Effluent Chemistry of Closed Sulfide Mine Tailings: Influence of Ore Type

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ABSTRACT

The seepage water quality of old sulfide mine tailings was studied from eleven closed sulfide metal mines in Finland to assess the influence of ore type on the drainage quality of tailings in the long-term. The studied sites represent a wide variety of ore types and commodities (e.g. Mo; Cu and Zn; Ni and Cu; Cu, W, As, Ag; Fe). Also the operation periods of the mines vary, from the 1750s to 1990s. Mining operations had ceased at the sites some 15–62 years ago (prior to sampling), but most of the facilities were left without any cover after mine closure. Only three of the tailings impoundments were covered with a thin layer of till or peat.

The seepage quality varied largely between the different mine sites. The pH of the seepages was between 2.8–7.3 and the total metal content (Zn + Cu + Cd + Pb + Co + Ni) between 0.004–207 mg/L. Overall, the high-acid, high-metal waters were related to Cu mine tailings, whereas the seepages from the other tailings deposits (i.e. from Ni, Fe, Mo, and Zn mines) were mostly near-neutral, low-metal containing waters. Unexpectedly, the most acidic seepage waters were found at the two sites where the tailings were covered with till after closure. One of the sites represented tailings from a Cu mine and the other from a Ni mine.

The results of the study show that there is a correlation between the tailings effluent quality and the ore type. However, other factors such as weathering period of the tailings also influence the seepage quality. These types of data contribute to the prediction of the long-term drainage quality of the new mining sites and in defining the requirements for their waste management and water treatment.

Keywords: Tailings, sulfide mining, seepage water, ore type, Finland

INTRODUCTION

Low-quality mine drainage from mine wastes is one of the major environmental issues that the mining operators struggle with. This is in particular at mines extracting base metals from sulfide-rich deposits, since the sulfide minerals oxidize once exposed to atmospheric oxygen and water. Depending on the balance between acid producing and neutralizing minerals in the waste, the drainage can be either low pH acid mine drainage ('AMD`) or non-acidic neutral mine drainage ('NMD') (Younger, 1995; Cravotta *et al.*, 1999; Pettit *et al.*, 1999; Heikkinen *et al.*, 2009). Regardless of the pH, these mining influenced waters can contain elevated to high concentrations of dissolved base and trace metals (*e.g.* Fe, Ca, Mg, Na, K, Cu, Ni, Zn) and sulfate originating from the weathering of sulfide, carbonate, oxide and silicate minerals in the waste (Pettit *et al.*, 1999; Plumlee *et al.*, 1999; Cravotta, 2008). Before opening a mine, quality of the future drainage should be estimated to assess the environmental impacts of the disposal of mine waste and to define the requirements for the waste management and water treatment techniques to prevent or minimize these impacts.

Prediction of the long-term quality of the waste effluents is a challenging task and is usually based on geochemical characterization of mine wastes using *e.g.* static and kinetic tests (White *et al.,* 1999; Sapsford *et al.*, 2008; INAP, 2009; Rousseau, 2012). Static tests provide one-time results and target to evaluate whether the waste is acid producing or not, whereas kinetic tests provide evaluation of the time dependent rates of chemical reactions in longer term. However, it has been recommended that the prediction should not be based on the tests alone, but they should be supplemented with water quality data from mine sites with similar types of geological deposits, because effluent chemistry shows ore type-specific features (Plumlee *et al.*, 1999; Lapakko, 2003; Seal & Hammarström, 2003).

In this study, preliminary results of the seepage water quality of tailings from eleven closed Finnish sulfide metal mine sites, representing six different genetic ore types, is presented to study the ore type-related effluent geochemical characteristics. At most of these sites the tailings have been uncovered since the mining operations ceased, and the tailings have had 15–62 years to weather. However, some of the tailings facilities were covered after closure with a layer of till or peat, so the influence of soil covers on the seepage water quality is also discussed. The focus of the study is in particular on the acidity and trace metal content of the seepages.

