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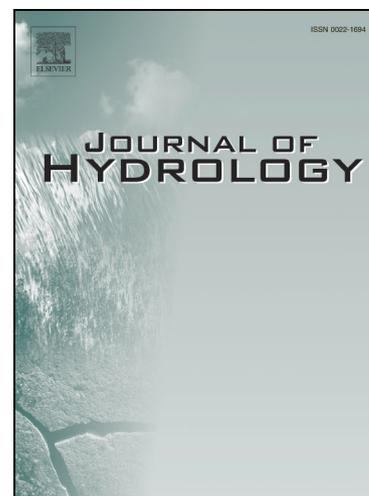
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Impacts of gold mine effluent on water quality in a pristine sub-Arctic river

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Abstract

Impacts of mining on water quality are a great concern in the Arctic region. This study evaluated the impact of pre-treated mine effluent on river water quality. The study was conducted along the Seurujoki River in sub-Arctic Finland, which is impacted by Kittilä gold mine. The study analyzed water quality and hydrological data upstream and downstream of the mining area over an eight-year period, including a tailing dam leakage event in 2015. The analysis focused on water quality determinants such as electrical conductivity (EC), sulfate, antimony, manganese, and total nitrogen (N_{total}). Descriptive statistics on river water at four stations along the river corridor showed negative impacts of mining activities on the recipient water body. In order to find an indicator for water quality, correlation analysis between the water quality determinants was carried out. It identified EC as a good indicator for continuous water quality monitoring, especially to detect mining accidents such as partial failure of a tailings dam. The results showed increasing contaminant concentrations due to mining as more mine effluent was generated over time. A linear mixed model was developed to predict the coefficient of different elements affecting EC at river water monitoring stations impacted by mining effluents. The results provide new information on how to assess mining water impacts and plan future water quality monitoring.

Keywords: Mining, environmental impacts, contamination, accident, Finland.

List of abbreviations

Q_{river} : Discharge of river

TP-A: Treatment peatland A

TP-B: Treatment peatland B

EC: Electrical conductivity

$I_{\text{TP-A}}$: Inflow water to treatment peatland A (Pre-treated process water)

$Q_{\text{TP-A}}$: Outflow from treatment peatland A

$EC_{\text{TP-A}}$: Electrical conductivity of water sample from outlet of treatment peatland A

$I_{\text{TP-B}}$: Inflow to treatment peatland B (Pre-treated drainage water)

$Q_{\text{TP-B}}$: Outflow from treatment peatland B

$EC_{\text{TP-B}}$: Electrical conductivity of water sample from outlet of treatment peatland B

EC_{Station} : Electrical conductivity of water at station #

SO_4^{2-} : Sulfate

Sb: Antimony

N_{total} : Total nitrogen, i.e., sum of nitrate (NO_3), nitrite (NO_2), organic nitrogen, and ammonia (NH_3).

Cl^- : Chloride

Mn: Manganese

Fe: Iron

Mg: Magnesium

Na: Sodium

Ca: Calcium

K: Potassium

O_2 : Dissolved oxygen

NH_4^+ : Ammonium

NO_3^- : Nitrate

As: Arsenic

Ni: Nickel

COD: Chemical Oxygen Demand

pH: Log_{10} hydrogen ion concentration in moles per litre

1. Introduction

Active and closed mines are recognized as serious and long-lasting threats for river systems all over the world (e.g., Beane et al., 2016; Garbarino et al., 2018; Hudson-Edwards et al., 1999; Monna et al., 2000). Effluent from mining enrichment processes is often treated with advanced purification methods, but ‘secondary’ effluent with lower contaminant concentrations, from mine drainage water, runoff from waste rock piles, and leaching from tailing dams, is often discharged directly into river systems or only partly treated in sedimentation ponds or different type of wetlands (Monna et al., 2000). In addition to mine-influenced waters during the normal operation phase, tailings dam and operational failures and accidents are a serious threat to river systems (World Information Service on Energy, 2019).

Mining effluents are typically acidic and saline, with high concentrations of sulfate (SO_4^{2-}), iron (Fe), and other metals/metalloids (e.g., Olías et al., 2004). These waters pose a risk to terrestrial and aquatic ecosystems (e.g., Graupner et al., 2014) by reducing both the species and functional richness of communities (Berger et al., 2018). Recent developments in mining technology (Luoto et al., 2019; Watling, 2014) and the growing need for raw materials have increased the pressure to open new mines in sensitive Arctic regions (such as Finland), where some of Europe’s largest metal mines already exist (Boyd et al., 2016). In northern Finland, mining is in conflict with other ecosystem services such as tourism, fisheries, and traditional reindeer herding. All these have raised great concerns about the safety and sustainability of the mining industry in general (Räsänen and Lindman, 2018). The best available techniques for lowering environmental impacts of the extractive industry have recently been updated by the European Commission (Garbarino et al., 2018). Despite the fact that mine effluent waters are typically treated and purified using different active (da Silveira et al., 2009; Mackie and Walsh, 2012) and passive (Palmer et al., 2015; Sheoran and Sheoran, 2006) treatment methods, the environmental impact of these pre-treated mine waters can be considerable, especially in pristine Arctic and sub-Arctic waters (Khan et al., 2020; Larkins et al., 2018; Lemly, 1994).

Past studies on the impact of mining on river water quality have focused on monitoring tailings dam failure effects (Yu et al., 2011), remediation of contaminated river systems (Byrne et al., 2018; Carmo et al., 2017; Klebercz et al., 2012; Mayes et al., 2011; Olías et al., 2012), assessment of the economic impacts of mining accidents (Kossoff et al., 2014; Rico et al., 2008), and pollution risk analysis and risk management (Burritt and Christ, 2018; Komnitsas et al., 1998; Xenidis et al., 2003). A major risk linked to mining activities is re-mobilization of pollutants from contaminated riverine sediments (Galán et al., 2003; Meck et al., 2006), which is intensified by high salinity (Olías et al., 2004; Riba et al., 2003). In particular, the ‘first flush’ after a relatively dry summer, or initial surface runoff of a

rainstorm, can deteriorate river water quality, an impact associated with re-dissolution of sulfate precipitated during the summer following intense natural weathering of sulfate minerals (Oliás et al., 2004). During winters, a slight increase in pH and decrease in pollutants have been observed, effects which have been associated with dilution (Oliás et al., 2004).

The aim of this study was to gain a better understanding of the impacts of pre-treated mining effluent waters on a sub-Arctic river system that has not been well studied (Tolvanen et al., 2019). In sub-Arctic climate conditions, rivers have their unique characteristics and flow regime varies both between and within years. The most important cause of this variation in snow cover properties such as maximum snow water equivalent and time of melting in May-June. There is a need for more research on the true impacts of mining in this type of sensitive system. Using an intensive monitoring set-up with continuous logger sensors, specific objectives of the study were to (i) assess the temporal and seasonal variation in water quality due to mine water discharge in a sub-Arctic climate and ii) assess impacts of pre-treated mining water on river water quality determinants in different stations along the river corridor. In contrast to previous studies on mining water impacts on Finnish lakes (e.g., Leppänen et al., 2019, 2017; Niinioja et al., 2003) and rivers (e.g., Salmelin et al., 2017), this study examined riverine impacts using data of high temporal resolution, which added knowledge on seasonal variation. Another unique feature of the study is that it provides data on river responses during a mining accident. The novel information obtained on the mining site water environment, and how it should be monitored and conceptualized, can be useful for future studies and numerical modeling.

2. Materials and methods

2.1. Study area

The river Seurujoki, with a total length of 37 km, is located in the north part of boreal zone in the municipality of Kittilä (67°55'N, 25°20'E) in northern (sub-Arctic) Finland (Fig. 1a). Its catchment (307 km²) is sparsely populated and is part of the Kemijoki catchment (51,127 km²). More than 83% of the Seurujoki catchment area is classified as forest and peatlands, while agricultural land accounts only for 0.12% of the catchment area (Fig. 1b). The river has been classified as a peatland-dominated, medium-sized river with high humic content (Pöyry, 2016). Mining area (Kittilä gold mine) accounts for 2.5% of the catchment area (857 ha). The area is one of the largest epigenetic gold deposits in Central Lapland Greenstone Belt and the mine is one of the largest active gold mines in Europe (Wyche et al., 2015). The mine includes two open pits, underground workings, ore processing and

water treatment facilities, two settling ponds, waste rock dumps, other mine facilities, and several treatment peatlands to treat different types of effluent generated during mining operations. Mining started in 2008 with open pit mining, which ceased in 2012, and has continued as underground mining since October 2010. Given the current ore deposits and production volume, mining is expected to continue until 2036. The lifespan of the mine may be prolonged after that date, depending on the results of ore prospecting (Agnico Eagle Finland, 2015).

