Capability of natural and constructed wetlands to mitigate acidic leakage from closed mine waste facilities – cases in Eastern Finland[1]

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ABSTRACT

The performance of a constructed passive system and a natural wetland in closed mine areas in eastern Finland was evaluated on the basis of water chemistry, i.e. pH, pe (Eh) and concentrations of S, Fe, Mn, Al, Zn, Cu, Ni, Co. The constructed passive system, which operated for 1.2 years, effectively removed Fe by direct precipitation as Fe oxyhydroxides. However, S, Al and trace metals were removed to a lesser extent. In the natural passive system, metals and sulfur were removed first by co-precipitation with iron in organo-Fe oxyhydroxides in a ditch. The final purification was, however, based on degradation of the colloidal precipitates by microbial reduction in the sediment layers of the wetland and subsequently by precipitation as sulfides and complexation with organic matter. Despite of the short operation time, the pore water data from the peat-limestone-peat substrate of the constructed wetland pond effectively generated more alkalinity via carbonate rock dissolution and sulfate reduction than sulfate reduction alone in the organic substrate of the natural wetland. Moreover, the recovery of acid generating tailings with alkaline (magnesite-rich) tailings had the greatest impact on leachate purification in the short-term, compared with the passive treatment system.

Key Words: wetland, mitigation, ARD, closed mines, iron, sulfur, aluminum, trace metals

INTRODUCTION

The passive treatment of mine effluents is proposed as an inexpensive treatment alternative to chemical neutralization and precipitation of metals and sulfate. Alternative passive methods include wetlands, limestone drains and channels, bioreactors, reactive barriers, and leach beds (PIRAMID Consortium, 2003, Champagne et al., 2005). For the treatment of acidic waters, generating alkaline conditions (raising pH) through sulfate reduction is the first priority, since the subsequent removal of metals is more effective at a higher than lower pH (Walton-Day, 2003). An organic substrate like peat or compost, strips oxygen from the water and traps sulfate-reducing bacteria, and therefore can be used to develop sulfate reducing conditions (Frostman, 1993). Moreover, limestone in the collecting drains and/or in the substrate of the pond further encourages the generation of alkalinity (PIRAMID Consortium, 2003). The removal of metals and S is based on the precipitation of metal sulfides in the presence of sulfate reducing bacteria and/or adsorption of organic matter and iron oxyhydroxides.

The objective of this study is to examine the removal of metals and sulfate in a constructed passive system at Luikonlahti and compare this with a naturally formed wetland at Otravaara, eastern Finland. Both wetland systems included aerobic and anaerobic processes and it was assumed that water purification was similar. The effectiveness of the metal removal is estimated by comparing metal concentrations in influent and effluent waters of the collecting

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[1] Paper presented at Securing the Future and 8th ICARD, June 23-26, 2009, Skellefteå, Sweden.

drains and wetland ponds. Changes in the concentrations were assessed on the basis of pH-Eh condition.

SITE DESCRIPTION

The mean annual rainfall in the study areas is between 600 to 700 mm, and there is a distinctive seasonal variation in precipitation, with the driest season in spring (March-April) and the heaviest rains in late spring and summer (May-July). Below zero temperatures with snow cover are generally from December to April. In 2008, May was exceptionally dry and heavy rains occurred in July and August.

Luikonlahti study site

The Luikonlahti tailings impoundment was constructed in 1968 for the disposal of tailings from the exploitation of the Luikonlahti massive Cu-Zn-Co-Ni sulphide ore (Eskelinen et al., 1983). The flotation plant and facility was used for the talc production starting in 1979 and continued from 1984 up to 2006 after metal production ceased in 1983. In addition to talc, a nickel concentrate was produced from the talc ore when feasible.

The Luikonlahti tailings impoundment in the drained Lake Petkellampi covers ca. 27 ha in formerly glaciated terrain with silt and till, and contains approximately 6 Mt of sulfide tailings overlain by 2.5 Mt of magnesite-rich tailings from talc production (Räisänen and Juntunen, 2004). Due to waterproof base and the gradient of the lake valley, seepage waters mainly discharge at two sites beside the western dam (Heikkinen et al., 2009). The major seepage (‘Toe seepage’) is located at the toe of the primary dam and is constructed of till and waste rock, and the other seepage (‘Upper seepage’) occurs below the upper section of the dam initially raised with sulfide tailings (Figure 1).