METHODOLOGY

Description of the studied sites

Seepage water samples were collected from tailings facilities of eleven closed sulfide metal mines in Finland (Figure 1), located in a temperate climate with an annual average precipitation of 600–700 mm. At all sites, the tailings were disposed above ground level into ponds constructed with earthen dams and utilizing natural depressions (*e.g.* small ponds/lakes or peatlands). The mines included Mätäsvaara Mo mine, Hammaslahti Cu-Zn mine, Laukunkangas Ni-Cu mine, Hällinmäki Cu-mine, Kotalahti Ni-Cu mine, Ylöjärvi Cu-W-As-Ag mine, Aijala Cu-Zn mine, Orijärvi Cu-Pb-Zn mine, Vihanti Zn-Cu-Pb-Ag mine, Rautuvaara Fe-Cu mine and Raajärvi Fe mine (Table 1). The ore deposit represented genetically 6 different ore types: volganogenic massive sulfide deposits, sedimentary exhalative type, porphyry copper deposits, magmatic nickel deposits, skarn and iron oxide ore types (Eilu *et al.,* 2012).

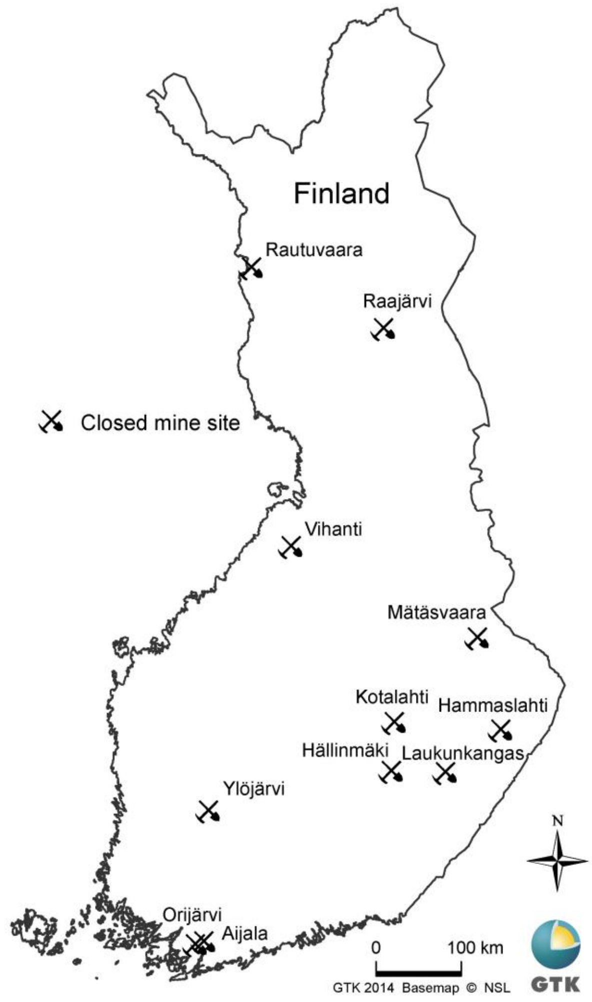

Figure 1 Locations of the studied mine sites (Contains data from the National Land Survey of Finland)

The mines operated variably from 1750s to 1990s (Puustinen, 2003, Eilu *et al.,* 2012), thus a variety of ore processing methods have been employed at the sites. Mining operations ceased at the sites some 15–62 years before the seepage water sampling (Table 1). Only three of the eleven tailings facilities (Hammaslahti, Laukunkangas, Vihanti) were covered after closure, so the tailings have been in most of the sites susceptible to oxidation. In Hammaslahti, the till cover was 35 cm thick in average (varying 10–60 cm) and it was partly covered with 10–15 cm layer of peat. Laukunkangas tailings area was covered with 50 cm till layer with 5 cm humus layer on top of it, whereas in Vihanti some 5 cm thick peat layer has been applied to revegetate the tailings facility. Covering of the sites has occurred only after some ten years after the mine closure and the end of the tailings disposal. Therefore, weathering of tailings has also initiated at these sites prior to covering.

