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# HYDROTECHNOGENICAL INFLUENCE OF THE OIL SHALE MINES TO THE WATER QUALITY OF THE NATURAL LAKES IN THE KURTNA LAKE DISTRICT, ESTONIA

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#### Abstract

Lakes' response to the chances in water chemistry were studied with the aim of acquiring new knowledge about protection of the anthropogenically influenced lakes. To evaluate the impact of oil shale mine water on the lake ecosystem status we compared macrophyte compositions of pre-industrial period to nowadays. For a more detailed picture of changes, we used also paleolimnological approach and in sediment cores from two lakes, we analysed sedimentary pigments. Kurtna Lake District is located in the northeastern Estonia, mostly in the territory of the Kurtna Landscape Conservation Area. Kurtna is the lake-richest region in Estonia: there are around 40 lakes per 30 sq. km, from which 18 belongs to the Natura 2000 network of the EU. Same time the Kurtna region is an environmental conflict area. In the beginning of the 1950s, the rapid expansion of oil shale mining and related industry started in the vicinity of Kurtna, raising the need for technological water. For this, the channel system was constructed. In order to increase the capacity for discharge waters pumped out from oil shale mines, the channel was further reconstructed in 1970. The waters from Estonia and Viru oil shale mines, with a distinctively different composition if compared with the natural surface waters, were directed through lakes Nõmmejärv, Särgjärv, Ahvenjärv, Peen-Kirjakjärv and Kirjakjärv. Inflowing mines water has increased the input of suspended matter (up to 40 t/yr). Another problems are the high mineralisation (due to SO<sub>4</sub><sup>2-</sup>, HCO<sup>3-</sup>, Ca<sup>+</sup>, Mg<sup>+</sup>) and different water pH. In 1937, when the lakes were in natural condition, the sulphates level in the lakes were in the range of 1.0-6.7 mg/l, while in the 1980s, the corresponding values were between 200 and 300 mg/l, and nowadays in some cases, it exceeds over 300 mg/l. Our results indicate an increase in the overall coverage and biomass of macrophytes. Major changes have taken place in Lake Kirjakjärv, where the number of species have diminished two times (from 30 to 15 compared to 1954). In Lake Nommejärv, the sedimentary pigments indicated a higher production before mine water influence, apparently because of the increasing human impact in catchment. In this case, the mine water has mitigated the effects of eutrophication. So the diversity has declined, but lake ecosystems are still in equilibrium state. The high flow-through rate and fast water circulation (up to 40 times per year in case of Lake Nõmmejärv) prevents the formation of anoxic conditions and the subsequent boost in primary production.

Keywords: water quality, mine water, human influence, lake ecosystem, sulphates, natural lakes, macrophytes

### **1 INTRODUCTION**

The chemical composition of lake water, determined by a combination of physical, chemical, and biological processes in the lakes and their catchments, can have considerable implications for biological diversity, ecological functioning and productivity of the lake. The long-term hydrotechnogenical influences have significant impact on the water cycle and its coupling with the energy flow and the sediment transport (Donohue & Molinos, 2009). As the total impact of different processes on ecosystem will have complex consequences on the lake water regime, chemistry and sedimentation, the determination of the acting mechanism and relations between them is extremely complicated. The quantification of interactions among multiple stressors comprises one of the most pressing problems in hydroecology. If not influenced by the direct pollution sources in the catchment, Water chemistry of small lakes can be expected to reflect regional characteristics of water chemistry, as well as of global anthropogenic processes such as climate and longrange air pollution (Müller et al., 1998; Moiseenko et al., 2001; Moiseenko, 2013). It has been shown (Yan et al., 2002) that the chemical composition of surface waters is closely connected with the rock type in the catchment and the mode of chemical weathering of rocks and soils, but environmental issues and extra load related to mining will boost the outcome. In case of Estonia, the chemical composition of mine water is somewhat specific – it is not acidic but alkaline, because local kukersite oil shale forms horizontal sequences in the limestone from which it is extracted in underground mines (Marzecovà et al., 2011). This requires drainage of large amounts of groundwater through artificial channels, which are in some cases connected with natural lakes. By discharging the mine drainage water through the lakes, a dilution effect is achieved at the expense of the lake ecosystems (Varvas, 1994). Problem is that although sediments are a natural and

