model the circulation of water between different depth regions in the lakes, the water columns of the lakes were divided into layers based on the depths of the thermocline and chemocline. The layers were 0–4 m, 4–8 m, 8–10 m, and 10–16 m. Several scenarios were run for both lakes. These scenarios represented differing amounts of mine water load and changing meteorological and climate conditions. For mine water load, scenarios of no mine load and 50% of current load were run. IPCC Fifth Assessment Report (AR5) greenhouse gas emission scenarios (representative concentration pathway, RCP) 2.6 and 8.5 were used for changing climate conditions (IPCC 2014). AR5 scenarios project increases of 1.5–2.0 °C (RCP2.6) and 5–7 °C (RCP8.5) for temperature and increases of 0–10% (RCP 2.6) and 10–20% (RCP 8.5) for precipitation, relative to the 1986–2005 period. Additionally, scenarios with increased wind speeds (an increase of 10%, and 2 m s^{-1}) were run. The timeframe for the plots was from AD 2010 to AD 2129, with annual plotting until AD 2020 and 10-year gaps between each plot afterwards.

Boundary, Forcing, and Initial Data

Meteorological variables (cloud coverage, air temperature, relative humidity, air pressure, wind speed, precipitation) were obtained from the closest meteorological station (Savonlinna Airport for Lake Sortavalanjärvi and Kuopio Ritoniemi for Lake Valkeinen) and the global radiation was calculated by MyLake. As there are no continuous discharge measurements for the study lakes, incoming discharges were evaluated based on available rain information. For Lake Valkeinen, the discharge was set at:

$$Q = \begin{cases} 0, \textit{if air temperature was} < -5 \\ \frac{3600*24}{1000}\left(\left(\sum_{i=n}^{n-2} rain(i)\right) * 0.174 + 0.61304\right) \end{cases} \quad (2)$$

For Lake Sortavalanjärvi, the inflow discharges were set to be almost uniformly distributed random variables between 62.68 and 187.90 m^3/day. Electrical conductivity was also assumed to be uniformly distributed random variables, between 789.50 and 2368.50 µS cm^{-1} for Lake Valkeinen and between 400 and 1200 µS cm^{-1} for Lake Sortavalanjärvi. Initial conditions for temperature and conductivity were derived from the water column measurements.

Results

Water Column Measurements

The water columns of both lakes remained stratified throughout the sampling period (Figs. 3 and 4). The chemocline occurred between 8 and 10 m with some fluctuation between seasons. Temperature, DO, EC, and pH showed clear differences between mixolimnion and monimolimnion throughout the year. In Lake Sortavalanjärvi, there was an increase in DO levels to 1.7 mg L^{-1} at the very bottom of the water column in February 2018 (Fig. 4). Concurrently, EC and ORP increased, and pH decreased.

Water Samples

Lake Valkeinen

The main anions in Lake Valkeinen were SO_4 and Cl, while the main cations were Ca, Mg, K, and Na (Fig. 5). All correlated with EC (r = 0.96–0.98, p < 0.0001). Other elements with notable concentrations were P, Mn, Ni, Cu, and Zn. Most of the main anion and cation concentrations were notably higher in the inflow from the mine area than from the southern inflow to the lake. The stream flow in the southern inflow was 0.7 l s^{-1} on Oct. 26th, 2019, and 1.1 l s^{-1} on Dec. 4th, 2019. Measurements could not be obtained from the mine inflow due to low flow rates. The corresponding outflow from Lake Valkeinen was 8.5 l s^{-1} (Oct. 26) and 11 l s^{-1} (Dec. 4).

During the autumn overturn in October 2017, most of the main elements had uniform concentrations in the mixolimnion and monimolimnion with a small increase at the chemocline. Other elements showed more variation with depth. P concentrations were very high and fluctuated slightly with depth in the mixolimnion. Fe concentrations were very low in the lake water. During the winter stagnation in February 2018, the increase in the concentrations of main elements with depth was stronger than in the corresponding autumn overturn samples.

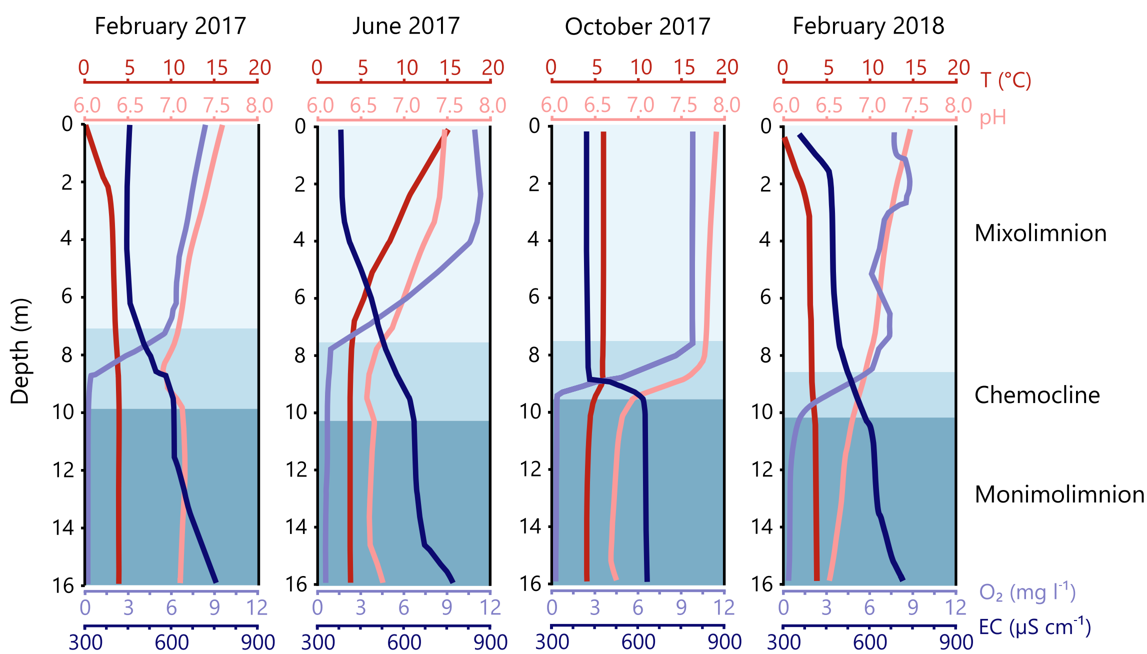


Fig. 3 Temperature, pH, oxygen, and electrical conductivity measurements from the water column of Lake Valkeinen