Table 1 Commodity, ore type, operation period and amount of tailings of the studied mine sites

Mine site	Commodity	Ore deposit type[1]	Operation period[2]	Tailings (Mt)[3]
Aijala	Cu, Zn	VMS (bimodal, felsic dominated)	1949–58	2.0
Hammaslahti	Cu, Zn	Sedimentary exhalative	1972–86	5.3
Hällinmäki	Cu	VMS? (mafic)	1966–84	4.1
Kotalahti	Ni, Cu	Magmatic Ni sulfide	1959–87	9.4
Laukunkangas	Ni, Cu	Magmatic Ni-Cu-PGE	1985–94	6.6
Mätäsvaara	Mo	Porphyry (Cu, Au, Mo, W, Sn, Ag)	1903, 1910s, 1920–22, 1932–33, 1939–47	1.0
Orijärvi	Cu, Pb, Zn	VMS (bimodal, felsic dominated)	1758–1882, 1929–55	1.0
Raajärvi	Fe	Skarn (Zn-Pb-Ag, Cu, Au, Fe, W)	1964–75	2.2
Rautuvaara	Fe, Cu	Iron oxide-copper-gold	1962–88	8.0
Vihanti	Zn, Cu, Pb, Ag	VMS (bimodal, felsic dominated)	1954–92	1.4
Ylöjärvi	Cu, W, As, Ag	Close to Cu porphyry	1943–66	2.8

[1]Eilu *et al.*, 2012; [2]Puustinen, 2003; [3]Räisänen *et al.*, 2013; VMS = volcanogenic massive sulfide deposit

Sampling and analytical methods

Seepage water samples were collected from locations where water surfaces through the tailings dams. Samples were collected during early summer to midsummer (27th May to 3rd July). One seepage water sample was taken from each mine site. EC, pH, O_2, O%, and redox potential were measured in the field using a multi-parameter field meter (YSI 556 Multiprobe system), and alkalinity was determined with a Hach digital titrator with 0.1600 N or 1.600 N H_2SO_4 to an end point of 4.5. Unfiltered samples were collected for anion measurements and filtered (0.45 μm), HNO_3-acidified samples were collected for dissolved major cation and trace metal analyses. Major anions were determined with ion chromatography and major cations and trace metals with ICP-OES or ICP-MS. Duplicate samples and field blanks were taken for quality control. All the laboratory analyses were carried out at the FINAS-accredited testing laboratory of Labtium Ltd.

RESULTS AND DISCUSSION

Overall chemistry of the seepage waters and influence of the ore type

The seepage water quality varied largely between the different mine sites. The pH of the seepage waters varied between 2.8–7.3 and the total metal content (Zn, Cu, Cd, Pb, Co, Ni) between 0.004–207 mg/l (Figure 2). In general, the most abundant trace metals in the seepages were Zn, Ni, Co, and Cu and the seepages contained also high concentrations of SO_4, Ca, and Mg, which are typically released in the weathering of sulfide minerals and subsequent weathering of carbonate and silicate minerals (Figure 3).

