Barrier system for treatment of heavy metal drainage at Ranstad, Sweden

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Abstract Alum shale was mined and processed (leaching with sulphuric acid for uranium recovery) at Ranstad, Sweden, during 1965—69, and the shale residues were deposited on site. A barrier system for treatment of leachates from the deposit was constructed in 1999: (1) Sedimentation pond (2) pre-treatment filter, and (3) final filter in sequence. Iron, Al and Mn were precipitated in section (2). Retention of other elements (As, Cr, Co, Ni, Zn and U) is attributed to adsorption by precipitated iron and components in section (3). More than 90% of the Fe and As and 50% of the Cr, Cd, Zn and U were removed from the leachates.

Key Words filters, uranium, iron, peat

Introduction

Drainage water from mine waste has been recognized as an environmental problem for a long time. With the drainage water, metals are released to the environment. To decrease the release of metals mine drainage can be treated. A conventional way of treating metal containing drainage is chemical treatment (i.e. liming). However, conventional treatment systems like liming often have high maintenance requirements and energy demands, and cause voluminous production of sludge. Recently, the interest in passive barrier systems have become popular as complement to the existing technology (Powell et al. 1998; Smyth et al. 2001). The construction costs for passive treatment systems can be large, but well built they are considered more cost-effective, as maintenance and energy costs are reduced.

In Ranstad, southwest Sweden, tailings after alum shale mining for uranium extraction (leaching using sulphuric acid) were deposited during the 1960s. In the 1990s an extensive restoration of the area was carried out. Lime was added to the tailings and a final cover consisting of till was installed. Production of drainage and metal concentrations decreased while pH increased from 4 to around 7. However, the remaining drainage is still treated in a chemical treatment plant. In 1999 a pilot-scale passive treatment system based on an aeration step, sedimentation pond, sludge separator, pre-treatment filter and filter step (filter sand and peat) were constructed.

The aim of the present study is to investigate the operation of a passive treatment system and the design of a filter, in order to determine the efficiency of such system.

Methods

Barrier design

An aeration step, a sedimentation pond, a sludge separator, a pre-treatment filter and a filter step (filter sand followed by peat) constitute the treatment system (fig. 1). Drainage water was pumped from the ditch surrounding the tailings to the sedimentation pond (10 m) via the aeration steps (flow rate 0.1—0.5 L/s) to remove the high concentrations of iron (tab. 1). After a hold up time of about 3.5—5 days in the sedimentation pond the water moved through the system by gravity to the filter furrow, via the sludge separator and the pre-treatment filter (coarse gravel, 16—32 mm).

The filter sections were constructed as a furrow, divided into two sections, 4 respectively 6 m (1 m depth, 1 m wide), with impermeable concrete walls. In both sections the bottom was covered with 0.1 m thick layer of coarse gravel serving as the drainage layer. In the present study, filter sand (quartz 0.2—3 mm) and peat (black peat, H_6-H_8, irregular shape 6—18 mm) was chosen as filter materials, based on experiences from conventional water treatment. Filter sand was used as a mechanical filter and the peat for sorption and ion exchange.

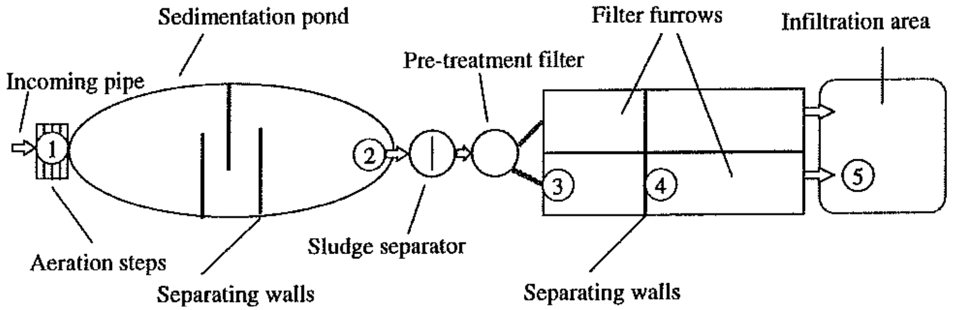

Figure 1 *The barrier system. Numbers indicate water sampling locations; (1) after aeration, (2) after sedimentation, (3) after the sludge separator and pre-treatment filter, (4) after the sand filter and (5) after the peat filter*