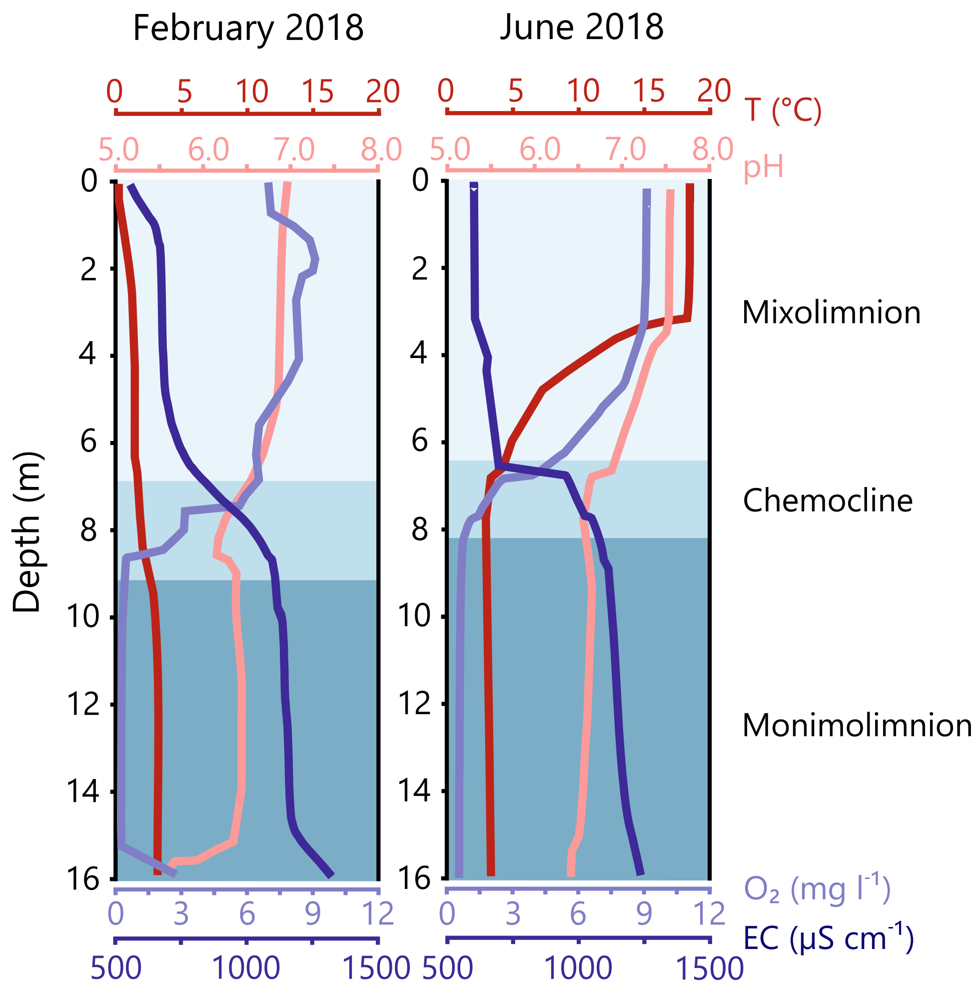


Fig. 4 Temperature, pH, oxygen, and electrical conductivity measurements from the water column of Lake Sortavalanjärvi

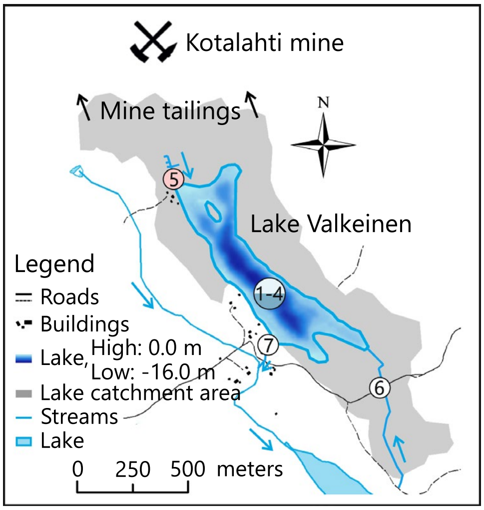
Lake Sortavalanjärvi

The main anions were SO_4 and Cl, and the main cations were Ca, Mg, Na, and K in Lake Sortavalanjärvi (Fig. 6). Notable trace elements included Mn, Fe, Ni, Cu, and Zn. The concentrations of main cations and anions and most trace elements were elevated in the mine stream compared to streams inflowing from elsewhere into the lake (Fig. 6). Dissolved organic carbon (DOC) and Fe were higher in the stream flowing from Muhelampi Pond east of the lake than in the mine area inflow. Mean flow in the stream from the mine area was 6.2 L s^{-1} (2.7.2019–4.12.2019, nine measurements), with minimum inflow 0.5 L s^{-1} on Oct. 3rd, and maximum inflow 15 L s^{-1} on Nov. 20th. Mean flow in the stream flowing out from Lake Sortavalanjärvi was 18.9 L s^{-1} (min 0.9 L s^{-1} on July 2nd, max 35 L s^{-1} on Nov. 20th).

The water column of Lake Sortavalanjärvi had an increasing trend in main anion and cation concentrations until the depth of 12 m during both sampling occasions. Concentrations of these substances showed little variation between the autumn and winter samples. In contrast, most trace elements had lower concentrations in the autumn (Nov. 2017) samples.