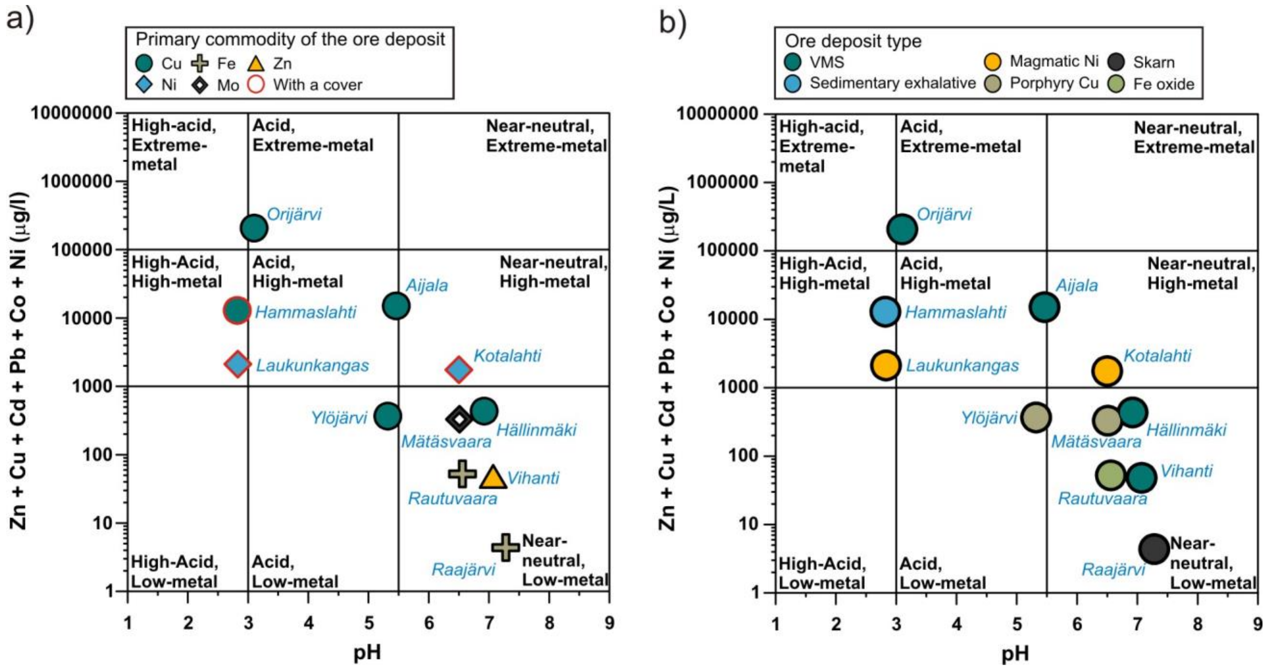

Figure 2 Ficklin diagrams (Plumlee *et al.*, 1999) of the tailings seepage waters showing the sum of dissolved heavy metals (Zn, Cu, Cd, Pb, Co, Ni) in µg/l plotted against pH value. Mine sites classified a) based on primary commodity of the ore deposit and b) based on genetic type of the ore deposit (VMS = volcanogenic massive sulfide deposit)

Based on the Ficklin diagram (Plumlee *et al.*, 1999), one third of the tailings seepages classified as high-acid/acid, high-metal waters, and half were near-neutral, low-metal drainages (Figure 2). Overall, the seepages at the sites where Cu ore had been extracted represented AMD type waters whereas the tailings effluents from most of the other types of deposits produced NMD (Figure 2). The most acidic seepage waters with highest metal contents were found at the sites where volcanogenic massive sulfide and sedimentary exhalative Cu ore types had been extracted, namely at Hammaslahti, Orijärvi and Aijala. These deposits contain typically high concentrations of sulfide minerals hosted by felsic rocks, having high acid production capacity and low neutralization capacity, and thus are prone to AMD. In contrast, the most neutral seepages with lowest metal contents were at the Raajärvi and Rautuvaara mine sites, which are iron oxide deposits containing only small amounts of sulfide minerals. However, the tailings produced in the extraction of magmatic Ni deposits (Laukunkangas and Kotalahti), porphyry deposits (Ylöjärvi and Mätäsvaara) and volcanogenic massive sulfide deposits (Vihanti, Hällinmäki, Aijala, Orijärvi) produced both AMD and NMD type seepage waters (Figure 2). These major ore type classes contain heterogeneous ore deposits, whose host rocks vary from ultramafic to felsic rocks, with varying buffering properties (Table 1). Host rocks of magmatic Ni deposits typically include ultramafic rocks with good buffering capacity, but in the Laukunkangas deposit the host rocks also contain black schists which are highly acid producing rocks (Grundström, 1985). Evidently, a more detailed classification of the ore type would likely better reveal the differences between the various ore deposits.