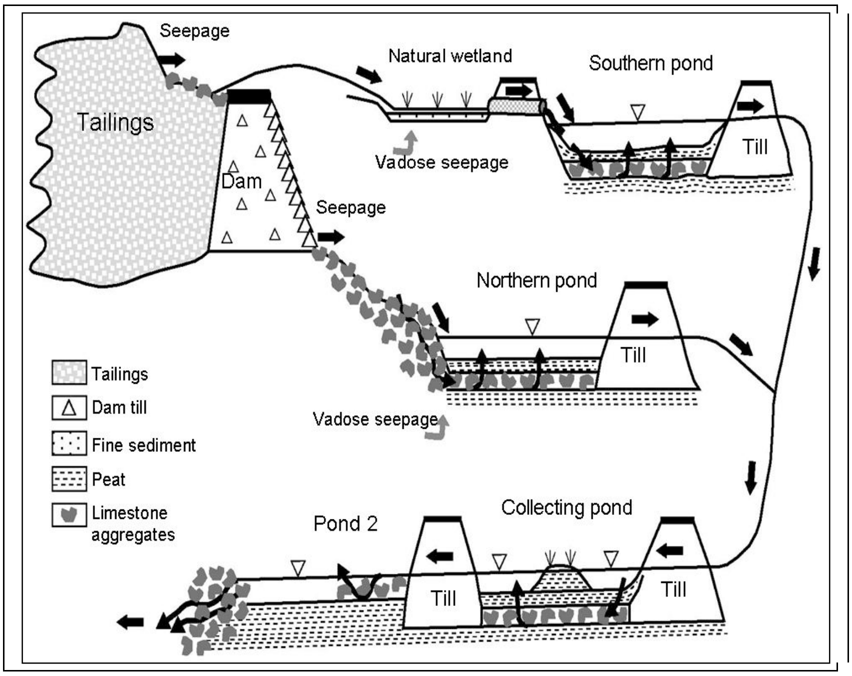

Figure 1. The sketch of the Suursuo passive treatment system, Luikonlahti, eastern Finland.

The decommissioning of the facility included the thickening of the magnesite-rich tailings and flooding to retard the sulfide oxidation (Räisänen and Juntunen, 2004). The purification of the leachate from the impoundment was based on a peat-limestone-based wetland-type passive system. In summer 2007, four wetland ponds (aerobic-anaerobic) were constructed in the Suursuo bog, west of the facility. A sketch of the water flow in the passive system and base structure of the ponds is presented in Figure 1. Seepage waters are collected via open limestone drains (OLD) into wetland ponds, the upper seepage flows first into a natural wetland (minerogenic fen) and then via a pipe below the road into the southern wetland pond, while the toe seepage flows into the northern wetland pond (Figure 1). The water runs from the pond into another via an open outlet in the middle of the dike, which is constructed of fine-grained till. In the end, the water from wetland pond 2 flows through a bank of limestone aggregates into a ditch and then into a pond for pumping back into the facility.

The thickness of the topmost peat layer and the mid limestone layer is ca. 30 cm, and that the thickness of the underlying peat layer is 50 cm or more in the southern part of the bog (max 3-4 m in the southern pond). Collecting drains for seepage waters and the bank of pond 2 for final outflow are filled with limestone aggregates with a diameter of 10-15 cm, whereas the diameter of the aggregates in the interlayer of the pond base is less than 10 cm. Limestone aggregates mainly consist of Proterozoic calcite-rich metasedimentary rocks with minor dolomite.

Otravaara study case

The Otravaara pyrite mine operated between 1919 and 1923. A 0.3 ha waste rock pile is located beside a small open pit on the slope of a moraine hill. Waste rocks consist of sulfide-bearing metavolcanic and metasedimentary rocks. Mine operations (on-site sorting) and acidic meltwaters from the highly weathered pile have caused forest death over an area of 0.5 ha on the hillside flanking the pile (Räisänen et al., 2001).