Modelling of Stratification

According to the MyLake model for EC (Fig. 7), the EC in Lake Valkeinen would approach zero, or equilibrium, around AD 2050, if the current loading from the mine area was to cease completely. For Lake Sortavalanjärvi, this would occur around AD 2094 (Fig. 8). Simulated epilimnetic and hypolimnetic EC values did not markedly differ and showed little seasonal fluctuation for Lake Valkeinen. For Lake Sortavalanjärvi, these values differed somewhat and showed some seasonal fluctuation, which lessened over time as the values reached zero. If the loading from the mine area to Lake Valkeinen were to decrease by 50% from the current EC to ≈ 200 µS cm^{-1}, equilibrium would be reached around AD 2040. For Lake Sortavalanjärvi, a 50% decrease from the current EC to ≈ 400 µS cm^{-1} means equilibrium would be reached around AD 2060. For both lakes, epilimnetic EC values were lower and showed less fluctuation than the hypolimnetic values in the 50% load scenario.



	Sample	Water body	Depth (m)	Description
Deep profile	1	Lake	2	Upper water
Deep profile	2	Lake	9.5	Water above chemocline
Deep profile	3	Lake	10	Water below chemocline
Deep profile	4	Lake	13-14	Bottom water
	5	Stream		Inflow from the mine area
	6	Stream		Inflow from south
	7	Stream		Outflow from the lake

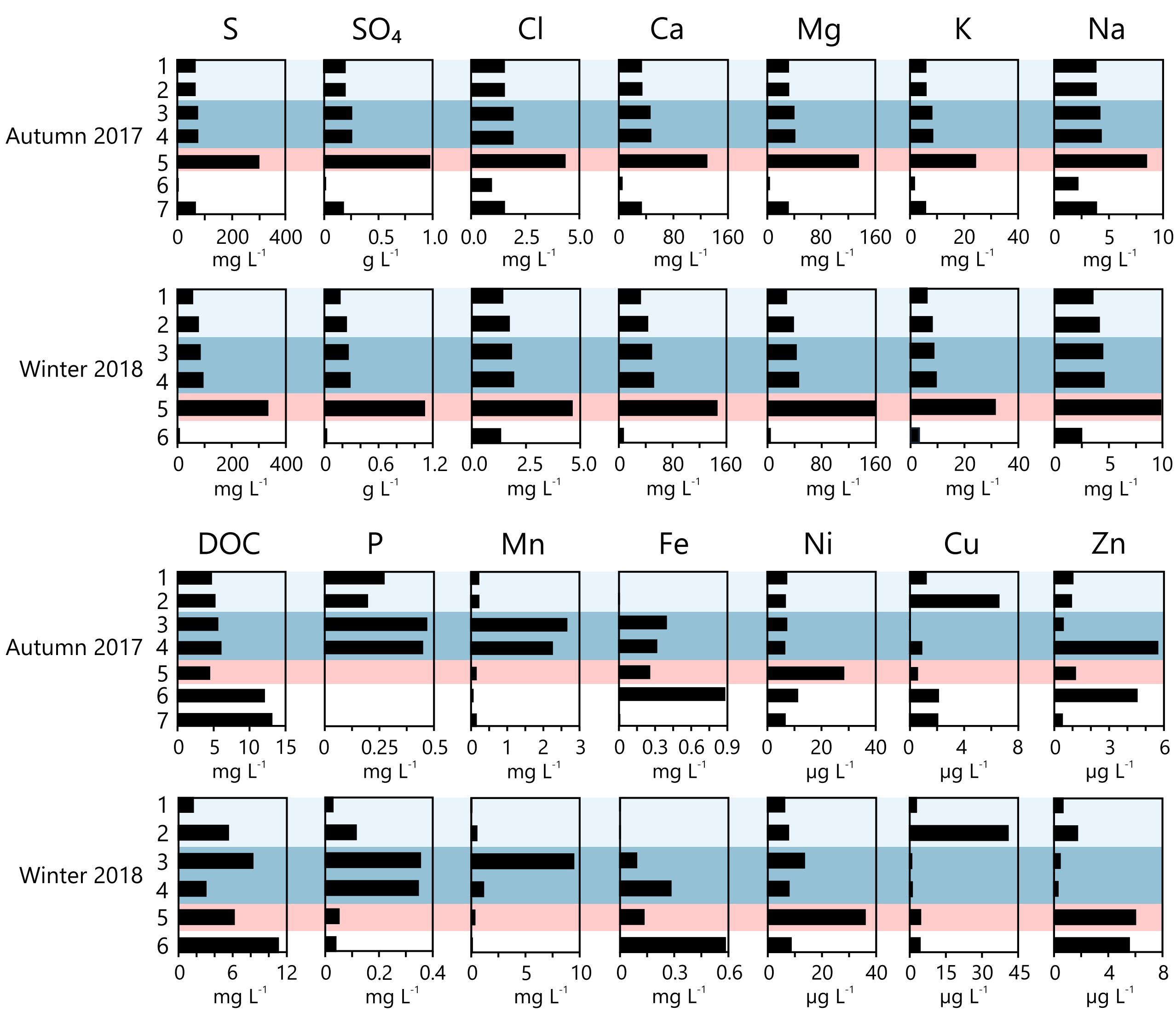
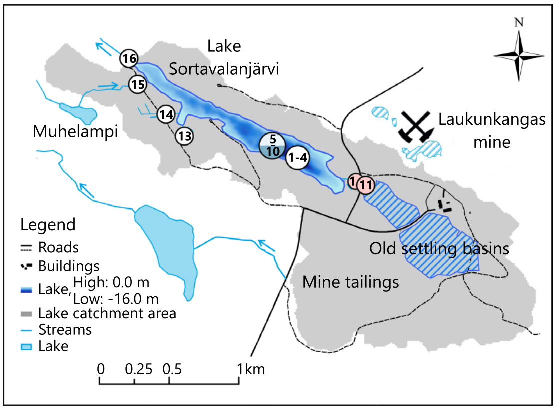