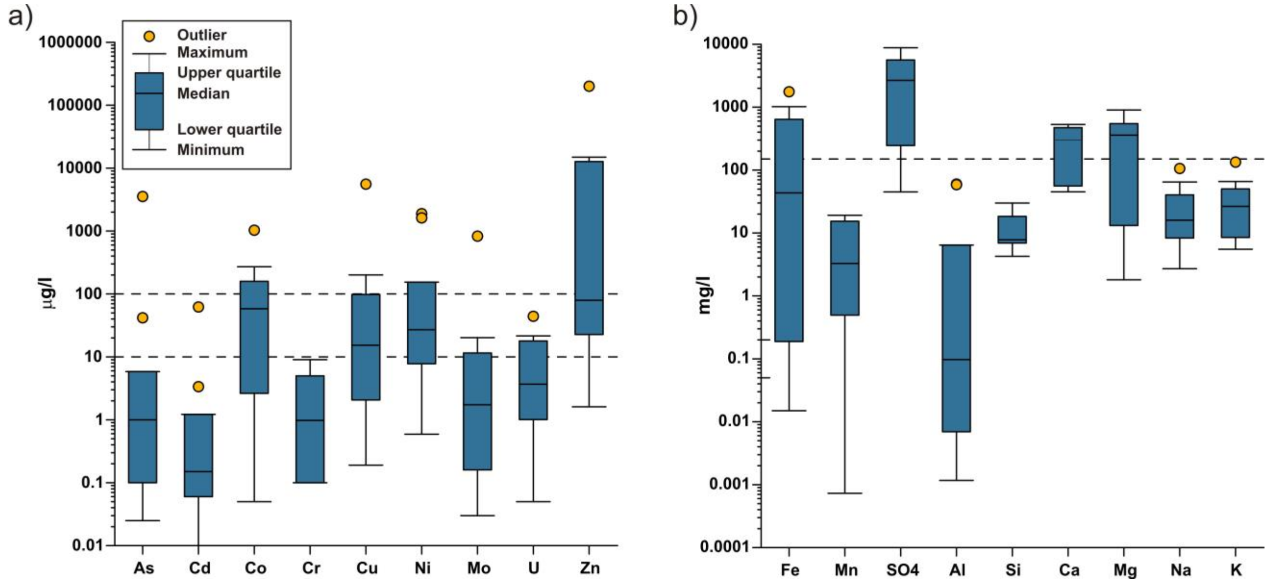

Figure 3 Box-Whisker plots for selected a) trace metal and b) major element and SO_4 contents of the tailings seepage waters

The loadings of trace metals in the seepages is a matter of the availability of the metals in the tailings (*i.e.* source of the metal) and the pH-Eh conditions controlling mineral weathering. Obviously, the tailings effluents of magmatic Ni deposits (Kotalahti and Laukunkangas) with abundant Ni sulfides in the tailings contained notably higher amounts of Ni than the other deposit types, whereas Cu concentrations were highest in the seepages of the Cu deposits (*e.g.* Orijärvi, Hällinmäki, Hammaslahti) rich in Cu sulfides (Figure 4). In addition, Zn was a typical trace metal in the seepages of the Cu deposit in which Zn occurred in exploitable amounts (Figure 4 and Table 1). However, occurrence of Zn in the seepages was also strongly linked to the pH conditions and thus in the weathering state of the tailings. For example, at the closed Vihanti Zn mine, the seepage water pH was > 7 and contained only minor amounts of Zn, even though there should be an abundant Zn source (sphalerite) in the tailings (Eilu *et al.*, 2012). The seepages of the Ylöjärvi porphyry deposit were distinct from the other seepages and had high As, U, and Co content originating from the weathering of arsenopyrite (As), uraninite (U) and presumably Co-bearing Fe-sulfides (Co). The seepages of the Fe deposits (Raajärvi and Rautuvaara) had in general very low contents of trace metals (Figure 4). The highest concentrations of Fe and SO_4 in the seepage waters occurred at the sites with the lowest pH and highest metal content, i.e. the sedimentary exhalative Cu deposit of Hammaslahti, Cu-rich VMS deposits of Orijärvi and Aijala, and the magmatic Ni deposit of Laukunkangas, indicating extensive sulfide oxidation in the tailings (Figures 2 and 4).