The Figure 2 depicts water flow in the passive treatment system and the structure of the main wetland. Initially, leachate from the waste rock pile seeped via a vadose zone of the dieback area into a ditch that bordered the mine area in north and discharged into a minerogenic fen (see main wetland in Figure 2).

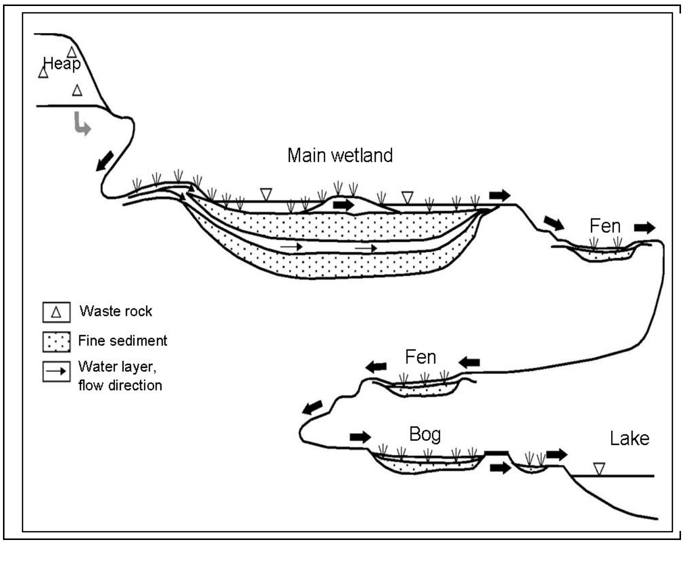

Figure 2. Sketch of the passive treatment system at Otravaara, eastern Finland. Black arrows show the flow path via small fens and a bog to the lake.

The main wetland was formed at the side of the former creek plateau due to the mineral material eroded from the mine area. In the mid 1990's, the dieback area was drained resulting in an increased sediment load into the fen, which developed into a water meadow-like wetland. In summer 2004, the landowner filled the drains in the dieback area. Since then, the leachate from the pile again seeps in the vadose zone of the deteriorated area and flows out into the ditch (at the original site). The mixing of acidic waters from the mine area and humus-rich waters from a bog results in Fe precipitation. The sediment drifting in the creek consists of organo-Fe precipitates, mainly schwertmannite and goethite, which effectively adsorb metals and S (Kumpulainen et al., 2007). At present, the precipitation is not as voluminous as it was during the drainage of the dieback area.

MATERIALS AND METHODS

In Luikonlahti, open water and pore water samples from the wetland's ponds were collected for the first time immediately after construction in August and October (only open water) 2007 and then in May and September 2008. The newest seepage water samples presented in this study were taken from the upper seepage in May 2007 (dried in autumn 2007) and from the toe seepage in May and September 2008. Comparative seepage data from 2003 represent the leachate before decommissioning (Heikkinen et al., 2009).

Samples from open waters were taken with a limnos sampler and from outlets of the ponds and from seepages with syringes into 500 ml polyethylene bottles. For pore water sampling, thin plastic tubes (PVC) with supporting structures were installed in the topmost peat layer and underlying limestone aggregates layer during the pond construction. The sample from tubes was taken by using a syringe and stored in 100 ml polyethylene bottles.

Water samples for dissolved elements were filtered through 0.45 μm filters into polyethylene bottles in the field. The filtered samples were immediately acidified with concentrated Suprapur® HNO_3 (0.5 ml acid addition per 100 ml sample). In 2003, seepage samples were filtered in the laboratory within 1-2 days after sampling. Similarly, the filters were acidified with concentrated Suprapur® HNO_3. The pH, redox (calculated as pe) and oxygen concentration were recorded with a portable multi-parameter field meter (YSI 556). The redox probe was a platinum electrode. Concentrations of the 30 elements were determined using the ICP-AES/MS technique. Analyses were done in the FINAS-accredited laboratory of Labtium Oy (former Geolaboratory of the GSF). This study presents data only for S, Fe, Mn, Al, Zn, Cu, Ni and Co.