important component of lake ecosystems, mine water is carrying mine clastic mineral debris, siliciclastic particles, and oil shale remains and is high in sulphate ions and calcium (Erg. 2003; Marzecovà et al., 2011). The elevated sulphate concentrations in mine water can have significant influence to the natural water bodies if environmental conditions are favourable, because microorganisms play an essential role in the sulphur cycle, catalysing both oxidation and reduction reactions of sulphur compounds (Sánchez-Andrea et al., 2014). The overall impacts of increased sediment loads on lake ecosystems is still not well studied, but indirect effects are likely also highly important, with increased sediment loads affecting both bottom-up and top-down ecological processes (Donohue & Molinos, 2009). In addition to changed water chemistry and extra nutrient load, the decreased transmission of light through the water column is among the most important of the physical effects of increased sediment loads on aquatic ecosystems. The increased mineral turbidity also influences the lake heat budgets by increasing the reflection of sunlight back to the atmosphere or through the absorption of heat by suspended particles. (Donohue & Molinos, 2009) Thermal regime of the lake is also affected by the mine water temperature (around 8°C when pumped out). The impact of temperature is season dependent; it is warmer than natural water during the winter and cooler during the summer. Thermal pollution from industry and beneficial and adverse effects of heated or cooled water has been recognized as a problem already for a long time (Davidson and Bradshaw, 1967). For example, the temperature alters the settling velocity of suspended sediment particles by modifying water viscosity and density (Kerr, 1995) but changes ecosystem also directly. The occurrence of macrophytes in lakes is affected by abiotic conditions (e.g. water quality, sediment properties and shore topography) together with physical stress (e.g. waves or ice, water currents, temperature conditions, water level fluctuations) and biotic factors (e.g. herbivory, competition, human influence) (Madson et al., 2001; Mäkela et al., 2004). However, the vegetation responses to environmental factors are not always linear - sometimes the highest macrophyte diversity has been observed in the mesotrophic or slightly eutrophic lakes (Madson et al., 2001). Some studies (Jeppesen et al., 2000) showed that while floating-leaved macrophytes tend to have a unimodal mesotrophic peak, the number of submerged plant species declined in eutrophic lakes and in dystrophic lakes, the species number is lower than in clear-water lakes within the same nutrient status (Mäkela et al., 2004). Traces of historical ecosystem changes and objective information about the development of lake can be obtained from the stratigraphic analysis of sediment cores, an approach that is widely used in landscape study (Last & Smol, 2001; Terasmaa et al 2013). Water chemistry in Kurtna Lake District has been previously studied at several occasions (Riikoja, 1940; Sagris, 1989; Varvas, 1994) as well as history of land-use and development of the lakes, and many scientific papers have been published (e.g. Ilomets, 1987; Ilomets 1989; Varvas and Punning, 1993; Koff, 1994; Punning, 1994; Punning et al., 2006; Marzecovà et al., 2011; Vainu & Terasmaa, 2014). This gives a good possibility to compare historical data with current findings. This work aims to acquire a new knowledge about the protection of the hydrotechnogenically influenced natural lakes. To evaluate the impact of oil shale mine water on the lake ecosystem status we compare water chemistry changes, sedimentary pigments, and macrophyte compositions of pre-industrial period to nowadays. The quantified variability and trends in water chemistry and magnitude of the ecosystem responses helps to predict effects of potential water regime changes in future.

## **2 STUDY AREA**

Kurtna Lake District is located in the northeastern Estonia in Alutaguse Lowland, mostly in the territory of the Kurtna Landscape Conservation Area (Fig. 1). Kurtna is the lake-richest region in Estonia: there are 38 natural lakes per 30 sq. km, from which 18 are protected under the EU's Natura 2000 network. The Kurtna Landscape Conservation Area is transitional zone between densely populated and heavily industrialised oil shale mining region and a sparsely inhabited territory with large forests and mires, which makes it predominantly a recreational area. The lakes of Kurtna have been deteriorated by the nearby peat fields, the mining of oil shale and the Pannjärve sand quarry. Also, the local ground water is pumped as drinking water to the towns of Ahtme and Jõhvi and the mine water is directed through several lakes. Profound human disturbances in studied lakes (L. Nõmmejärv, L. Särgjärv, L. Ahvenjärv, L. Peen-Kirjakjärv and L. Kirjakjärv) started in the first half of the 20<sup>th</sup> century. At the latest by 1920, the natural flow of Raudi Creek was modified, changing the lakes from the closed to flow-through type (Varvas, 1994).