Fig. 5 Water samples taken from Lake Valkeinen 24.10.2017 and 20.2.2018. Concentrations of selected elements from water samples from Lake Valkeinen and inflows/ouflows. Note the differing x-axes between years for some elements. S is total sulfur concentration. P concentration in the inflow from south (sample 6), Oct 2017, was discarded for anomalously high value



Sample	Water body	Depth (m)	Description
1	Lake	2	Upper water
2	Lake	8.5	Water above chemocline
3	Lake	9	Water at chemocline
4	Lake	12	Bottom water
5	Lake	2	Upper water
6	Lake	8	Water above chemocline
7	Lake	8.5	Water at chemocline
8	Lake	12	Water below chemocline
9	Lake	14.5	Water below chemocline
10	Lake	15.5	Bottom water
11	Stream		Inflow from mine, before road
12	Stream		Inflow from mine, after road
13	Stream		Inflow from the south
14	Stream		Inflow from Sortavalansuo
15	Stream		Inflow from Muhelampi
16	Stream		Outflow from the lake

Deep profile: samples 5–10

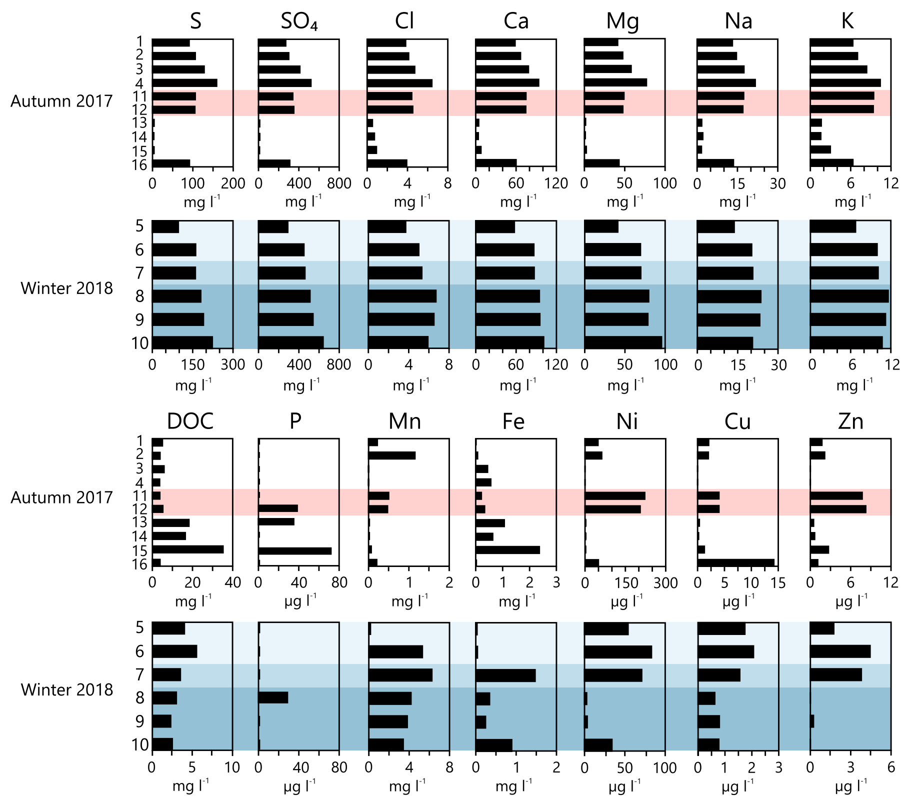


Fig. 6 Water samples taken from Lake Sortavalanjärvi 2.11.2017 and 21.2.2018. Concentrations of selected elements from water samples from Lake Sortavalanjärvi and inflows/ouflows in 2017 and 2018. Note the differing x-axes between years for some elements. S is total sulfur concentration

The water circulation scenarios showed that if the loading from the mine area was 50% of the current load (‘50% mine load-scenario’), the water would circulate briefly between the different layers of Lake Valkeinen (Fig. 8) during the autumn overturn, but there would be very little circulation for the rest of the year. Lake Sortavalanjärvi (Fig. 9) also showed some circulation during spring. If there was no load from the mine area (‘no mine load-scenario’), the autumn overturn lasts longer, more water is transferred, and there is additional mixing during winter and spring at both lakes. Scenarios where wind speeds were increased (not shown), either by 2 m s^{-1} or by 10%, also led to the water column circulating during the autumn but less so at other times of the year. At Lake Valkeinen, the more severe climate change scenario (annual temperature increased 7 °C and precipitation by 10%) also generated prolonged autumnal mixing and some additional mixing during spring. At Lake Sortavalanjärvi, this scenario produced less autumnal mixing and very little mixing during spring compared to the other scenarios. Instead, the RCP 2.6 scenario with increases of