The water quality of influents and effluents in the Otravaara passive system was monitored 3-4 times per year during the unfrozen season (May and/or June, August, October) between 2000-2005. The element concentrations were analyzed with ICP-AES/MS from water samples that were filtered and acidified with concentrated Suprapur® HNO_3 in the laboratory 1-2 days after sampling in the period from 2000 to 2004, and in the field immediately after the sampling in 2005. This study presents mean concentrations of the above mentioned elements for 2000-2004 and separately for 2005. The pH was measured with a WTW pH330/SET field meter and the oxygen concentration was measured with a WTW oxi330/SET field meter. In 2005, the pH and redox potential was measured with a WTW 340i/SET field meter equipped with a platinum electrode.

Unfiltered 500 ml samples for analyses of the total solid concentration were taken from five pond outlets at Luikolahti in May and September 2008 and from three sites (collecting ditch, wetland, final effluent) at Otravaara in 2005. Measurements were taken using the SFS-EN872 method in the FINAS-accredited laboratory of Savo-Karjalan Ympäristötutkimus Oy.

RESULTS AND DISCUSSSION

Impact of the remediated tailings facility on seepage water quality, Luikonlahti

In 2003, before the remediation of the tailings facility, effluents from the tailings were mainly net acidic with a high trace metal (Mn 32.4 mg/l, Zn 13.7 mg/l, Ni 1.5 mg/l, Cu 1.9 mg/l, Co 1.0 mg/l; on average) and S, Fe and Al content (S 1250 mg/l, Fe 890 mg/l, Al 17.6 mg/l; on average, Table 1). The pH <3 at the upper seepage point in 2003 indicates that sulfide mineral oxidation was extensive before the decommissioning (Heikkinen et al. 2009).

In 2006, the thickening of the magnesite-rich tailings overlying the sulfide tailings quickly raised the pH of the seepage up to 6 (Heikkinen et al., 2009). The flooding during the thickening in 2006 was, however, temporary and the water table gradually decreased and remained at the level of the main dam surface at the end of 2008 (Unpublished monitoring data). As a result, the upper seepage dried in autumn 2007 and the seepage predominantly shifted into the toe section of the primary dam. Despite the water table drop, pH slightly raised over six from 2007 to 2008, and the pe of the toe seepage was low indicating less oxidizing conditions in the tailings and dam (Table 1). Obviously, the thickening of magnesite tailings has reduced vertical oxygen diffusion and amplified the alkalinity of the vadose water. Consequently, sulfide oxidation was retarded and/or the buffering capacity was improved.

A total reduction in concentrations of 99.6%, 97%, 96%, 95%, 92%, 88%, 86% and 70% from the upper seepage in 2003 to 2007 was measured for Al, Fe, Mn, Zn, Co, Ni, Cu and S, respectively. The change in the toe seepage content was minor despite the pH increase. Concentrations of Al and Cu decreased by over 99 %, and for Mn and Zn, the decrease was 87% and 82%, respectively. For Ni and Co, however, the reduction was much lower, about 60% and 40%, respectively. The decrease in S concentration was surprisingly small (<10%). However, the concentration of Fe in the toe seepage increased almost by one-third. Obviously, a shift to a more reducing phase has caused the dissolution of Fe oxyhydroxides precipitated in shallow tailings before the remediation (Ribet et al., 1995).

Efficiency of the constructed passive system, Luikonlahti

The removal of Fe in the constructed wetland system was overall more marked than that of S or the other metals (Table 1). In September 2008, the total decrease for Fe was 96% from the mean Fe concentration of the toe seepage (i.e. from the main contamination source). The reduction for S was only 30% while the reduction varied between 12% and 27% for the trace elements (Mn, Zn, Ni, Co). As seen in Table 1, the removal of Fe and several other metals was more effective in October 2007 (two months after the construction) and in May 2008 than in September 2008. Obviously, the difference is caused by the seasonal variation in precipitation. Autumn 2007 and spring 2008 were exceptionally dry (less rainfall) compared to July and August 2008. In September 2008, the ponds were almost full, which promoted quick flow-through and also retarded natural spreading of vegetation.