The kame field around the lakes consists of kames ranging from 40 to 70 m a.s.l, separated by small depressions (Ilomets & Kont 1994). The glacial deposits in the area are 50-60 m thick; consist of medium- to coarse-grained glaciolacustrine and glaciofluvial sands (Erg, 1994). The climate of the region is continental. Monthly average air temperature is below 0°C from November to March (annual average air temperature is 4.7°C). Average annual precipitation is 684 mm (Vainu et al., 2014).

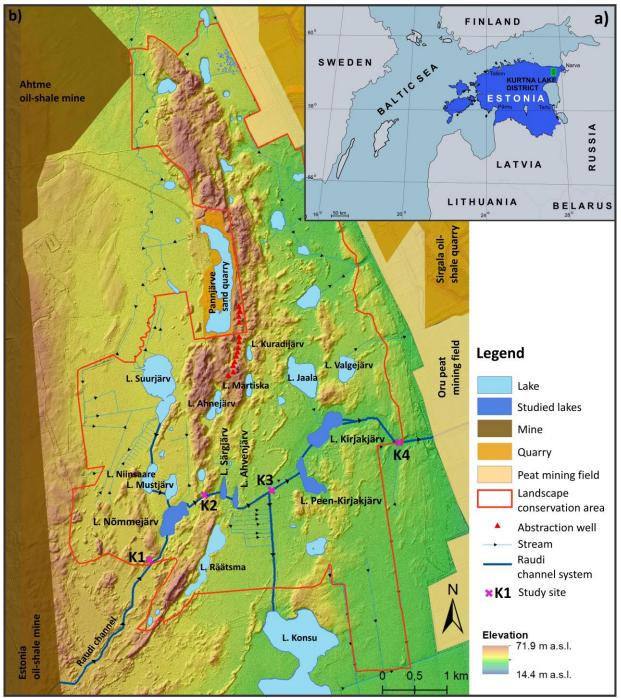


Figure 1. Location of the Kurtna Lake District (a) and studied lakes (b) with pressure factors.

### **3 MATERIAL & METHODS**

For comparison with available historical data, we carried out water chemistry survey of Raudi channel system in a variety of flow conditions during the ice-free period of years 2012-2013. Water chemistry was measured from four sites in Raudi channel system before, after and in between the lakes (K1-K4, Fig. 1). In some cases, water chemistry was also measured in lakes and directly after mines pumping sites outside of Kurtna Landscape Conservation Area.

*In situ* measurements were made with the YSI 556 multiparameter system. YSI probe measures temperature with an accuracy of  $\pm 0.15$  °C, conductivity with an accuracy of  $\pm 0.001$  mS cm<sup>-1</sup>, dissolved oxygen with an accuracy of  $\pm 0.2$  mg l<sup>-1</sup> and pH with an accuracy of  $\pm 0.2$  unit.

Water samples for laboratory analysis were collected in non-sorbing plastic bottles. In the field, the bottles were rinsed with the same water, and then filled to the top to ensure minimum analytical error from carbon dioxide degassing. Water samples were transported to the laboratory as soon as possible (within 2-3 days, normally next day). In the beginning of measurement period, we analysed up to 14 parameters, later

fewer. In this paper we use a selection of following ions:  $SO_4^{2-}$ ,  $Ca^{2+}$ ,  $Mg^{2+}$  and  $Cl^-$ . Analyses were made in Estonian Environmental Research Centre using the following methodologies: for  $SO_4^{2-}$  (mg l<sup>-1</sup>) turbidimetric method (AOAC 973.57); for  $Ca^{2+}$  (mg l<sup>-1</sup>) and  $Mg^{2+}$  (mg l<sup>-1</sup>) atomic absorption spectrometry (EVS-EN ISO 7980:2000); for  $Cl^-$  (mg l<sup>-1</sup>) silver nitrate titration with chromate indicator (ISO 9297).