The Seurujoki river receives treated effluents from the gold mine, but also loads from scattered settlements and runoff from agricultural fields (Fig. 1d). Before discharging to the river, excess mining process waters are first treated in a gypsum precipitation unit (since 2017), and then polished in a treatment peatland (TP-A, around 44 ha). The drainage water from the mine area is purified in another treatment peatland (TP-B, around 17 ha). The mine drains the surrounding landscape, including groundwater, to the underground mine pit. All this drainage water is pumped to TP-B, where it flows eventually to the river system. Due to this, the general direction of groundwater flow is towards the mine pit (Fig. 1d). The groundwater outside the mining area follows natural flow paths and discharge to the river channel (Eurofins, 2019). Generally, the soil in the area and below the peat is predominantly glacial till with low hydraulic conductivity and there are no significant alluvial aquifers in the mine area or close by (AVI, 2013), suggesting little contact with the deeper groundwater. Both TPs have worked quite efficiently as buffer zones between the mine and the river (Khan et al., 2019; Palmer et al., 2015), meaning that all water quality criteria for mine effluent waters must be met already in the inflow waters to the TPs. Mean inflow to TP-A in the period 2010-2018 was 3100 m³day⁻¹, whereas it was somewhat higher to TP-B (around 7000 m³day⁻¹). More detailed descriptions of the mine (Larkins et al., 2018) and the TPs with their removal efficiencies (Khan et al., 2019, 2020; Kujala et al., 2019; Palmer et al., 2015) can be found in previous studies.

In the study period (2010-2018), mean annual temperature was 0.6°C and mean annual precipitation was 515 mm (Finnish Meteorological Institute, 2018). Typical permanent snow cover lasts from October to May and mean maximum snow depth is 80 cm (normal period 1981-2010) (Finnish Meteorological Institute, 2017). The region is classified as Dfc (snow climate characterized by moist, cold winters) in the Köppen climate classification system (Chen and Chen, 2013).

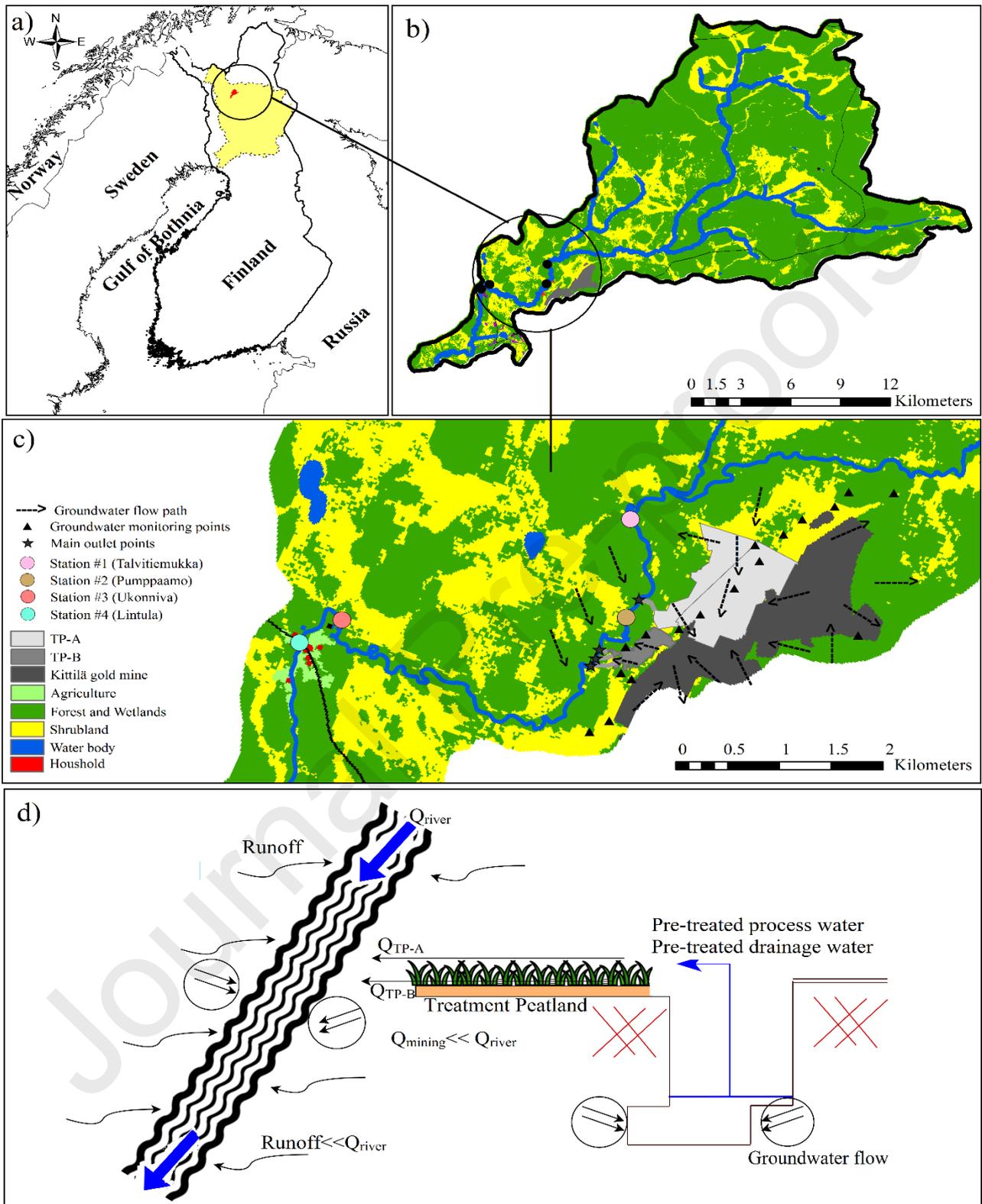


Fig. 1: Maps showing (a) the location of the Seurujoki river in northern Finland. (b, c) Land uses in the catchment; water quality monitoring stations used in this study are represented by circles and two treatment peatlands polishing mine-influenced waters (TP-B and TP-A) are shown, with the main outlet points and groundwater flow paths. d) Conceptual model of mine and river section water balance, indicating mine effluent inflow to the Seurujoki River.

2.2. Data on water quality in the Seurujoki River

Water quality data for the years 2010 to 2018 were obtained from four monitoring stations along the Seurujoki river (Fig. 1c). These were: Station #1, before the mining area (Talvitiemukka); Station #2, 0.3 km after the main discharge ditch from TP-A (Pumppaamo); Station #3, about 6.4 km after TP-A (Ukonniva); and Station #4, about 7.2 km after TP-A (Lintula). The distance between TP-A and TP-B is around 1.3 km.

In the present study, we monitored electrical conductivity (EC), sulfate (SO_4^{2-}), antimony (Sb), total nitrogen (N_{total}), chloride (Cl^-), manganese (Mn), iron (Fe), magnesium (Mg), sodium (Na), calcium (Ca), potassium (K), dissolved oxygen (O_2), chemical oxygen demand (COD), ammonium (NH_4^+), nitrate (NO_3^-), arsenic (As), nickel (Ni), and pH in the river. Based on the most recent environmental impact assessment report (from 2016), the most significant impacts on river water quality are increases in the concentrations of different nitrogen compounds, SO_4^{2-} , Sb, Fe, and Mn (Pöyry, 2016). Therefore these elements were selected for analysis in this study. We included Cl^- , K, Na, Mg, Ca, As, and Ni as local environmental authorities lists these as major concerns (AVI, 2013). We included EC, O_2 , COD, and pH as general indicators of water quality variations. Gold mine effluents typically also contain cadmium (Cd), lead (Pb), and mercury (Hg), but these have been determined or predicted to be negligible in the Seurujoki river (AVI, 2013) and were not included in the present analysis. All water quality data were downloaded from the open database HERTTA provided by Finnish Environmental Institute (HERTTA, 2018). Water samples were collected at different frequencies (2 or 3 times per month) over the study period and generally well represented the seasonal variability. Additionally, over the period July-November 2015, EC was measured at 60-min intervals at four river stations, in a joint effort by the Geological Survey of Finland (GTK) and the Water, Energy and Environmental Engineering (WE3) research unit at the University of Oulu. Cross-sectional measurements using multi-frequency and multi-constellation Trimble R10 GNSS receivers were made at the stations, in order to comprehensively chart the situation in the river and its discharge.

Since 2007, the Finnish Environmental Institute has been continuously measuring river discharge, based on the common rating curve method (Sauer, 2002), at Station #1, which is located 1.5 km above the mine water discharge point in the river. Based on the data obtained, the mean flow rate is $3.8 \text{ m}^3\text{s}^{-1}$ and maximum discharge typically occurs in May, during the snowmelt period.

Data on water quality at the inlet and outlet and volume of water inflow to TP-A and TP-B were provided by the mining company (as part of a monitoring program required by the environmental permit for the mine). All water samples were analyzed at accredited laboratories (AHMA Ympäristö Oy., Ramboll Analytics Oy., EUROFINS environment testing Finland Oy., and Lounais-Soumen

vesi- ja ympäristötutkimus Oy.), using standard methods specified by the Finnish Standards Association (SFS) and certified by the Finnish Accreditation Service (FINAS) (Fig. 2).

Some groundwater monitoring wells are located around mining area and have been sampled a few times per year since 2009. Groundwater quality determinants (e.g., EC, O₂, pH, N_{total}, NO₃⁻, NH₄⁺, SO₄²⁻, Cl⁻, Sb, As, Ni, Fe) are analyzed in these samples. This dataset was used to assess groundwater quality before mining activities started and was compared with data collected after the activities started, in order to identify changes due to mining.