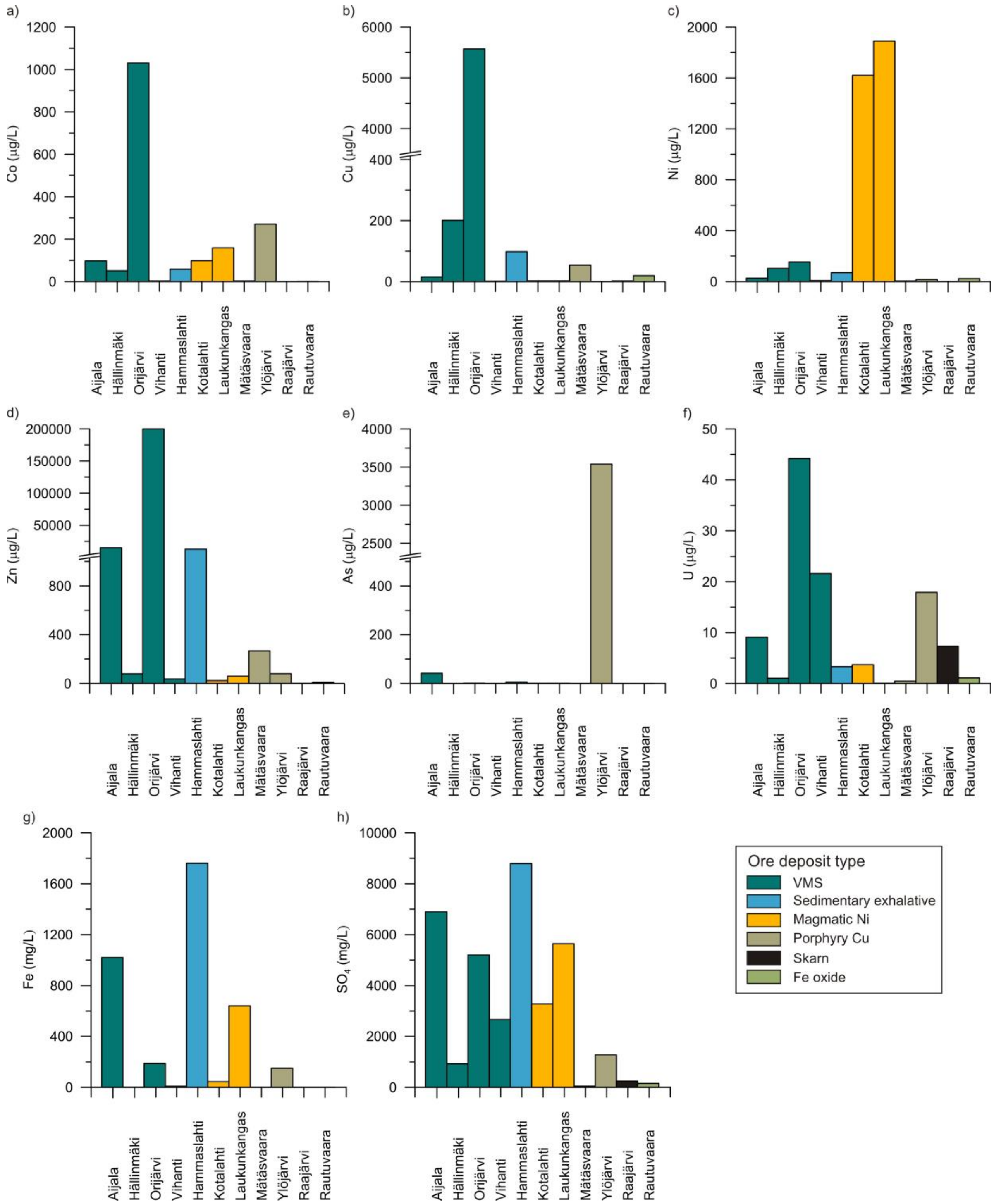

Figure 4 Concentrations of a) Co, b) Cu, c) Ni, d) Zn, e) As, f) U, g) Fe and h) SO_4 in the tailings seepage waters

The influence of the weathering period of the tailings on the drainage trace metal content was especially well seen in the seepage quality of the Orijärvi and Aijala sites. At these sites, the highest metal contents were measured and the time period after the cessation of tailings disposal was the longest (> 40 years, Table 1). In particular, seepage waters of the Orijärvi tailings showed high concentrations of Cu, Co, Zn and U (Figure 4). High metal loadings at Orijärvi can also be related to the undeveloped processing methods of the ore leaving notable amounts of valuable metals in the tailings, since the mine operated in the early 20th century (Table 1).

Influence of the tailings covers

Covering of the tailings facility appeared to be a less important factor than the deposit type in controlling the seepage quality. Hammaslahti and Laukunkangas tailings facilities were covered in the 1990's and 2000's with less than a half a meter thick layer of till and Vihanti tailings with a 5 cm layer of peat. Despite this, the tailings facilities of the Hammaslahti and Laukunkangas generated seepages with some of the highest metal contents and lowest pH values (Figure 2). The applied cover structures were only thin soil covers that are not regarded as sufficient to prevent infiltration of rainwater into the tailings and to prevent the oxidation of sulfide minerals in the tailings (INAP 2009). Presumably the primary aim of the covers has been to prevent the dusting and to promote the spreading of vegetation. In addition, the covering of the tailings was made only after some years from the cessation of the tailings disposal. The delay in the covering of sulfide tailings contributes to the onset of sulfide oxidation and AMD production in tailings (*e.g.* Heikkinen 2009).