Iron oxidation and the precipitation of Fe oxyhydroxides seem to be the main mechanism for Fe removal in the natural wetland, northern pond and collecting pond. According to field observations, Fe precipitation was most abundant in the northern pond and caused the increase of solids in the effluent in September 2008 (Table 2). The Fe precipitation was followed by a drop in pH (<4) and consequently, concentrations of Al and the trace metals slightly increased (Table 2). Desorption of cationic metals is a result of the acid pH as the net surface charge of the Fe precipitates becomes more positive (Dzombak and Morel, 1990).

Table 1. pH, pe (Eh) and concentrations of O_2, S, Fe, Mn, Al, Zn, Cu, Ni and Co of the upper and toe seepage before and after the remediation of the tailings facility, and comparative data from the effluent of the end pond 2 of the passive system, Luikonlahti, Eastern Finland.

	Upper seepage		Toe seepage				Effluent from the Pond 2		
	Before	After	Before		After				
	May03	May07†	May03	Oct07	May08	Sept08	Oct07	May08	Sept08
pH	2.7	6.0	4.5	6.1	6.5	6.3	5.2	5.3	4.2
pe	-	-0.2	-	1.5	-0.1	0.1	2.0	4.1	6.4
	----- mg/l -----		-------------- mg/l -------------				-------- mg/l ---------		
O_2	1.4	-	3.4	2.6	3.6	3.1	11.2	7.9	8.4
S	1900	389	600	501	418	548	325	335	372
Fe	1730	30.9	50.6	25.8	38.0	88.2	0.24	1.68	3.57
Mn	49.3	0.92	15.6	1.38	1.01	1.98	1.03	0.99	1.45
Al	29.6	0.07	5.66	0.03	0.01	0.02	0.38	0.21	0.63
Zn	21.2	0.52	6.10	1.19	0.53	1.02	0.61	0.39	0.74
Cu	3.73	0.35	0.14	0.0002	0.001	0.001	0.004	0.01	0.01
Ni	1.94	0.16	0.97	0.33	0.14	0.37	0.24	0.18	0.28
Co	1.86	0.10	0.20	0.07	0.03	0.11	0.07	0.06	0.10

† The upper seepage dried in autumn 2007

- Not measured

Table 2. pH and pe (Eh) values and concentrations of solids, O_2, S, Fe, Mn, Al, Zn, Cu, Ni and Co of influents and effluents in the constructed passive system in September 2008, Luikonlahti, Eastern Finland.

	Southern pond		Collecting pond		Pond 2
	Influent*	Effluent	Influent**	Effluent	Effluent
pH	3.8	5.5	3.5	3.5	4.2
pe	7.4	4.5	7.1	7.5	6.4
	------ mg/l ------		------ mg/l ------		-- mg/l --
solids	12	5.0	42	23	17
O_2	5.3	10.3	9.9	9.5	8.4
S	313	234	483	412	372
Fe	4.47	0.55	25.6	10.3	3.57
Mn	1.38	0.97	1.65	1.61	1.45
Al	8.32	0.22	1.00	0.93	0.63
Zn	0.19	0.19	1.78	1.29	0.74
Cu	0.02	0.01	0.02	0.02	0.01
Ni	0.44	0.23	0.40	0.35	0.28
Co	0.08	0.06	0.16	0.13	0.10