Sampling

From the first day of operation, water samples (unfiltered) were taken weekly at 4—5 sampling locations (fig. 1). Water samples for metal analysis were acidified using concentrated nitric acid. Oxygen and water temperature were measured weekly in the field using an oxygen probe (WTW OXI 330).

Analytical methods

Water samples have been analysed for metals, as well as physical chemical parameters. ICP-OES was used to determine concentrations of major elements (Ca, K, Mg, Na and Fe) while concentrations of trace elements were determined using ICP-MS (As, Cd, Co, Mn, Ni, Pb, Zn and U). Sulphate and chloride were determined by ion chromatography. Alkalinity was determined by titration.

Results and discussion

The treatment system was run for 14 months (september 2000 to november 2001) with only minor interruptions. Major parameters were barely changed through the treatment system (sampling locations 1—5; fig. 1). On average alkalinity decreased from 224 to 135 mg/L HCO_3^-, probably due to the production of protons when ferric iron was hydrolysed.

Sulphate (around 900 mg/L), chloride (around 8 mg/L), calcium (around 350 mg/L), magnesium (around 50 mg/L), sodium (around 11 mg/L) and potassium (around 35 mg/L) remained fairly constant through the system. Oxygen concentrations increased slightly after the aeration step (from 6 to 8 mg/L).

Table 1 *Metal concentrations (µg/L) in the deposit area, Ranstad: Inlet to and outlet from the barrier, reduction (%) through the barrier system and local background levels. Averages for the 14 month monitoring period*

	Drainage (inlet)	Drainage (outlet)	Reduction (%)	Background (local)	Quality goal
pH	7.9			7.8	
As	15	0.6	96	0.5	2.0
Cd	0.46	0.16	65	<0.02	-
Co	14	8	43	0.26	2.0
Fe	38 000	1 450	96	510	1 000
Mn	4 100	2 400	41	140	-
Ni	150	39	74	1.2	10
Pb	5.2	0.2	96	0.1	0.7
Zn	75	22	70	2.0	8
U	38	9.7	74	2.1	11

Calcium concentrations were fairly high in the system due to the addition of limestone to the tailings residue prior to covering. This is also noted in the high pH (tab. 1). Geochemical calculations also indicate equilibrium with calcite ($CaCO_3$) and gypsum ($CaSO_4$) in the water. Trace metal concentrations prior to the treatment system compared to average concentrations after the system can be found in tab. 1.

Iron and arsenic

Oxidation, hydrolysis and precipitation of iron as (hydr)oxides are expected to be an important removal mechanism for trace elements (Lee et al. 2002). Illustration of decreased iron concentrations through the system can be found in fig. 2. Iron concentrations are reduced by 96% through the system (tab. 1), even though the reduction is not as high as expected in the initial parts of the system (aeration pond and sludge separator). A large part of iron is retained in the filter sand, probably due to physical filtering of precipitated iron(hydr)oxides. At the end of the operation period slightly increasing iron concentrations were observed after the peat filter, indicating possible reductive dissolution of iron(hydr)oxides.

Arsenic concentrations (fig. 2) are found to be co-varied with the iron concentrations through the system indicating a significant impact on the arsenic concentrations. This is most likely an effect of co-precipitation and some adsorption as the high concentrations of iron precipitates. This is partly confirmed by the fact that the major decrease in arsenic happens prior to the peat filter sections.