2.3. Data visualization and statistical analysis

In order to derive an indicator for water quality in the river, Spearman correlation between EC and other determinants was examined. The significance level was set to $p \leq 0.05$, at which the null hypothesis of no correlation between EC and other determinants was rejected (H_0 = significant correlation). Moreover, regression analysis between EC and determinants that showed a significant correlation with EC was performed, using a linear regression model. Daily discharge observations were used to find the correlation between EC in natural conditions and discharge of the river, again using linear regression analysis.

Principal component analysis (PCA) was used to visualize differences in water quality for samples taken at the river stations and the inflow and outflow waters of the two treatment peatlands. The following water quality determinants were considered in PCA: O₂, pH, EC, SO₄²⁻, N_{total}, Mn, Fe, As, Sb, Ni, and Cl⁻. However, Mg, Na, K, and Ca were excluded from the PCA, as they were not routinely sampled in the treatment peatland inflow and outflow waters. Any samples (i.e., combinations of sampling points and dates) with incomplete data (i.e., missing values for one or more of the selected determinants) were removed. Prior to PCA, the data were standardized (z-score normalization to 0 means and unity standard deviations) to allow for comparison of determinants with different scales and units. Samples from different sites were analyzed together, to show overall differences in water quality, and separately for the river stations, to show seasonal variations in water quality. Calculations were conducted in R using the vegan package (functions “decostand” and “rda” to perform standardization and PCA, respectively; Oksanen et al., 2019). To illustrate grouping of data points in different seasons, centroids of each season and 95% confidence ellipses were constructed in the same color as the datapoints, using the “ordiellipse” function.

In order to analyze seasonality of water quality in the river, available meteorological data were used. Daily temperature and precipitation data from three meteorological observation stations near the study site (Kittilä kirkonkylä, Kittilä Pokka, and Kittilä Kenttäröva; about 30-35 km from the site)

were obtained from the Finnish Meteorological Institute. The values for the three stations were combined and the overall mean was calculated. A configuration of four seasons was considered, as described for northern Finland by the Finnish Meteorological Institute (2017), but with some modifications based on the meteorological data and to fit calendar months into seasons. The seasons were categorized as: spring (April and May); summer (June, July, and August); autumn (September and October); winter (November, December, January, February, and March) (Fig. 2).

The standardized dataset used for the PCA was also used for cluster analysis of the water quality determinants in different seasons. Cluster analysis was conducted using the function “hclust” in R, with Euclidean distances as input. Dendrograms were created for each season using the unweighted pair group method with arithmetic mean (UPGMA).

To identify the determinants affecting $EC_{Station\#2}$ and $EC_{Station\#4}$, we used linear mixed effects regressions (LMM) fitted with maximizing the restricted log-likelihood (REML) (function “lme” from R package “nlme”; Pinheiro *et al.*, 2019) in R v.3.5.3 (R Core Team, 2019). The best models were found to be:

$$\text{Model I: } Y_{EC(station\#4)} = \alpha + \beta_1(Q_{TP-B}) + \beta_2(EC_{Station\#2}) + \beta_3(EC_{TP-B}) + \beta_3(Q_{River}) + a(\text{month}) + \varepsilon$$

$$\text{Model II: } Y_{EC(station\#2)} = \alpha + \beta_1(Q_{TP-B}) + \beta_2(EC_{Station\#2}) + \beta_3(EC_{TP-B}) + \beta_3(Q_{River}) + a(\text{month}) + \varepsilon$$

where in both models the variable month (a) was considered a random effect nested in eight years. Determinants that affected $EC_{Station\#2}$ in model I were EC_{TP-A} , Q_{TP-A} , and Q_{River} . Parameters that affected $EC_{Station\#4}$ in model II were $EC_{Station\#2}$, EC_{TP-B} , Q_{TP-B} , and Q_{River} . All the predictors were centered on the mean 0, to remove potential multi-collinearity. The best model was chosen based on the Akaike information criterion (AIC), by including the parameters that minimized AIC. This method was chosen since it is good for dealing with the risk of overfitting and under fitting. The residuals were tested for absence of temporal pattern and autocorrelation. Standard error (SE) was calculated for each . The model fit was examined by removing one variable at a time and seeing how this affected the AIC.

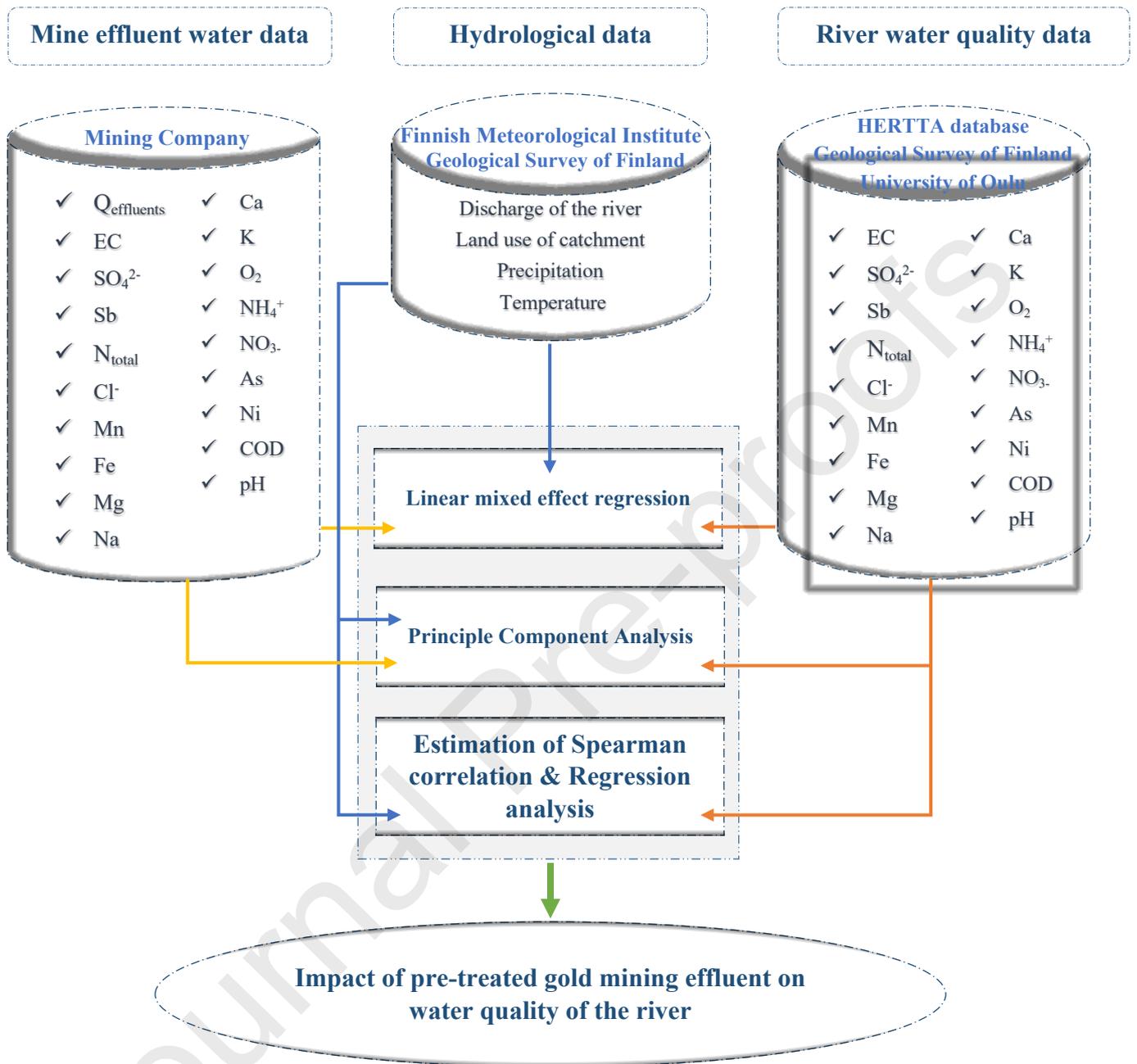


Fig. 2: Flowchart of available data and methodology applied in analysis of the Seurujoki river

3. Results and Discussion

3.1 Comparison of water quality in upstream and downstream river reaches

There was an obvious impact of mining on river water quality, as seen from water samples taken downstream of the mining area (Fig. 3 and Fig. 5a). EC and concentrations of N_{total} , SO_4^{2-} , and Sb were clearly elevated after the points of TP-A and TP-B discharge to the river. For instance, EC was $9.3 \pm 0.2 \text{ mS m}^{-1}$ at Station #1 (reflecting the baseline values without mining influence), but downstream from the mining site it increased to 23.4 ± 1.5 , 27.7 ± 1.0 , and $26.4 \pm 1.4 \text{ mS m}^{-1}$ at Station #2, Station #3, and Station #4, respectively (Fig. 3). These values are 2.5- to 3-fold higher than the upstream EC values, and are also higher than values reported for mining-impacted rivers in the UK (Jarvis et al., 2019). However, they are lower than values reported for mining-impacted rivers in China (86.5 and 223 mS m^{-1} in wet and dry season, respectively) (Sun et al., 2013). Mean EC in inflow water to TP-A ($\text{EC}_{\text{TP-A}}$) and TP-B ($\text{EC}_{\text{TP-B}}$) was 712 and 186 mS m^{-1} respectively. These values are within the range reported in previous studies of other gold and copper mines (e.g., Edraki et al., 2005). Based on the results of this study, the observed increase in river EC might have been caused by mining effluents (Fig. 4).