*Outflow from the natural wetland

**Outflow from the northern pond

In comparison, the effluent from the southern pond contained minor amounts of soluble Fe and solids, indicating better removal (Table 2). In addition, the pH was moderate (>5) and concentrations of Al and trace metals were lower in the southern pond than in the northern and collecting ponds from August 2007 to September 2008. The retention of S was, however, small but better than in the northern pond. Overall, the performance of the substrate was more favorable for sulfate and Fe reduction in the southern than in the northern pond. According Todorova et al. (2005), the optimum for pyrite formation is between pH 6 and 9 and pe of -3 and -6. This is true for the pore water in the limestone aggregate layer but not for the overlying peat layer of the southern pond (Table 3). In the northern pond, the pH values of the pore water in both layers were in the optimum range, but pe values were above the range (Table 3). The concentration of S was, however, great in the pore waters of both pond's substrate. Therefore, sulfate reduction and precipitation of Fe and trace metal sulfide have not reached an equilibrium in 1.2 years' of passive treatment. This argument is based on the fact that there no H_2S smell was observed during pore water sampling. Sparse vegetation suggests minor productivity of decomposed organic matter and therefore, a low activity of living organisms.

In the collecting pond, concentrations of S, Fe, Mn, Al, Zn, Ni and Co remained high and were only slightly reduced despite mixing with the less contaminated effluent from the southern pond (Table 2). However, the effluent from the collecting pond continued to improve in pond 2 before the outflow. The pH rose from 3.5 to 4.2 and the concentration of soluble Fe decreased about 65% (Table 2). A minor reduction could be seen in the quantity of solids and concentrations of Al and trace elements.

Table 3. pH and pe (Eh) values and O_2, S, Fe, Mn, Al, Zn, Cu, Ni and Co concentrations of the pore water in the top peat and mid-limestone aggregates layer of the constructed southern and northern ponds, Luikonlahti, Eastern Finland.

	Southern pond				Northern pond			
	Peat (top)		Limestone (mid)		Peat (top)		Limestone (mid)	
	Aug07	Sept08	Aug07	Sept08	Aug07	Sept08	Aug07	Sept08
pH	6.7	6.8	6.5	7.0	6.7	6.3	6.7	6.6
pe	-0.1	-2.1	-1.2	-5.2	-0.6	-0.4	-1.4	-1.3
	------------- mg/l --------------				------------- mg/l ---------------			
O_2	4.5	2.1	1.9	0.6	3.5	4.5	2.2	2.6
S	315	180	358	396	458	696	446	667
Fe	72.0	1.21	197	0.13	67.1	150	85.8	154
Mn	1.36	0.89	1.66	0.49	1.46	2.36	1.42	2.48
Al	0.06	0.03	0.13	0.02	0.03	0.10	0.05	<0.02
Zn	0.04	0.02	0.09	0.03	0.04	0.04	0.05	0.04
Cu	------------ <0.0005 -----------				------------ <0.0005 -----------			
Ni	0.07	0.01	0.27	0.01	0.03	0.15	0.04	0.06
Co	0.03	0.001	0.06	0.001	0.03	0.18	0.03	0.07

Efficiency of the natural passive system, Otravaara

The precipitation of organo-Fe oxyhydroxides in the tributary junction and downstream ditch was the first stage to the removal of metals and S at Otravaara (Räisänen et al., 2001). During 2000 and 2004, the mean total reduction from seepage concentrations in the ditch was 97% for Fe, 96% for Al, Zn, Cu, Ni and Co, 95% for S and 94% for Mn. The removal increased ca 3% in the main wetland and to a minor (≤1%) extent in downstream fens. The exception was Mn, which had somewhat higher concentrations in the effluent that flowed into the lake than in the surface water of the main wetland (Table 4).

Table 4. Means of pH and pe (Eh), and mean concentrations of solids, O_2, S, Fe, Mn, Al, Zn, Cu, Ni and Co of seepage from the waste rock heap and surface waters of the passive system in 2000-2004 and 2005, Otravaara, Eastern Finland.