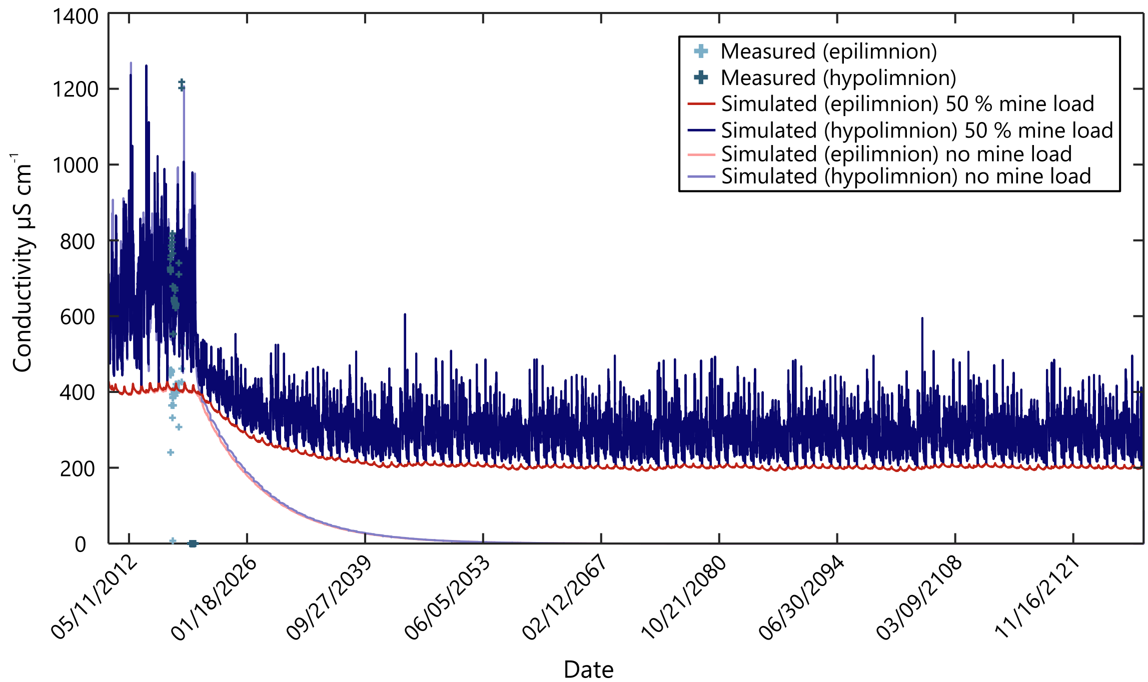


Fig. 7 Measured and simulated electrical conductivity timeseries for surface (epilimnion) and bottom (hypolimnion) waters at Lake Valkeinen AD 2010–2129. "50% mine load"-scenario in darker and "no mine load"-scenario in lighter colours

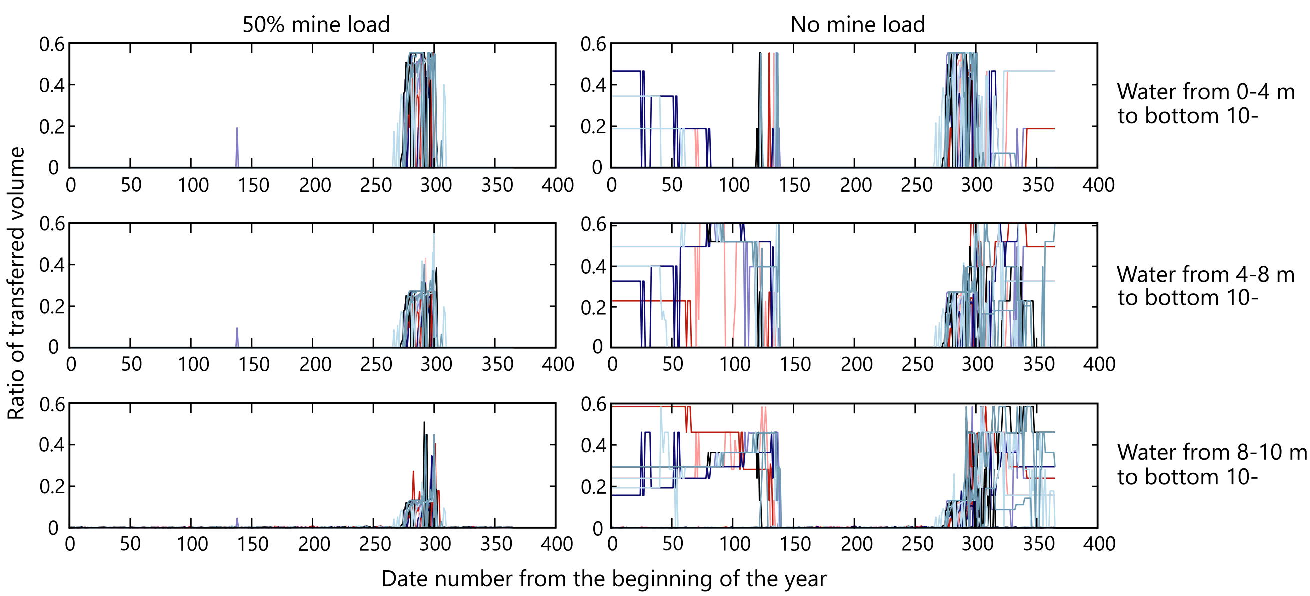


Fig. 8 Modelled ratio of transferred water volume between different water depth layers at Lake Valkeinen in the "50% mine load"-scenario (on the left) and "no mine load"-scenario (on the right). Different colours represent different plots, with each plot being one year. Plotting is annual from AD 2010 to AD 2020, after which there is a 10-year gap between each plot until AD 2129

2 °C in temperature and 10% in precipitation led to more circulation during both autumn and spring. The models performed better for Lake Valkeinen because the limited data hampered the usability of the MyLake models for Lake Sortavalanjärvi.