Mean N_{total} concentration increased from $142 \mu\text{g l}^{-1}$ upstream of the discharge point of both TPs (Station #1) to more than $600 \mu\text{g l}^{-1}$ downstream of the TP-B discharge point. Elevated nitrogen concentration in the river is stated as one of the major concerns in the environmental impact assessment of the mine (Pöyry, 2016). The SO_4^{2-} concentration also increased, from $5000 \mu\text{g l}^{-1}$ to $80,000 \mu\text{g l}^{-1}$, in the river (Fig. 3). Despite the clear increase in SO_4^{2-} and N_{total} concentrations, the values were generally lower (Edraki et al., 2005; Sun et al., 2013), but sometimes higher (e.g., Kusimi and Kusimi, 2012), than reported in other cases. Observed values were also below the limit values set by the World Health Organization (WHO, 2011) for drinking water (maximum $500,000 \mu\text{g l}^{-1}$ for SO_4^{2-} and $10,000 \mu\text{g l}^{-1}$ for N_{total}). The concentration of Fe was found to be 30% higher than the maximum permissible value for drinking water ($200 \mu\text{g l}^{-1}$) (Kumar and Puri, 2012) at Station #1, and 40-70% higher than the permissible value downstream of the mining area. However, Fe concentrations can be naturally high in Finnish waters (Helenius, 1981), especially in rivers with peatland-dominated catchments such as the Seurujoki. The Fe values observed did not exceed the toxicity threshold for aquatic life ($1000 \mu\text{g l}^{-1}$) (Kumar and Puri, 2012). Elevated Fe and SO_4^{2-} concentrations in discharge from mining activities are partly due to extraction of gold encapsulated within the crystal matrix of iron sulfide minerals (Fleming, 2010). Elevated nitrogen concentration has also been associated with use of explosives in mining activities (Chlot et al., 2011; Ernawati et al., 2018), which is common in Finland (Kujala et al., 2019; Mattila et al., 2007). Besides explosives,

certain mineral processing activities, including pH regulation, use of cyanide in gold extraction, and use of ammonia as a lixiviate, can generate significant nitrogen loads to the environment (Jermakka et al., 2015).

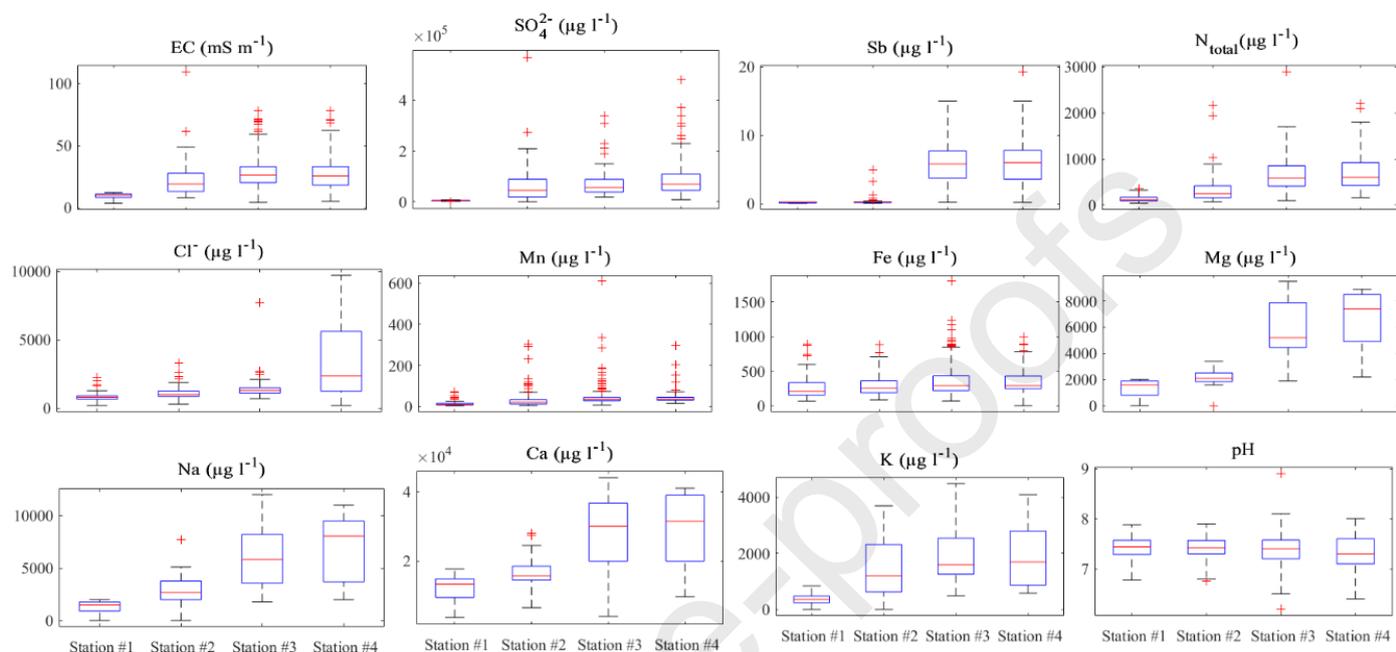


Fig. 3: Boxplot of water quality determinants at four monitoring stations on the Seurujoki river, based on data for the period 2010-2018. Station #1 is before the mining area, Station #2 is located 0.3 km downstream from the main discharge point of water from treatment peatland A (TP-A), Station #3 is located 6.4 km after TP-A, and Station #4 is located 7.2 km after TP-A.

Higher concentrations of different contaminants were observed at Stations #2-4 compared with Station #1 (Fig. 3), and this increase was likely caused by the discharge of mining-influenced waters. Although the levels of Cl^- , K , Ca , Na , and N_{total} increased due to mining, these substances were not present in river water in potentially harmful concentrations (WHO, 2011) (Supplementary information Table S1). There were no significant changes in the concentrations of Ni , COD , and As at different stations due to mining activities compared with the natural condition. The reason that Sb , Mg , K , and Na showed higher levels at Stations #3 and #4 was because of contributions of these contaminants from TP-B (Fig. 4). The efficiency of the TPs was evident, since the quality of pre-treated mine water improved after passage through these wetlands (O/TP-A and O/TP-A in Fig. 4). However, while these treatment peatlands work as a buffer zone, they contain large amounts of different chemicals that have been retained and could be transported away by heavy precipitation (Khan et al., 2019)

Comparison of data collected during the years before and after mining activities began in the area indicated that river water quality was similar at different stations in the time before mining (data for

2007-2010). Even though sampling was sparse and some mine-related construction had already started in 2008, water samples taken at Station #2 and Station #4 showed similar EC and pH levels as samples taken at Station #1, located upstream of the mine.

In general, the groundwater component is important in Arctic rivers and typically contributes the majority of water to river systems during low-flow conditions. In the Seurujoki catchment, groundwater monitoring wells outside the active mine area showed unchanged water quality values before and after mine construction (data for the period 2009-2018) (Supplementary information Fig. S1). Baseline groundwater quality is relatively similar to the river water quality at Station #1 (pre-impacted station). However, groundwater quality data collected from the monitoring well within the active mining area showed a clear increase in determinants (Supplementary information Fig. S1). This indicates that mining activities have a clear impact on groundwater quality. However, groundwater flow paths are towards the underground mine pit and do not directly influence the river water. Nevertheless, comprehensive research is needed to investigate and fully reveal possible impacts of mining activities on groundwater flow paths and quality in the catchment, since mining activities markedly alter groundwater flow paths and groundwater recharge and discharge areas.