For lakes' water and hydrochemistry budget, we carried out repeated discharge measurements (volume per unit of time) of the Raudi channel system. Measurements were done using a *Valeport flow meter model 801* in the four cross-sections (K1-K4) and standard methodology (ISO 748:2007). For measurement locations, cross-section with as uniform bottom as possible and strong clear bank were selected. The speed of the current was measured every 30 cm, flow-meter were held on the 60% of the distance from the bottom. Discharges were calculated as m<sup>3</sup> s<sup>-1</sup> for every cross-section in a given time.

To evaluate the influence of mine water on the lake ecosystem status, we compared macrophyte compositions of pre-industrial period to nowadays. In order to describe and map the modern vegetation in lakes, the whole littoral zone was traversed by boat and the species composition and depth limits of different plant groups were recorded. For submerged aquatic plants, which were not readily visible, five throws with a grapple were made around the boat. As the past vegetation records are mainly descriptive, therefore the comparison and analysis is more of qualitative than quantitative nature.

Sediment cores from L. Nõmmejärv and L. Kirjakjärv (86 and 103 cm lengths respectively) were analysed for loss on ignition (LOI) according to the standard methodology (Heiri et al., 2001, Boyle 2001). The sediment core from Nõmmejärv was used for <sup>137</sup>Cs- and <sup>210</sup>Pb-dating by gamma-spectroscopy. These analyses were conducted at the Ukraine Hydrometeorological Research Institute. The sediment core chronology was determined according to the model of Constant Rate of Supply (CRS) (Appleby & Oldfield, 1978). The sediment cores show a similar stratigraphic pattern with depth and good stratigraphical correlations of LOI in cores allowed transferred the <sup>210</sup>Pb dating result of core from L. Nõmmejärv to the L. Kirjakjärv. Sediment cores for pigment analysis were sectioned at 1 cm intervals in field, and placed into plastic boxes, flushed with argon and hold in dark and cold conditions within a collection day. The sediments were extracted overnight at -4°C in a 90% acetone. Chlorophyll derivatives (CD), a proxy for algal chlorophyll, were measured at 665 nm. Myxoxanthophyll (MYX) was measured using the separation method and calculations according Swain (1985). The chlorophyll values were expressed in relative units and pigments values were normalized to organic matter.

### 4 RESULTS & DISCUSSION

Although the water chemistry in Kurtna lakes has been previously studied (Riikoja, 1937; Sagris, 1989; Varvas, 1994), the measurement methods and sampling location has somewhat changed. The earliest published data from the end of the 1930s (Riikoja, 1940) show prevailing natural conditions ( $SO_4^{2-}$  concentration in the water of L. Nõmmejärv 5.8 mg l<sup>-1</sup>, L. Särgjärv 2.9 mg l<sup>-1</sup> and L. Kirjakjärv 4.8 mg l<sup>-1</sup>). Comparing data from end of the 1930s (Riikoja, 1940) with those from 1980s and the present, it is possible to evaluate the extent of the man-made changes in the lakes. Of course, we must keep in mind that the oldest data were produced by a single measurement made in the lakes in August 1937, while recent measurements were made from in- and outflows of the lakes and were collected over several years. The increased content of sulphate in the surface water indicates directly the influence of the mine water (Table 1, Fig 2).

	2010s. Locations of the sites are on Fig. 10.										
	SO₄²- (mg l⁻¹)		Cl <sup>-</sup> (mg l <sup>-1</sup> )		Ca²+ (mg l-¹)		Mg <sup>2+</sup> (mg l <sup>-1</sup> )				
	1986-87 (n=10)	2012-13 (n=9)	1986-87 (n=10)	2012-13 (n=2)	1986-87 (n=10)	2012-13 (n=2)	1986-87 (n=10)	2012-13 (n=2)			
<b>K</b> 1	250	286	13.8	5.1	84.4	125.0	50.6	47.5			
K2	255	240	12.9	5.0	66.8	100.9	45.7	56.8			
K3	223	227	12.4	5.2	64.4	91.7	44.9	53.0			
K4	126	194	6.9	5.2	34.8	95.8	24.1	41.1			

**Table 1.** Comparison of average values of selected anions and cations from the 1980s (Sagris, 1989) and2010s. Locations of the sites are on Fig. 1b.