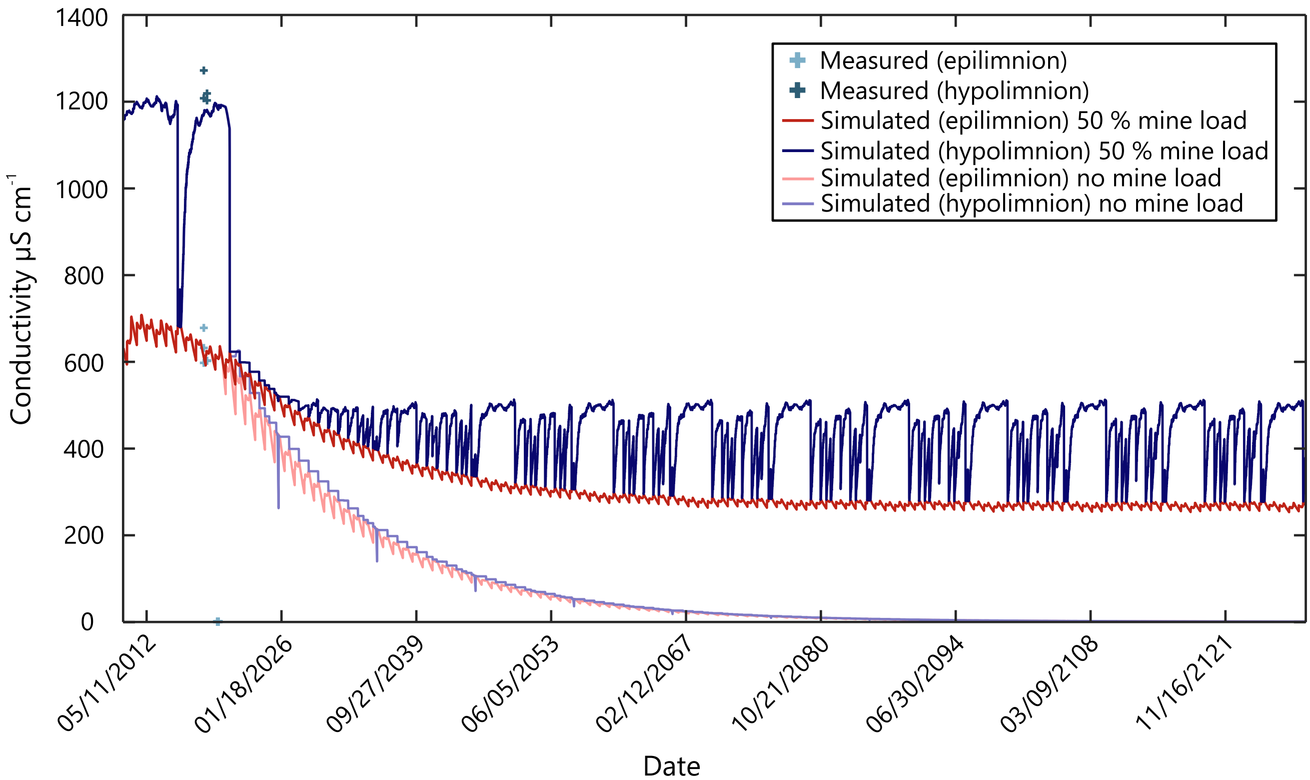


Fig. 9 Measured and simulated electrical conductivity timeseries for surface (epilimnion) and bottom (hypolimnion) waters at Lake Sortavalanjärvi AD 2010–2129. "50% mine load"-scenario in darker and "no mine load"-scenario in lighter colours

Discussion

Stratification of the Lakes

The water columns of Lakes Valkeinen and Sortavalanjärvi were chemically stratified throughout the sampling period. The mixolimnion of both lakes exhibited temperature, DO, pH, and EC depth profiles typical of dimictic lakes (Figs. 3 and 4) with inverse stratification in winter, thermal stratification in summer, and an overturn in spring and autumn (e.g. Fojutowski et al. 2021). The depth and gradient of the chemocline varied seasonally in both lakes, as has been observed in other meromictic lakes (Boehrer et al. 2017). During the autumn overturn, the chemocline was eroded by the well-circulated mixolimnetic waters and there were clearer shifts with depth in pH and EC (Figs. 3 and 4). The monimolimnion remained mostly anoxic and was thermally very stable at 3–4 °C throughout the sampling period in both lakes. This reflects the temperature of maximum density of water and the average annual temperature of the area (Table 1). Seasonal changes in the other variables were subtle, with some variation in the values of EC and pH. During the autumn overturn, the values in the monimolimnion were noticeably more uniform than in the other seasons, reflecting the similarly uniform values in the mixolimnion.

Lake Sortavalanjärvi had an exceptional, slightly more oxic (up to 1.6 mg L^{-1}) layer at the deepest part of the lake in February 2018 (Fig. 4), which was not observed at other sampling depths. Concurrent with this unexpected increase in oxygen, a decrease in pH and an increase in ORP was observed. The origin of this more oxic water layer is unknown, but it may have resulted from an increased dense drainage from the mine area caused by increased inflow. This interpretation is supported by the data of the Finnish Meteorological Institute (2021), which indicates that, in December 2017, precipitation was 170–220% of the 1981–2010 monthly average at the nearby weather stations in Kuopio, Mikkeli, and Joensuu. The less reducing conditions in this bottom layer may have induced precipitation of Fe and Mn oxyhydroxides resulting in the observed decrease in pH and elevated turbidity (not shown) between 14.5 and 15 m.

Chemistry of the Water Columns

Most of the main cations and anions had higher concentrations in Lake Sortavalanjärvi (Fig. 6) and the main anion and cation concentrations also increased more notably with depth than in Lake Valkeinen (Fig. 5). The main anions and cations of the mine water samples typically reflect the composition of the tailings material, i.e. mineralogy of the mined material and remains of chemicals used in the mining processes (Alopaeus et al. 1986; Heikkinen et al. 2009; Räisänen 2015; Turunen 1960). The observed high concentrations of the main anions S and SO_4 originate from the weathering of the sulfide minerals in the mine wastes, and from the remains of chemicals, such as sulfuric acid, xanthates, and copper sulfate, used in ore processing. The main cations, Ca and Mg, originate from the weathering of the gangue minerals, which are mostly pyroxenes and amphiboles (Grundström 1985; Papunen and Koskinen 1985), as well as from the lime used in ore processing and in water treatment as a buffering agent. K and Na concentrations, originating from gangue minerals and xanthates, such as Na-IBX and potassium amyl xanthate, were also rather high in the studied lakes. However, both mines in question ceased operations years ago and the effect of processing chemicals has likely decreased significantly.

Fe concentrations were higher below the chemocline in both lakes. This is to be expected as Fe bound to insoluble ferric hydroxides ($Fe(OH)_3$) is reduced to soluble forms in the hypoxic monimolimnion (Davison 1993). Overall, the Fe concentrations were higher in Lake Sortavalanjärvi, possibly due to the humus-rich inflow from Muhelampi pond. Mn concentrations were elevated below the chemocline in Lake Valkeinen, whereas in Lake Sortavalanjärvi, the highest concentrations of Mn occurred at the chemocline or just above it. The maximum concentration of Mn occurring at a shallower depth than Fe can be attributed to the higher standard potential of the Mn redox reaction and the slower oxidation kinetics of dissolved manganous ions compared to dissolved ferrous ions (Davison 1993). This phenomenon has been observed in several seasonally anoxic and meromictic lakes, e.g. in Esthwaite Water (Davison 1993; Hamilton-Taylor et al. 2005) and Lake Sammamish (Balistrieri et al. 1992). The mechanism likely returns some redox sensitive metals from the lower part of the mixolimnion back to the monimolimnion that would otherwise be lost due to diffusion and mixing.