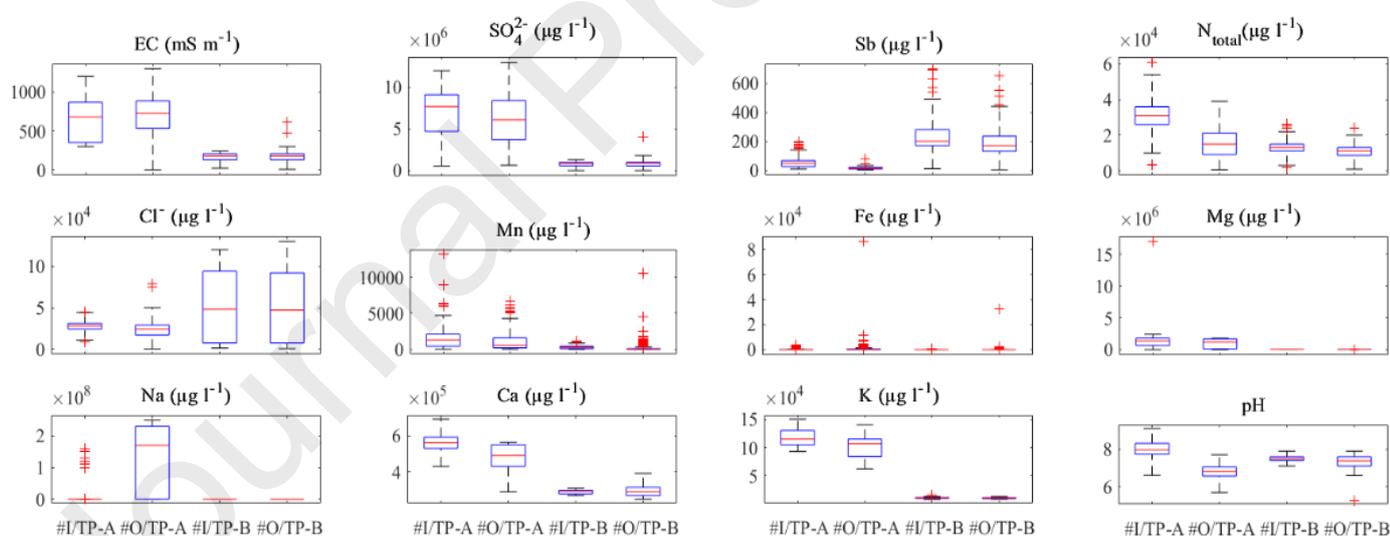


Fig. 4: Boxplot of different determinants of water quality of: pre-treated process water inflow to treatment peatland A (I/TP-A), outflow from treatment peatland A (O/TP-A), pre-treated drainage water inflow to treatment peatland B (I/TP-B), and outflow from treatment peatland B (O/TP-B).

Although TP-A and TP-B both treat mine water, they do not remove all substances (Fig. 4). The level of Sb was not decreased by TP-B and it was just conveyed through this peatland. The low concentration at Stations #3 and #4 is due to dilution (Fig. 4). The K and Na concentrations at the TP-A discharge point were elevated compared with those in common Finnish stream sediment (Lahermo

et al., 1996), but the impact of TP-A was very local and the K and Na concentrations in the river at different stations were within the acceptable range (WHO, 2011). Minimum, maximum, average, median, and standard deviation values of each determinant in all stations are presented, and compared with the requirements and guide values defined in Finnish regulations on drinking water quality (Mäkinen, 2008) and by WHO (2011), in Table S1 in Supplementary information.

3.2 Relationship between electrical conductivity and water quality determinants

Electrical conductivity showed a significant positive linear correlation with SO_4^{2-} , Sb, N_{total} , Cl⁻, Mg, Na, Ca, K, and Mn at the different stations (Table 1). A similar correlation has been found in previous studies (Ataee-pour and Rezaei, 2019; Luoto et al., 2019; Njinga and Tshivhase, 2017). There was a significant negative linear correlation between EC and Fe, Mn, COD, and As (Table 1). Electrical conductivity provides a useful water quality indicator, as it can be monitored continuously in order to detect sudden tailing dam leakages or irregularities in water treatment of mining waters. Additionally, EC measurements could provide the possibility to estimate other determinants (SO_4^{2-} , Sb, N_{total} , Cl⁻, Mn, Fe, Mg, Na, Ca, K) with significant reliability ($p < 0.05$). In natural conditions, the level of chemical substances in river water is very low and in some cases there is no significant correlation (e.g., EC and Sb, NH_4^+) while in other cases a significant negative linear correlation is apparent, as between EC and N_{total} and Mn.

Table 1: Spearman correlation and regression analysis between electrical conductivity (EC) and other determinants. Significant results of Spearman correlation (RHO>0.5, p<0.05) are highlighted and regression analysis are performed for these (EC*b+α= Determinant, p<0.05). RHO: Spearman correlation coefficient, n: Number of samples, b= regression coefficient.

EC	Station #1					Station #2					Station #3					Station #4				
	Spearman Correlation			Regression analysis		Spearman Correlation			Regression analysis		Spearman Correlation			Regression analysis		Spearman Correlation			Regression analysis	
	RHO	p	n	b	p	RHO	p	n	b	p	RHO	p	n	b	p	RHO	p	n	b	p
SO ₄ ²⁻	0.81	<0.05	92	509.83	<0.05	0.97	<0.05	92	5447.14	<0.05	0.95	<0.05	25	5100.00	<0.05	0.88	<0.05	82	4012.70	<0.05
Sb	0.18	<0.05	92			0.23	0.02	93			0.44	0.00	89			0.46	<0.05	92		
N _{total}	-0.53	<0.05	92	-20.09	<0.05	0.60	<0.05	93	14.74	<0.05	0.63	<0.05	89	20.19	<0.05	0.61	<0.05	92	14.22	<0.05
Cl ⁻	0.50	<0.05	92	82.88	<0.05	0.58	<0.05	92	21.03	<0.05	0.59	<0.05	25	85.38	<0.05	0.60	<0.05	79	80.47	<0.05
Mn	-0.63	<0.05	92	-3.93	<0.05	0.62	<0.05	93	2.03	<0.05	0.13	0.23	89			0.20	0.05	92		
Fe	-0.79	<0.05	92	-61.63	<0.05	0.21	0.04	92			-0.53	<0.05	89	-8.19	<0.05	-0.55	<0.05	91	-6.18	<0.05
Mg	0.94	<0.05	13	200.91	<0.05	0.66	<0.05	13	92.52	<0.05	0.93	<0.05	13	274.10	<0.05	0.84	<0.05	10	244.05	<0.05
Na	0.92	<0.05	13	161.11	<0.05	0.79	<0.05	13	313.60	<0.05	0.96	<0.05	13	357.20	<0.05	0.94	<0.05	10	327.46	<0.05
Ca	0.95	<0.05	24	1561.66	<0.05	0.54	<0.05	24	612.40	<0.05	0.90	<0.05	24	1110.78	<0.05	0.83	<0.05	18	1062.70	<0.05
K	0.86	<0.05	13	62.28	<0.05	0.67	<0.05	13	163.75	<0.05	0.88	<0.05	13	103.74	<0.05	0.94	<0.05	10	104.57	<0.05
pH	0.17	0.11	92			-0.13	0.22	93			0.02	0.86	89			0.06	0.57	87		
COD	-0.91	<0.05	92	-809.60	<0.05	0.20	0.05	93			-0.39	0.01	50			-0.49	<0.05	92		
O ₂	0.25	0.02	95			0.06	0.59	95			-0.05	0.82	25			0.05	0.66	82		
NH ₄ ⁺	-0.06	0.60	92			0.56	<0.05	93	6.79	<0.05			NDA			0.50	<0.05	92		
NO ₃ ⁻	0.75	<0.05	90	7.17	<0.05	0.33	<0.05	91					NDA			0.75	<0.05	90	14.53	<0.05
Ni	0.03	0.76	92			0.31	<0.05	93			0.41	<0.05	89			0.39	<0.05	92		
As	-0.61	<0.05	92	-0.12	<0.05	0.12	0.24	93			-0.46	<0.05	89			-0.39	<0.05	92		

3.3 Seasonality of water quality determinants in the Seurujoki River

The seasonal variation in water quality was rather small at the river sampling points (Fig. 5 c-f), except for EC at Station #1, which is explained in more detail later. At Station #2, winter samples clustered separately from spring-autumn samples in the PCA, while at Stations #3 and 4 winter and spring samples clustered separately from summer and autumn samples (Fig. 5). Winter and spring samples showed higher inter-annual variability than summer or autumn samples, which might be due to differences in snowmelt onset in different years. Similar results have been reported by Sun et al. (2013), who found no clear seasonal variation between wet and dry periods in non-impacted catchments, but dramatic seasonality in mine-impacted catchments, especially for SO_4^{2-} and Fe.