In Estonia, the first oil shale power plants were built in the late 1930s, but in the vicinity of Kurtna rapid expansion of oil shale mining and related industry started in the beginning of the 1950s. According to the sparse hydrochemical and -biological data, the lakes retained the stable conditions at least until the 1950s, thus the values reflect the natural conditions of the lakes (Mäemets, 1968). Certainly, the atmospherically emitted alkaline fly-ash, which is characterized by the elevated pH values (up to 12) and high contents of several chemical substances (Varvas, 1994) also caused some changes in the lakes (Mäemets, 1987; Varvas & Punning, 1993), yet the rising need for the technological water fact by the developing industry was more important stressor

Therefore, a water supply system was built from the Lake Konsu. In the 1970s, the channel system was reconstructed in order to increase its capacity for the discharge waters, which were pumped out from the oil shale mines (Marzecovà et al., 2011). The waters from Estonia and Viru oil shale mines, having a distinctively different composition if compared to the natural surface waters, were directed through lakes Nõmmejärv, Särgjärv, Ahvenjärv, Peen-Kirjakjärv and Kirjakjärv (Fig 1b). These measures triggered visible changes in the lakes (Mäemets, 1987). As Fig. 2 shows, in sites K2 and K3, the sulphate concentrations in the water have remained at the same level, when compared between the decades. In the inflow (K1) and outflow (K2), the concentrations have risen. If we compare data within the different time periods, we can see decline in the flow direction, however nowadays, the sulphate concentrations in the outflow from Kurtna Lake Distric (K4) are much higher. This phenomenon could be explained by the changes in water budget; however, there may be other alternative causes. It is possible that lakes have reached the maximum level of sulphate sequestration from water and now increasingly higher amount flows first through the lakes and then to the Baltic Sea.

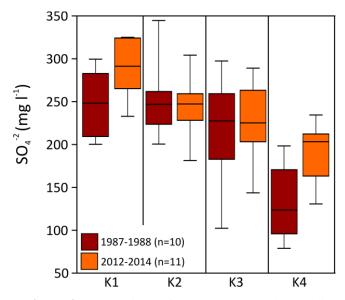


Figure 2. Comparison of sulphate content in Raudi channel (locations of the sites are on Fig. 1b). Boxplots represents quartiles and average values with min and max.

**Table 2.** Total average amount of the sulphates (t) carried through the studied cross-sections per day.

SO4-2 t d-1	K1	K2	<b>K</b> 3	K4
1986-1987	7.1	6.9	5.6	0.7
2012-2013	12.5	16.0	9.5	6.6

To some extent, the rise of the sulphate content also reflects the atmospheric source, but concentrations up to several hundred mg per litre have been only found in the pumped-out mine waters, which makes it good indicator to analyse. The source of this compound is the ordovician bedrock, from which it was released through the oxidation of pyrite. Sulphates are known as a potential threat to the water environments, because under the anoxic and low pH conditions, the sulphate-rich sediments can release the toxic  $H_2S$ , leading to the phosphorus resuspension from sediment to lake water (Perens et al 2006). The high flow-through rate and fast water

circulation (up to 40 times per year in case of L. Nõmmejärv) have prevented the formation of anoxic conditions in the lake bottom. Since the Viru mine closedown (summer 2013), the water circulation is much lower (~17 times per year). Critical limit for the water circulation in L. Nõmmejärv is hypothesized to be less than 7 times per year. This limit can be reached in case when the mine waters will be completely redirected or the artificial channel system will be closed. Another factor, which has been contributing to the mitigation of the sulphates problem is the pH of the mine water. In years 2012-2014, the pH in the site K1 has been in average 7.98, but the value has been declining in the end of the period (in average 7.18 in 2014). This can be a result of the smaller proportion of the alkaline mine water in the whole water budget due to Viru mine closedown.