The Fe and SO_4 in lakes Valkeinen and Sortavalanjärvi likely play significant roles in the P retention capacity of the lake sediments. In boreal freshwater lakes, SO_4 concentrations are typically low, and P is coprecipitated with Fe oxides and oxyhydroxides. If SO_4 concentrations increase sufficiently and conditions are reducing, Fe reacts with the sulfides formed from SO_4 reduction to form insoluble Fe sulfides, which may lead to Fe exhaustion and consequently increased P release from sediments (e.g. Caraco et al. 1989, 1993; Smolders and Roelofs 1993). Based on the very high P concentrations in Lake Valkeinen, particularly in the monimolimnion, the lake would be considered eutrophic or even hypereutrophic (Wetzel 2001) despite its small, forested watershed. The high amount of P in the water mass of Lake Valkeinen is likely due to the use of phosphates as reagents in mineral processing, combined with slow removal through the outlet and into sediments. The Fe entering Lake Valkeinen may be reacting with sulfides instead of coprecipitating with P, which may have led to increased P release, low retention in sediments, and consequently the eutrophication of Lake Valkeinen (Kleeberg and Grüneberg 2005). In contrast, the P concentrations in Lake Sortavalanjärvi were mostly below detection limit (20 μg L^{-1}) apart from one measurement from the monimolimnion at 28.7 μg L^{-1}. The Fe-rich inflow from Muhelampi pond may keep the P concentrations in Lake Sortavalanjärvi relatively low but detectable.

Trace metals such as Ni, Cu, and Zn were elevated above the chemocline and generally decreased to a few μg l^{-1} in the monimolimnion of both lakes. Overall, these decreases likely stem from the change to reducing conditions at and below the chemocline, which may lead to the removal of these metals from the water column through sulfide mineral precipitation (e.g. Denimal et al. 2005; Martin et al. 2003; Moncur et al. 2006). A small Ni peak in the bottommost sample of Lake Sortavalanjärvi in February 2018 probably originates from the exceptional event of more oxygenated conditions at the bottom.

The concentrations of the main cations and anions in the outflow were ≈ 18%–45% of the concentrations in the inflow from the mine at Lake Valkeinen (Oct. 2017). This suggests that most of the loading from the mine area is detained in the lake basin. In contrast, the concentrations of the main cations and anions in the outflow from Lake Sortavalanjärvi were ≈ 67%–90% of the concentrations in the inflow from the mine (Nov. 2017). This implies that most of the loading from the mine area is flushed through Lake Sortavalanjärvi.

The Stability of the Meromixis

Irregular circulation, particularly in spring, is a common natural feature in boreal inland lakes that have high humus contents and are ice covered for several months each winter (e.g. Hakala 2004; Hongve 2002; Simola and Arvola 2005). The isolating effect of ice cover leads to oxygen depletion, which can result in anoxia particularly in small, shallow lakes with limited inflow (Kirillin et al. 2012). If the duration of autumn circulation is also very short, due to e.g. lake morphometry, catchment characteristics, and climate, a persistent solute gradient may form in the lake, leading to meromixis. A shift from holomixis to meromixis can also be affected by additional anthropogenic factors. In Finland, these shifts have historically been associated with the onset of agriculture and subsequent eutrophication due to increased influx of minerogenic electrolytes and organic load (Hakala 2004). According to Hakala (2004), these human-impacted meromictic lakes typically have high alkalinity and pH values, and increased concentrations of basic elements, metals, and nutrients. Lakes Valkeinen and Sortavalanjärvi are both small but relatively deep, sheltered by forests and inclines, and elongate in shape, which restricts the mixing effect of wind to directions parallel to the long side of the lakes. Furthermore, the meromixis in both seems to be mostly due to superficial loading from the mines in the catchment. Sporadic measurements done on both lakes by environmental authorities and the Geological Survey of Finland indicate that the lakes already had bottom water anoxia in the 1990s and may have shifted to meromixis by 2008 (Kauppila et al. 2017).

The MyLake model scenarios suggest that the external load from the mines would continue affecting the lakes for several decades even if the lakes no longer received any inflow from the mine area (Figs. 7 and 9). This is perhaps not surprising, since in meromictic lakes, the monimolimnion is isolated from the atmosphere and accumulates dissolved substances very effectively while transport upwards through density gradients is slow (Boehrer et al. 2017). Precipitation reactions at the chemocline may also return dissolved substances to the monimolimnion.

Woolway and Merchant (2019) modelled the effect climate change could have on lake mixing regimes worldwide. Their findings suggest that many lakes could mix less frequently by the end of the twenty-first century, particularly those that already display anomalous mixing behaviour, meaning that many currently dimictic lakes would turn monomictic due to changing winter ice conditions. However, our model scenarios with IPCC AR5 projections increase the circulation in both lakes (Figs. 8 and 10), with the more severe RCP 8.5 projection increasing the circulation in Lake Valkeinen more than the "50% mine load-scenario" but less than the "no mine load-scenario." The lakes studied by Woolway and Merchant (2019) were quite large (> 27 km^2) compared to lakes Valkeinen and Sortavalanjärvi, which presumably have different ice phenology due to their smaller size and sheltered surroundings. Additionally, the dimictic lakes that Woolway and Merchant (2019) projected shifting to monomictic are situated in more temperate regions (40° N–60° N), where many lakes are projected to experience ice-free winters in the future (Sharma et al. 2021).