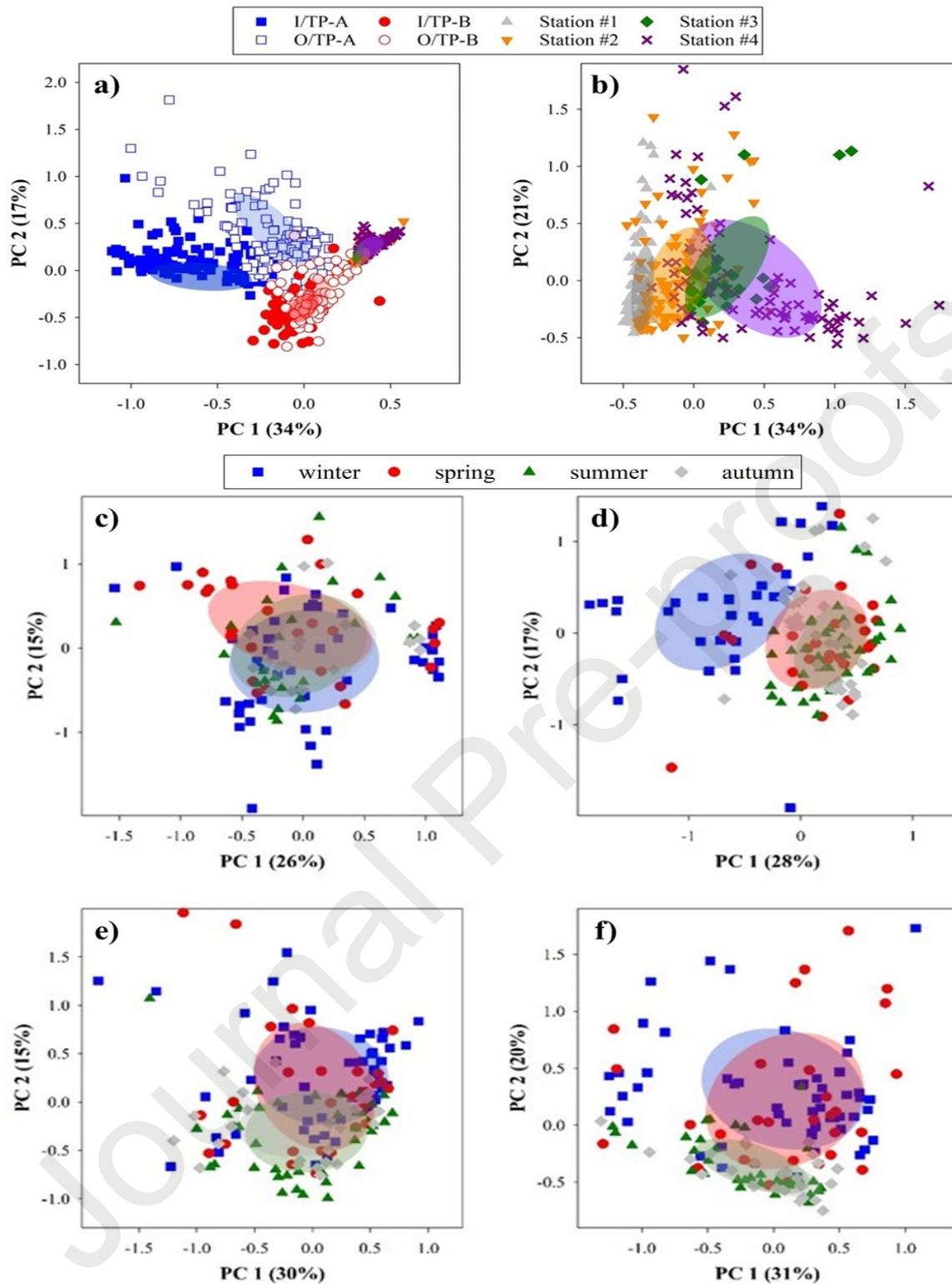


Fig. 5: Results of principal component analysis (PCA) illustrating the differences in water quality for all (a) and river (b) sampling points and seasonal variations in water quality at river stations #1 (c), #2 (d), #3 (e), and #4 (f). PCA was performed using the R *vegan* package with z-score transformed data. To illustrate grouping of data points in different seasons in panels c) through f), centroids of each season and 95% confidence ellipses were constructed in the same color as the data points, using the 'ordiellipse' function. The percentage of variance explained by each principal component (PC) is given in brackets. For the PCA in panel e) O₂ and SO₄²⁻ data were not used as these determinants were not measured on all sampling occasions at station #3.

Cluster analysis of water quality determinants from different river stations showed similar clustering in winter, summer, and autumn (Fig. 6). In these seasons, EC and SO_4^{2-} clustered closely together in the dendrogram, indicating that those determinants are highly correlated. N_{total} , Sb, and Cl^- fell near the EC/ SO_4^{2-} cluster, while pH, O_2 , and As were the most distant from EC/ SO_4^{2-} , indicating that those determinants behave quite differently. SO_4^{2-} is one of the major contaminants in mining-affected waters, and EC could thus be a feasible indicator for the influence of mining-affected waters on river water quality in winter, summer, and autumn. In spring, EC did not cluster closely with SO_4^{2-} but rather with Sb and Cl^- (Fig. 6). This suggests that slightly different mechanisms are active in spring than in the other seasons, which might be caused by the snowmelt events.

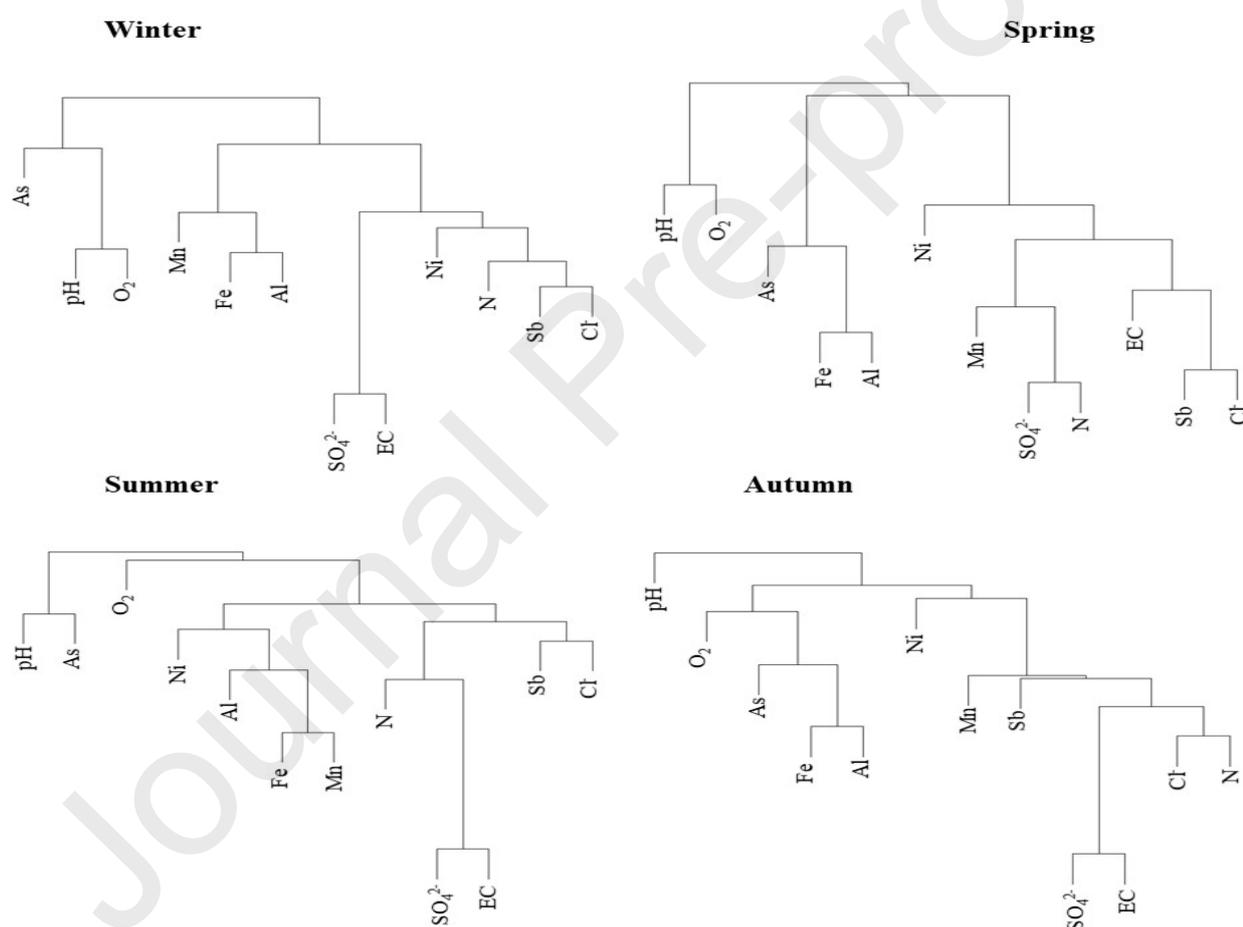


Fig. 6: Cluster analysis of water quality data from the river sampling stations in different seasons. Dendrograms were constructed using hclust with average linkage on Euclidean distances of z-score-transformed data in R. Seasons: Winter= November-March, Spring= April-May, Summer=June-August, Autumn=September-October.

Even though seasonal variation in overall water quality was not pronounced in the section with mining impacts, single determinants could be used to illustrate seasonal variations in the river sampling points. EC was used as an indicator for the influence of mining-affected water on the river, as suggested by the outcome of the cluster analysis.

The seasonal variation in EC was assessed using 2840 observations. A clear seasonal pattern in EC with the natural flow regime was observed at the point of no mining influence (Station #1) (Fig. 7b). At the sites influenced by mining effluent discharge, the fluctuation was different (Fig. 7c and Fig. 7d). The EC values were lowest in high-flow conditions during snowmelt (April and May) and in late autumn-early winter (October and November), when precipitation is typically high and evapotranspiration is low in the study region (Fig. 7a). This was confirmed by correlation analyses, where a negative correlation between $EC_{\text{Station\#1}}$ and Q_{River} was observed ($p < 0.05$) (Fig. 7b). The results confirm previous findings that river EC is a good predictor for Q_{River} and vice versa (Comina et al., 2013; Weijs et al., 2013). Decreasing EC with increasing river discharge due to snowmelt and runoff would be explained by the larger amount of water causing dilution, and thus fewer anions and cations per unit volume of river water. However, increased runoff (resulting in higher Q_{River}) could also increase dissolution of contaminants, as found in studies in abandoned mines (Jarvis et al., 2019)

Due to dominating impacts of mining activities on the river section downstream of the mine, there was no linear correlation between EC and river discharge at Stations #2-4 (Fig. 7c and Fig. 7d). It is well known that EC indicates the amounts of anions and cations in water and, since the concentration of sulfate and other substances is increased by mining water entering the river, EC at Stations #2 and Station #4 did not follow the natural pattern caused by seasonal variation in river discharge as was the case at Station #1 (Fig. 7b).