CONCLUSION

The tailings seepage water data collected from eleven closed mine sites in Finland, representing six different genetic ore types, showed that the ore type can be a starting point for the prediction of the future drainage quality of the tailings at new mine sites. Data indicated that especially tailings from processing of volcanogenic massive sulfide Cu deposits and sedimentary exhalative Cu deposits are susceptible to produce high-metal (*e.g.* Cu, Zn), high-acid drainage waters containing also high SO_4 and Fe concentrations. In addition, tailings effluents of certain types of magmatic Ni deposit may also be highly acidic with high metal (particularly Ni), SO_4 and Fe contents, whereas the Fe deposits seemed to be the least problematic concerning the tailings water quality. Tailings seepages from porphyry deposits may contain high levels of As, U and Co.

Covering of the tailings had little or no effect on the quality of the tailings seepage waters, because of the delay in the covering and too thin cover layers. The covers were made in the 1990's and in 2000's and apparently the primary aim was then to prevent dusting and promote the spreading of vegetation.

These types of data are valuable in defining the requirements for water treatment and tailings disposal at new mine sites. However, a larger data set and more detailed descriptions of the ore types are needed to better define the relationship between the seepage quality and the ore type. In a future study, the data set will be enhanced by collecting tailings seepage water data also from operating mine sites in Finland.

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