According to the MyLake model scenarios, the lack of sufficient wind mixing seems to be a strong factor in sustaining the meromixis in lakes Valkeinen and Sortavalanjärvi. Increasing the wind speeds by only 2 m s^{-1} resulted in the waters circulating during spring and autumn, with the main mixing event taking place during autumn. Woolway and Merchant (2019) had a very small number of the shallowest lakes studied shifting from dimictic to polymictic, which they speculate to be due to some change in the meteorological drivers acting at the lake surface, e.g. surface wind speed. Bartosiewicz et al. (2021) also noted the importance of wind shear, or lack thereof, for better predictions of lake ice phenology in the future.

Additional anthropogenic nonpoint stressors that might influence lakes Valkeinen and Sortavalanjärvi increasingly in the future include so-called water browning due to increasing DOC concentrations (e.g. De Wit et al. 2016; Monteith et al. 2007) and freshwater salinization syndrome (e.g., Cunillera-Montcusí et al. 2022; Kaushal et al. 2018;

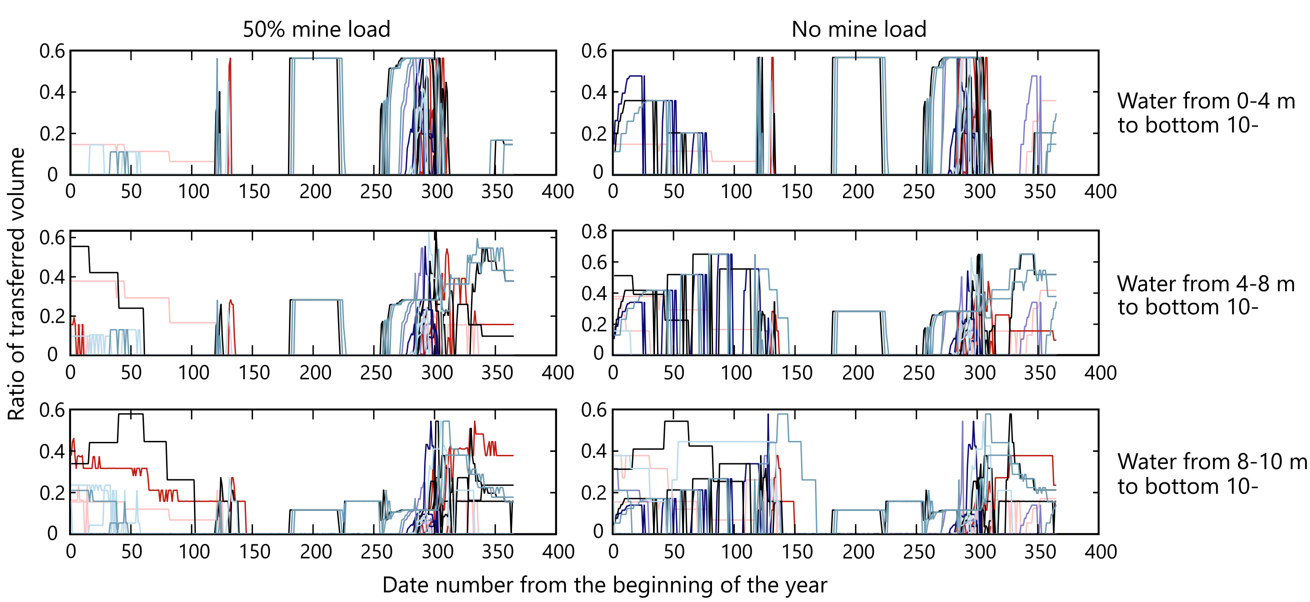


Fig. 10 Modelled ratio of transferred water volume between different water depth layers at Lake Sortavalanjärvi in the "50% mine load"-scenario (on the left) and "no mine load"-scenario (on the right). Different colours represent different plots, with each plot being one year. Plotting is annual from AD 2010 to AD 2020, after which there is a 10-year gap between each plot until AD 2129

Sibert et al. 2015; Tammelin and Kauppila 2018). These stressors may interact synergistically with climate change and lead to stronger stratification (Bartosiewicz et al. 2019, 2021). As such, it becomes even more important to limit point source pollution, such as mines, to lessen the anthropogenic loading into these lakes and facilitate their return to holomictic conditions. The effects of mining, in Finland and globally, can only be expected to increase as the demand for minerals increases and technological advances make mining of lower grade ores feasible (Mudd 2007). Mining lower grade ore results in larger individual mines and more waste rock, which increases the amount of mine waters, particularly when low-grade sulfidic ore deposits are mined.

However, the sudden full circulation in these lakes might not be desirable either. Since the monimolimnion is enriched in several substances, some in ecologically hazardous concentrations, a sudden overturning of the whole water column would almost certainly lead to deteriorating lake water quality, with severe impacts on the lake ecosystem, at least temporarily (Heikkinen and Väisänen 2007; Lehmann et al. 2015). Therefore, it would be better to ensure that lakes do not turn meromictic in the first place by e.g. setting stricter environmental load permits for SO_4 from mine areas.

Conclusions

We investigated two natural lakes situated near closed Ni-Cu mines to verify their shift from holomixis to meromixis. The lakes were found to be permanently stratified with a chemocline separating the circulating upper water from a more stagnant, anoxic bottom water. The main reason for this denser layer of bottom water is the SO_4-rich water accompanied by an abundance of cations such as Ca, Mg, and K flowing from the nearby mined areas. This load is relatively small but enough to sustain the meromixis in these small headwaters. In addition, the morphology and sheltered surroundings of these lakes may help sustain the meromixis.

If the load from the mined areas were to decrease or stop completely, the studied lakes would revert to holomictic within decades, according to modelling. Wind conditions also seem to be decisive for the water circulation, particularly in spring, as increasing wind speeds led to more circulation. Furthermore, climate change may play a role, but its effects are not as clear as it does not necessarily influence the ice conditions in small boreal lakes as much as in temperate lakes. The possible recovery of these meromictic lakes to holomictic conditions needs to happen slowly, since a sudden, complete overturn in the water column could be very detrimental to the biota of the lakes. As per usual, prevention is of key importance.

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Data availability The raw data supporting the conclusions of this manuscript will be made available by the corresponding author on reasonable request. The MyLake model code is available online under the CC BY-NC-SA 3.0 NO license.

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