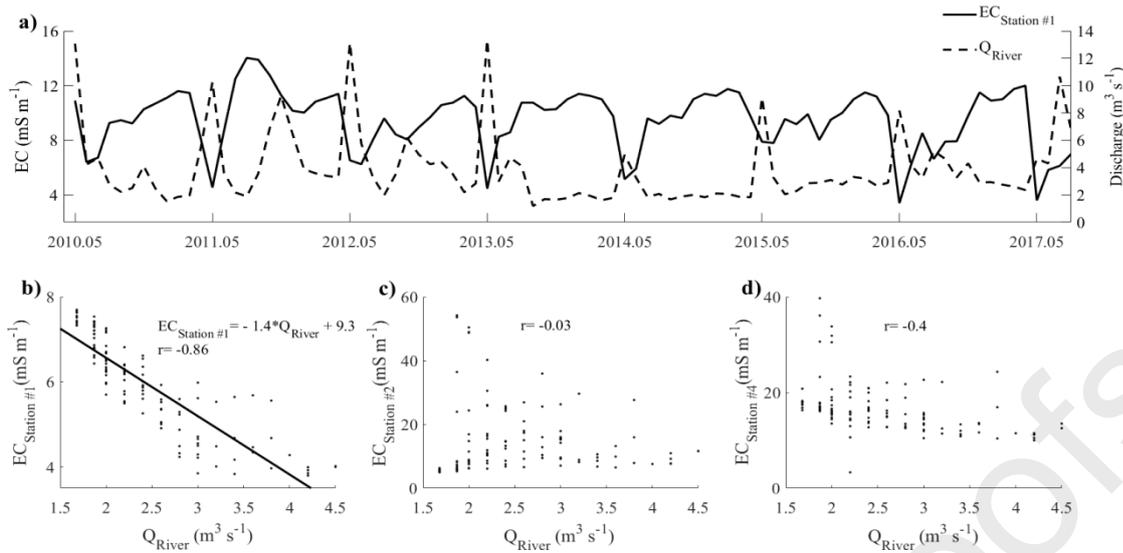


Fig. 7: a) Variation in electrical conductivity at Station #1 ($EC_{\text{Station}\#1}$) and river discharge (Q_{River}) representing natural river water quality without the influence of mining water, based on data for 2010-2018. b) Linear regression between $EC_{\text{Station}\#1}$ and Q_{River} (Pearson correlation coefficient $r = -0.86$); c) linear regression between $EC_{\text{Station}\#2}$ and Q_{River} ($r = -0.03$); and d) linear regression between $EC_{\text{Station}\#4}$ and Q_{River} ($r = -0.4$), based on data for July 2015 to November 2015.

The EC values at Stations #2 ($EC_{\text{Station}\#2}$) and #4 ($EC_{\text{Station}\#4}$) were significantly affected by the discharge from TP-A ($Q_{\text{TP-A}}$) and TP-B ($Q_{\text{TP-B}}$), respectively ($p < 0.05$) (Table 2). However, the estimated coefficient of $Q_{\text{TP-A}}$ (0.263) was higher than that of $Q_{\text{TP-B}}$ (0.094). Moreover, $EC_{\text{Station}\#4}$, but not $EC_{\text{Station}\#2}$, was significantly affected by Q_{River} ($p < 0.05$) (Table 1). $EC_{\text{TP-A}}$ contributed to $EC_{\text{Station}\#2}$, but the effect was small compared with the estimated effect of $Q_{\text{TP-A}}$ (Table 1). On the other hand, $EC_{\text{TP-B}}$ did not have significant impact on $EC_{\text{Station}\#4}$. The main reason might be the long distance between the TP-B outlet and Station #4 (around 7.2 km). $EC_{\text{Station}\#4}$ was significantly affected by $EC_{\text{Station}\#2}$, as illustrated in Fig. 8b. $EC_{\text{Station}\#2}$ and $EC_{\text{Station}\#4}$, with approximately 10 hours travel time in between, followed the same pattern, which was affected by $Q_{\text{TP-A}}$ and Q_{River} , respectively (Fig. 8b).

Table 2: Parameter estimates from analysis of the linear mixed model effect (lme) of electric conductivity (EC) at Stations #2 and #4 ($EC_{\text{Station}\#2}$ and $EC_{\text{Station}\#4}$), 2010-2018. SE = standard error, AIC = Akaike information criterion

	Parameter	Estimated coefficient	SE	P-value	AIC
Model I ($EC_{\text{Station}\#4}$)	Intercept	26.495	2.01	0.001	669.5
	$Q_{\text{TP-B}}$	0.094	0.03	0.006	
	$EC_{\text{Station}\#2}$	0.765	0.12	0.001	
	$EC_{\text{TP-B}}$	-0.008	0.01	0.557	

	Q_{River}	-1.325	0.47	0.006	
Model II ($EC_{\text{Station}\#2}$)	Intercept	22.358	0.91	0.001	
	$Q_{\text{TP-A}}$	0.263	0.05	0.001	647.2
	$EC_{\text{TP-A}}$	0.01	0	0.019	
	Q_{River}	-0.367	0.4	0.355	

3.4 Tailing dam accidental leakage

A tailing dam leakage occurred in September 2015 and the exposure was illustrated in EC records at 22:00 h on 9 September (Fig. 8b). The discharge from the leak was estimated to be $340 \text{ m}^3 \text{ h}^{-1}$. The immediate action was to pump the leakage water back into the tailing dam pool and block the leak by adding $36,000 \text{ m}^3$ of moraine to the tailing dam. Even though the accident was controlled immediately, its impacts were evident in Seurujoki river water quality (Fig. 8). The leakage accident increased river EC significantly, to approximately 60 mS m^{-1} (Fig. 8b). The EC was higher than the threshold value of 50 mS m^{-1} considered harmful for aquatic life (Abah et al., 2018) for about 10 days after the accident.

At Station #4, 6.4 km below TP-A, EC increased to 43 mS m^{-1} compared with a mean value of 26 mS m^{-1} in the period 2010-2018. At the station before the mine (Station #1), EC was 5.8 mS m^{-1} during the period of the accident. While EC increased downstream of the mine, the mean monthly $EC_{\text{TP-A}}$ and $EC_{\text{TP-B}}$ did not change markedly. For September 2015, $EC_{\text{TP-A}}$ and $EC_{\text{TP-B}}$ was 795 and 205 mS m^{-1} , respectively, compared with 712 and 185 mS m^{-1} , respectively, in the period 2010-2018. Mean $Q_{\text{TP-A}}$ and $Q_{\text{TP-B}}$ was 0.03 and 0.08 $\text{m}^3 \text{ s}^{-1}$, respectively, in 2010-2018 and 0.01 and 0.1 $\text{m}^3 \text{ s}^{-1}$, respectively, in September 2015 (Fig. 8a). This confirmed that the observed peak in EC was not due to a peak in the mine effluent flowing through TP-B and TP-A. The leakage accident also had an impact on SO_4^{2-} concentration, which increased dramatically at stations below the mining area. The average value of SO_4^{2-} during 2010-2018 was 66.54 mg L^{-1} at Station #2, while in September 2015 it increased by 62%. Similar results were obtained for Station #3, where the increase was around 40%, and Station #4, where the increase was 48%.

This leakage accident was reported to the authorities and has been well covered by the media due to high public interest following a leakage accident at the Talvivaara mine in 2008 (Parviainen and Loukola-ruskeenieni, 2019; Sairinen et al., 2017). However, in the Seurujoki river system, the EC in river water during the accident was not high and even higher values have been recorded in the river during the history of the mine (Fig. 8a). In particular, high EC values recorded in October 2012 and November 2013 were almost twice the values detected during the accident in 2015 (Fig. 8b). These

two peaks in EC and their causes were not assessed at the time, and thus any possible environmental impacts were not reported. Even though the mining company is required by law to meet all water quality regulations and even though the discharge is passed through quite efficient treatment peatlands (Khan et al., 2019), there have been quite high peaks in river EC that pose concerns regarding aquatic life and ecosystems in the river.

