



Efficient removal of arsenic, antimony and nickel from mine wastewaters in Northern treatment peatlands and potential risks in their long-term use



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ABSTRACT

The number of metal mines, the amount of ore processed, and thus the amount of mine wastewaters have been rapidly increasing in recent decades. Pretreated process effluent and drainage waters containing high concentrations of e.g., arsenic (As), antimony (Sb), and nickel (Ni) have been purified for up to 6 years in treatment peatlands at a gold mine in Finnish Lapland prior to their release into downstream water bodies. Retention efficiencies based on monitoring of peatland inflow and outflow waters indicated that retention of As, Sb, and Ni was generally good (up to 95%) but temporarily variable. Other contaminants such as sulfate or iron were retained less efficiently or were even leaching from the peatland. Most of the As, Sb, and Ni was retained close to the wastewater distribution ditch, and there was a near-linear increase in As, Sb, and Ni concentrations in the peatlands over time. Adsorption experiments indicated the high potential of peat soils to adsorb As, Ni and especially Sb. Accumulation of contaminants has led to or will lead to concentrations exceeding higher guideline values for contaminated soils within the next 1–18 years, which indicates a limited lifetime for treatment peatlands. Leaching of contaminants is anticipated in situations where clean natural waters mix with mine waters, e.g., during snowmelt or after mine closure, and desorption experiments suggest that especially easily mobilized (i.e., loosely-bound) fractions of As, Sb, and Ni are likely to leach from the treatment peatlands. The collective data indicate (i) high purification efficiencies for As, Sb, and Ni in the peatlands, (ii) high adsorptive capacity of the peat soil for those contaminants, (iii) increased risk of contaminant leaching e.g., after mine closure, and (iv) a need for further studies of environmental parameters and retention processes affecting contaminant removal efficiencies and potential lifetimes of treatment peatlands.

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1. Introduction

Mining is an important industrial branch worldwide. The increasing number of operating mines raises concerns for health and environment, and many of those concerns are related to mine water issues. Mining operations often produce substantial amounts of acidic wastewater, including mine drainage waters from pits and underground mines (groundwater and intruding surface waters), process waters stemming from the metal beneficiation process, and seepage waters from mine tailings and waste rock piles. Even though process-based and drainage wastewaters can be recycled in the enrichment process to a certain extent, most of the water is finally discharged from the mine to downstream water bodies after clarification, chemical and/or

passive water treatment (Kauppila et al., 2011; PIRAMID Consortium, 2003; van Berkel, 2007). Constructed wetlands (CW) are one of the passive treatment methods which are successfully and widely used for different types of runoffs and wastewaters including acidic mine waters (Koskiaho et al., 2003; Liikanen et al., 2006; Sharma et al., 2013; Sheoran and Sheoran, 2006; Silvan et al., 2003; Vymazal, 2011). Wetlands function either alone or in combination with chemical or physical pretreatment. They are generally well-suited for purification purposes as they retard the flow of water and provide a large filtration network with many adsorptive surfaces on plant roots, tissue or soil organic and mineral particles (Ronkanen and Kløve, 2009; Vymazal, 2011).

Mine wastewaters contain highly elevated concentrations of potentially harmful substances derived from process chemicals, explosives, or sulfate (SO_4^{2-}) minerals, including trace metals and metalloids (e.g., nickel (Ni), arsenic (As), lead (Pb), antimony (Sb)), nitrogen (N) compounds, SO_4^{2-} , and other contaminants such as carboxymethylcellulose, phosphate (PO_4^{3-}), oil, xanthates, and