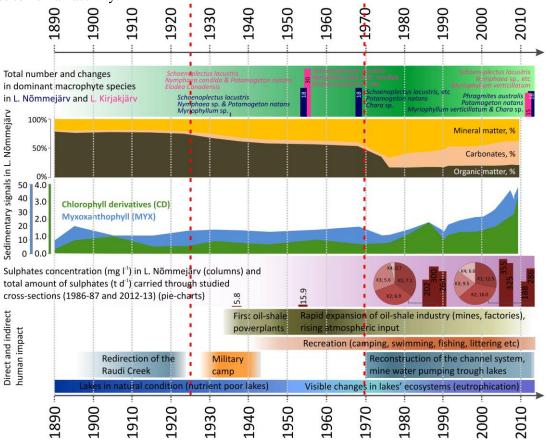
When comparing macrophyte compositions of pre-industrial period to nowadays, the main difference is the increasing overall coverage and biomass of macrophytes. While in L. Nõmmejärv and L. Särgjärv the number and composition of species has remained relatively stable, major changes have taken place in Lake Kirjakjärv. The number of species has decreased to 15 which is a drastic change when compared to 30 species found in 1954. Also the once dominant *Elodea Canadensis* Michx., *Potamogeton* 

lucens L. and Utricularia vulgaris L. are either gone or scarce and have been replaced mainly by Myriophyllum verticillatum L.

The sedimentary record in L. Nõmmejärv show minor variations in the pigment values, starting at the beginning of the 20th century, while in L. Kirjakjärv only the cyanobacterial pigment MYX showed a slight increase. Sedimentary pigments, produced by plants and algae, can provide important information about changes in phytoplankton community and productivity, and therefore they are often used as markers to assess the lake ecosystem status (Leavitt & Hodgson, 2001). In both lakes, mineral matter content increased at the same time, suggesting that the changes in pigment record were caused by the alteration of Raudi Creek. The considerable rise of sedimentary pigments occurs in 1970s. In the L. Nõmmejärv, the pigment values are still very unstable, increasing and declining from time to time. By contrast, in the L. Kirjakjärv has steady increase of pigment values and cyanobacterial myxoxanthophyll abundance is three times higher then in L. Nõmmejärv during the time period. Period of stable increase of MYX, CD and TC in L. Nõmmejärv marks the time after the oil shale mining where reduced (1990s). This confirm the tendency to a stronger biological activity in L. Nõmmejärv, apparently because of increasing human impact in catchment, which was temporarily mitigated or diminished by inflow of mine waters. However, in the L. Kirjakjärv the dilution effect of mine drainage water is not noticeable and lake ecosystem status has deteriorated since 1970s when the Raudi channel system was renovated.

#### **5 CONCLUSIONS**

Main findings of this paper are presented in the graphical summary (Fig 3). Different natural conditions (topography, catchment area, depth and volume, water retention time, etc.) and different pollution loads on the lakes and their parts have resulted in different resistances of their ecosystems and different responses to human activity.



**Figure 3.** Graphical summary of the main findings. From bottom to top: changes in direct and indirect human impact; concentration and total amount of sulphates; sedimentary signals in L. Nõmmejärv; changes in macrophytes in L. Nõmmejärv and L. Kirjakjärv. Red dotted lines denotes two main switches in the human impact. Human impact summary is compiled from the Mikomägi et al. (2012); precipitation data is from the Estonian Environmental Agency.

Discharge of mine drainage waters into the lakes has changed the lake water hydrochemical characteristics and affected lakes ecosystems. During the last decades, the studied lakes shifted toward more eutrophic state and there are notable changes in sediment and macrophytes composition. Nevertheless, the lakes are still in equilibrium state, most probably because of high flow rate and fast water circulation (up to 40 times per year in case of L. Nõmmejärv), which prevents the anoxic conditions. If those conditions would change (closing down the mines, redirection of the mine water, etc.), the ecological state of lakes can be shifted dramatically. For management and protection of Natura 2000 network, lakes the long-term stability of the lake ecosystems has to be the main goal.

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