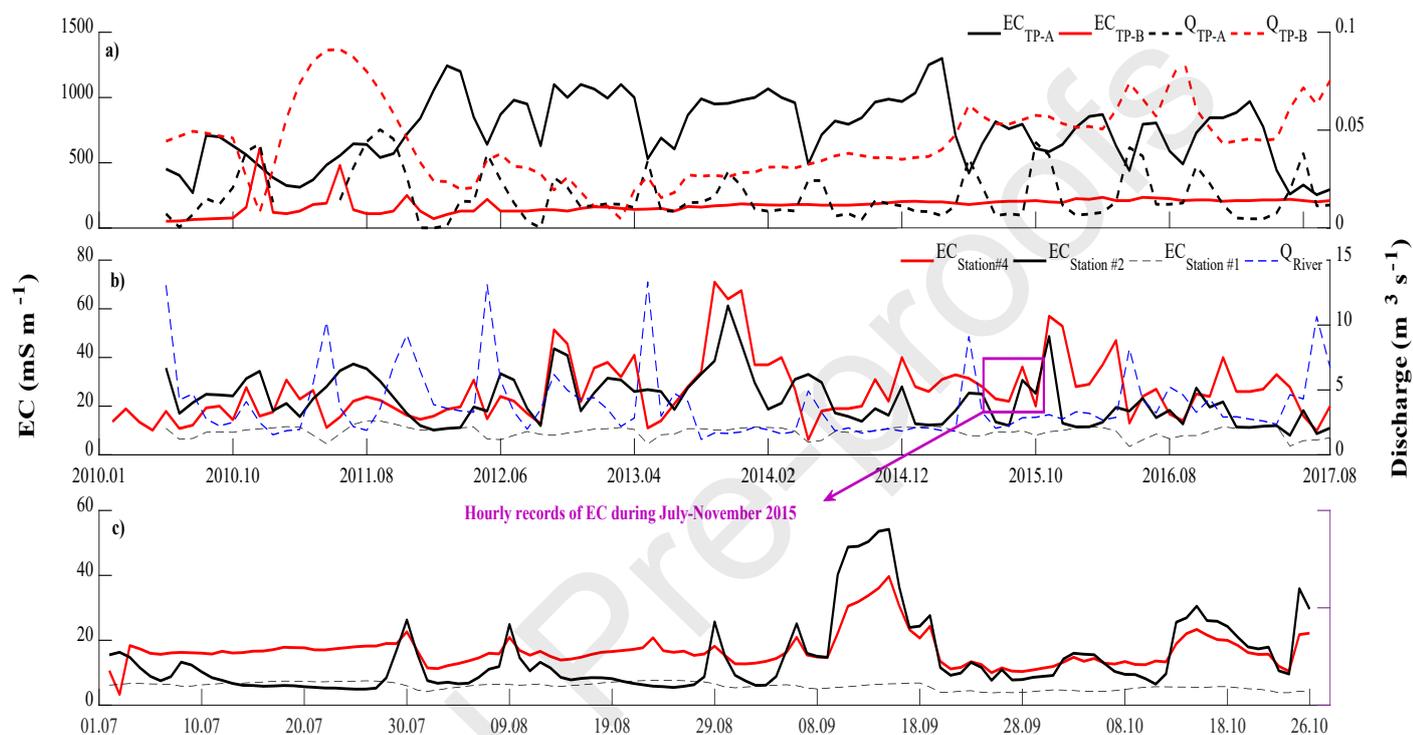


Fig. 8: Electrical conductivity (EC) and discharge (Q) fluctuations a) in treatment peatlands (TP) A and B; b) at Stations #1, #2, and #4 based on monthly data; and c) at Stations #1, #2, and #4 based on hourly data for July-November 2015.

3.5 River water quality monitoring and assessment of mining impacts

Monitoring of river water quality is an essential part of environmental management to assess the efficiency of water treatment measures and detect potential impacts of mining, including mining accidents. A comprehensive water quality monitoring program identifies all water sources and operating units in the overall balance equation, monitors all key determinants, measures determinants reliably and at sufficient intervals, and associates these data with the water management program. As the mine moves through the different phases of its operating life, the water monitoring program may need to change. Following a phase of gathering information and data, the knowledge obtained can be used to identify the need to revise and update the monitoring program (Debén et al., 2017).

Changes in ore extraction and beneficiation processes or changes in active water purification processes at the mine typically influence the ratio of different contaminants in mine waters. Events

such as spring snowmelt, heavy rainfall, or mine closure will also influence the concentrations of contaminants, by diluting the inflow waters to treatment peatlands and ultimately affecting surface water quality in the recipient river (Khan et al., 2019). Natural peatlands are used as mine water treatment systems in Finland (Isokangas et al., 2019; Khan et al., 2020; Larkins et al., 2018), and temporal fluctuations in site chemistry may lead to these peatlands becoming a net source of contamination for the adjacent river system (Palmer et al., 2015).

Our results show the benefit of continuous monitoring of EC upstream and downstream of mining sites. As the river concentration is variable, we recommend that such monitoring be a mandatory precondition for any environmental permit. With future advances in technology, on-line and continuous measuring sensors could be available to provide early warning indicators and monitor sudden changes and problems in mining activities based on high-quality data. The EC values obtained should be below 50 mS m^{-1} to enable the survival and growth of diverse aquatic life (Behar, 1997; EPA, 2011; Tziritis, 2014), a condition which was always fulfilled at Station #1, but not at other stations in this study.

4. Conclusions

Long-term water quality data from monitoring stations upstream and downstream of a gold mine were analyzed and compared with available data for the pre-mining period, in order to determine the impact of mining activities on the river in a sensitive sub-Arctic region where mining, tourism, and natural values are in conflict. The pre-treated mine water changed the seasonal patterns of water quality determinants along the river. In the river section before mining effluent entered, EC measurements showed a strong seasonal pattern and correlation with river discharge, but downstream of the mine the correlation decreased or disappeared. The data also indicated clear impacts of pre-treated mine waters in the river studied, with e.g., marked increases in N_{total} , SO_4^{2-} , and Sb concentrations in river water. The level of water quality determinants remained high for about 7.2 km from the uppermost discharge point of mine waters (the outlet of a treatment wetland). Furthermore, groundwater quality has changed in the mining area and outside the mine pit, towards which groundwater flows. Although the contaminant concentrations measured were below the maximum permissible concentrations for drinking water, they were 4- to 16-fold higher than the natural concentrations in the river.

Continuous EC monitoring proved useful for detecting and monitoring changes in river water quality and can serve as a cost-efficient early-warning method to detect sudden changes water quality in mine-impacted catchments. It could be used to detect leakages from tailings dams etc. and to provide valuable information on how far contaminated waters travel in downstream water systems. In this

study, high peaks observed in EC reflected irregularities in water treatment processes at the mine, or even some other unreported or undetected leakage accidents. Continuous EC monitoring along the river would allow mining companies and environmental authorities to determine the impacts of mining over time.

CRedit authorship contribution statement

Navid Yaraghi: participated in planning of the work, field work, interpretation of data, preparation of illustrations, writing and overall writing work flow administration.

Anna-Kaisa Ronkanen: participated in planning and supervision of the work, field work, interpretation of data, preparation of illustrations, writing and review of the work.

Ali Torabi Haghighi: participated in planning and supervision of the work, field work, interpretation of data, preparation of illustrations, writing and review of the work.

Mahdi Aminikhah: carried out the work related to executing linear mixed model and writing of related sections.

Katharina Kujala: participated in planning of the work regarding overland flow fields, field work, PCA and cluster analysis, and writing.

Björn Klöve: participated in planning and supervision of the work, interpretation of data, design of illustrations, writing and review of the work.

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Impacts of gold mine effluent on water quality in a pristine sub-Arctic river

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Abstract

Impacts of mining on water quality are a great concern in the Arctic region. This study evaluated the impact of pre-treated mine effluent on river water quality. The study was conducted along the Seurujoki River in sub-Arctic Finland, which is impacted by Kittilä gold mine. The study analyzed water quality and hydrological data upstream and downstream of the mining area over an eight-year period, including a tailing dam leakage event in 2015. The analysis focused on water quality determinants such as electrical conductivity (EC), sulfate, antimony, manganese, and total nitrogen (N_{total}). Descriptive statistics on river water at four stations along the river corridor showed negative impacts of mining activities on the recipient water body. In order to find an indicator for water quality, correlation analysis between the water quality determinants was carried out. It identified EC as a good indicator for continuous water quality monitoring, especially to detect mining accidents such as partial failure of a tailings dam. The results showed increasing contaminant concentrations due to mining as more mine effluent was generated over time. A linear mixed model was developed to

predict the coefficient of different elements affecting EC at river water monitoring stations impacted by mining effluents. The results provide new information on how to assess mining water impacts and plan future water quality monitoring.

Keywords: Mining, environmental impacts, contamination, accident, Finland.

Credit author statement

Navid Yaraghi: participated in planning of the work, field work, interpretation of data, preparation of illustrations, writing and overall writing work flow administration.

Anna-Kaisa Ronkanen: participated in planning and supervision of the work, field work, interpretation of data, preparation of illustrations, writing and review of the work.

Ali Torabi Haghighi: participated in planning and supervision of the work, field work, interpretation of data, preparation of illustrations, writing and review of the work.

Mahdi Aminikhah: carried out the work related to executing linear mixed model and writing of related sections.

Katharina Kujala: participated in planning of the work regarding overland flow fields, field work, PCA and cluster analysis, and writing.

Björn Klöve: participated in planning and supervision of the work, interpretation of data, design of illustrations, writing and review of the work.

Highlights

- Pre-treated mine effluent elevated water quality determinants in a sub-Arctic river
- Determinant's level remained high for about 7 km after the discharge point
- Pre-treated mine effluent changed seasonal patterns of water quality determinants
- EC proved to be a good water quality indicator in a pristine sub-Arctic river
- Continuous EC monitoring is recommended to monitor changes in river water quality

Declaration of interests

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

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



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