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salts (Bissen and Frimmel, 2003; Kauppila et al., 2011; Nordstrom, 2011; van Berkel, 2007). Elevated concentrations of metals such as copper (Cu), zinc (Zn), nickel (Ni), or mercury (Hg) are toxic to many organisms, as they are non-biodegradable and accumulate in cells and tissues (Fu and Wang, 2011). Also metalloids such as arsenic (As) accumulate in plant and animal tissue, which can lead to acute or chronic arsenicosis, depending on the amount and duration of As intake (Bissen and Frimmel, 2003; Ratnaïke, 2003). As toxicity largely depends on its bioavailability, thus more bioavailable inorganic As compounds such as arsenate and arsenite are generally more toxic than organic As compounds (Ratnaïke, 2003). The example of As illustrates why it is of the utmost importance to remove contaminants from mine wastewaters prior to their discharge into natural water bodies. Removal of metals and metalloids in CWs is based on a variety of chemical, physical, and biological processes such as adsorption, chemical or biological oxidation or reduction, precipitation and sedimentation, or plant uptake (Dunbabin and Bowmer, 1992; Fu and Wang, 2011; Kosolapov et al., 2004; Sheoran and Sheoran, 2006). The prevailing processes and the efficiency of contaminant removal are dependent on various factors, including input water quality, wetland hydrology, system pH, oxygen saturation, redox conditions, and temperature (Sheoran and Sheoran, 2006; Webb et al., 1998). Permanent or temporary water saturation is of critical importance in the clean-up process, as carbon, N, sulfur, metals, and metalloids accumulate in peatlands under anoxic conditions (Sheoran and Sheoran, 2006; Shotyky, 1988). Due to the high degree of water saturation, especially the lower layers in wetlands are often anoxic and allow for the reduction of NO_3^- , Fe^{3+} , Mn^{4+} , or SO_4^{2-} by microorganisms, creating reduced metal forms as well as sulfide (S^{2-}) (Gadd, 2004; Sheoran and Sheoran, 2006). Metal sulfides are largely insoluble, thus the formation and precipitation of metal sulfides is the main way of metal removal in wetlands under anoxic conditions (Sheoran and Sheoran, 2006; Webb et al., 1998). On the other hand, metal(loid)-hydroxides are formed in oxic layers of wetlands and near plant roots, as plants introduce oxygen into the soil and thus create steep oxygen gradients in the surroundings of plant roots (Sheoran and Sheoran, 2006; Webb et al., 1998). This can moreover lead to the precipitation and coprecipitation of iron (Fe) and manganese (Mn) oxides and As oxides, respectively, on the root surface (Dunbabin and Bowmer, 1992; Kosolapov et al., 2004; Vale et al., 1990).

Wetland-based mine water treatment is especially challenging in boreal environments (e.g., in Finland), which experience long and cold winters and short growing seasons. In Finland, seasonal snow cover is encountered from November to May and mean annual temperatures range between -3 and 6°C (Pirinen et al., 2012). Thus, temperature dependent biogeochemical purification processes in northern treatment wetlands are strongly impacted by the highly variable climatic conditions throughout the year. Moreover, mine sites can be located in remote areas, e.g., in Finnish Lapland, which are often surrounded by pristine environments which should be protected from environmental damage. However, the number of metal mines and the mined ore levels are rapidly increasing in Finland, driven by the general rise in the price levels of base metals, Fe, and gold (Kauppila et al., 2011; Tuusjärvi, 2013). This trend has raised fears of environmental destruction, and the mining industry is already facing more intensive opposition than ever in its history in Finland (Tuusjärvi, 2013).

The use of pristine peatlands for purification of municipal wastewaters and runoff from forestry, peat extraction, and agriculture are quite well studied (e.g., Hynninen et al., 2011; Liikanen et al., 2006; Ronkanen and Kløve, 2009; Vymazal, 2011) while knowledge about retention of mine water contaminants in natural northern pristine peatlands is scarce. Processes occurring during wastewater purification in natural wetlands are more

Table 1
Chemical composition of influent water to treatment peatlands 1 and 4. Total concentrations in unfiltered water samples were determined during the environmental monitoring by the mining company.

Parameter	pH	EC ^c (mS/m)	Total solids (mg l ⁻¹)	Alkalinity (mmol l ⁻¹)	Total P ^d (μg l ⁻¹)	Total N ^e (mg l ⁻¹)	SO ₄ ²⁻ (mg l ⁻¹)	HCO ₃ ⁻ (mg l ⁻¹)	Ca (mg l ⁻¹)	Mg (mg l ⁻¹)	Na (mg l ⁻¹)	K (mg l ⁻¹)	Li (mg l ⁻¹)	Mn (mg l ⁻¹)	Fe (μg l ⁻¹)	Ni (μg l ⁻¹)	As (μg l ⁻¹)	Sb (μg l ⁻¹)
Peatland 1 ^a	7.18–7.84	94.3 ± 1.70	2.00 ± 0.14	2.26 ± 0.03	8.41 ± 0.37	8.83 ± 0.43	374 ± 11.2	138 ± 1.56	153 ± 3.01	31.8 ± 0.73	14.8 ± 0.42	4.67 ± 0.15	2.80 ± 0.19	0.42 ± 0.02	67.2 ± 10.8	68.9 ± 1.85	41.0 ± 1.79	160 ± 6.24
Peatland 4 ^b	7.80–8.60	97.9 ± 4.38	7.03 ± 0.24	2.05 ± 0.06	25.1 ± 0.41	25.3 ± 0.08	896.2 ± 51.7	123 ± 4.79	450 ± 20.4	1488 ± 30.6	114 ± 2.29	118 ± 6.29	0.22 ± 0.005	1.33 ± 0.02	109 ± 10.6	37.0 ± 0.98	140 ± 2.36	35.5 ± 0.38

^a Based on 13 measurements conducted in 2010–2011.

^b Based on 21 measurements conducted in 2013–2014.

^c Electrical conductivity.

^d Total phosphorus.

^e Total nitrogen.

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