	Seepage water		Ditch (collecting)		Wetland		Effluent	
	2000-2004†	2005‡	2000-2004†	2005‡	2000-2004†	2005‡	2000-2004†	2005‡
pH	2.2	2.6	2.9	2.9	3.4	3.4	3.6	3.6
pe	-	8.9	-	8.4	-	7.3	-	6.8
	mg/l							
O_2	3.3	4.7	4.3	4.0	4.8	5.2	4.0	4.1
Solids	-	-	-	30.7	-	13.0	-	4.5
S	2813	1118	132	378	60.7	40.2	36.0	28.6
Fe	2567	962	71.2	220	45.5	16.3	9.4	12.4
Mn	11.0	3.74	0.61	2.17	0.29	0.25	0.46	0.35
Al	307	81.5	12	57.1	3.9	5.0	3.2	1.8
Zn	9.96	2.46	0.37	1.15	0.10	0.10	0.06	0.05
Cu	3.10	1.03	0.11	0.37	0.02	0.02	0.01	0.01
Ni	1.55	0.41	0.06	0.24	0.02	0.03	0.02	0.02
Co	1.08	0.46	0.04	0.20	0.02	0.01	0.02	0.01

† 15 samples filtered in the laboratory

‡ 3 seepage water samples and 4 surface water samples filtered in the field

\- Not measured

In 2005, after the ditch bordering the waste rock heap was filled, the seepage concentrations of Zn, Al and Ni decreased from 73% to 75% (least for Zn), and those of Co, S, Fe, Mn and Cu from 57% to 67% (least for Co). The pH remained acidic. The retention deteriorated 1% to 2.5% in 2005 from that in 2000-2004, which is within the accuracy and precision of analytics and sampling. An exception to this is that the removal of Mn decreased ca. 5%. Nevertheless, the actual mean concentrations in the effluent flowing into the lake were lower in 2005 than in 2000 and 2004 (Table 4). The only exception was Fe, which had a somewhat greater mean concentration in the effluent in 2005 than in 2000 and 2004.

The retention of metals and S in the natural passive system is mainly based on the adsorption and fixation on organo-Fe oxyhydroxides, which precipitates in the tributary junction after the mixing of the humus-rich water with the acidic seepage (Kumpulainen et al., 2007). In the wetland, the precipitates were decomposed in the anaerobic layers and, consequently, released Fe and sulfate that were then reduced and reprecipitated as secondary Fe sulfides (Räisänen, 2003). However, the sulfate reducing within the wetland sediments has not raised the pH in the surface water running over the wetland and downstream fens (Table 4). Räisänen et al. (2001) reported that the hydrolysis of Al is stabilizing the pH at a constant level. Obviously, the precipitation of Fe oxyhydroxides is also active in surface waters, which maintains the acid pH.

CONCLUSIONS

Two years after the facility's closure, the thickening of the magnesite-rich tailings had the greater impact on leachate purification than the constructed passive treatment system at Luikonlahti. The net acidic seepage from the oxidized sulfide tailings changed to net alkaline and consequently, concentrations of metals reduced from 40% to 99%, depending on the seepage point. Moreover, the seepage changed from oxidative to reductive (negative pe) thus maintaining Fe dissolution. The change results in the quick oxidation of Fe and precipitation of Fe oxyhydroxides in the constructed pond that received the most seepage in autumn 2008. As a result, the removal of Fe was greater than that of S, Al and trace metals.

In the natural passive system at Otravaara, the first stage for treatment was the precipitation of Fe oxyhydroxides fixed with organic matter. Despite of low pH, this type of Fe precipitate had better adsorption capacity to retain S, Al and trace metals than that formed in the constructed system at Luikonlahti. In the natural passive system, the final purification was based on degradation of the colloidal precipitates by microbial reduction in the sediment layers of the wetland and subsequently by precipitation as sulfides and complexation with organic matter. This stage was insufficiently achieved in the constructed passive system. However, alkalinity was more effectively generated via carbonate rock dissolution and sulfate reduction in the peat-limestone-peat substrate of the constructed ponds than via sulfate reduction alone in the organic substrate of the natural wetland.

ACKNOWLEDGEMENTS

The author thanks Mondo Minerals B.V. Branch of Finland and Finn Nickel Oy for providing the opportunity to study water quality in the constructed passive system at Luikonlahti. Funding in 2007 was provided by Mondo Minerals Oy (former name of the company) and in 2008 Finn Nickel Oy and the Geological Survey of Finland. The Otravaara study was financed by the Geological Survey of Finland.

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