Divalent cations

Typical cations such as Cd, Co, Ni, Pb, Zn and Mn decreased (between 41—96%) through sorption at a pH above 7. This is confirmed by the fact that the major part of the decrease is in the sand filter sections indicating co-precipitation and sorption onto iron(hydr)oxides retained in the sand (zinc is found as an example in fig. 3). However, the decrease is lower than expected (except for lead) considering the high pH and the high concentrations of precipitating iron.

Uranium

Uranium concentrations decreased fairly evenly through the treatment system (fig. 3). Uranium also showed some co-variation with iron indicating sorption as the major removal mechanism. Uranium occurs as UO_2^{2+} in oxygenated waters and due to the high alkalinity in the system, uranium most likely exist as dissolved carbonate complexes ($UO_2(CO_3)_2^{2-}$ or $UO_2(CO_3)_3^{4-}$).

Iron(hydr)oxides are important for uranium sorption, but the adsorption decreases when the uranyl ions are complexed with carbonate (Duff and Amrheim 1996). This is also observed as the adsorption/removal of uranium is increased as the alkalinity decreases through the system. Removal of uranium happens between sampling locations 1 and 4 (fig. 3) indicating that the removal in the peat filter section is insignificant.

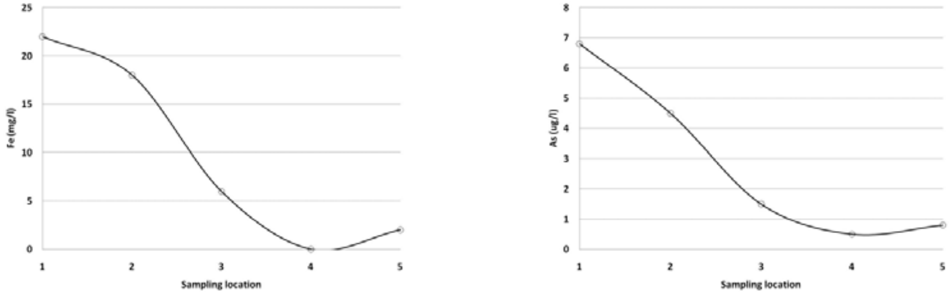

Figure 2 *Iron (mg/L) and arsenic (µg/L) concentrations (averages during the entire operation period September 2000-October 2001) through the treatment system steps. See fig. 1 for details around sampling locations*

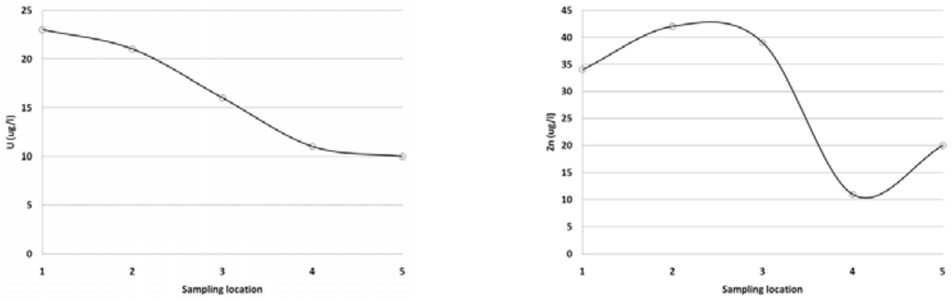

Figure 3 *Uranium and zinc (µg/L) concentrations (averages during the entire operation period September 2000-October 2001) through the treatment system steps. See fig. 1 for details around sampling locations*

Conclusions

In general, the results obtained indicate satisfactory trapping of metals during the operation of the system. About 95% of the total iron and arsenic were trapped in the treatment system. Further, about 70% of the total Zn, Ni, Cd and U content were retained in the treatment system, while manganese was removed about 20%.

The sedimentation pond, the pre-treatment filter and the sand filter functioned as the main iron traps in the treatment system. Furthermore, most of the trace metal retention in the treatment system seemed to be associated with the iron precipitates.

A more extensive retention of the iron before the final filter step is of importance, partly to avoid clogging of the filter surface, and partly to hinder reduction of ferric iron to ferrous iron if the redox potential is lowered in the peat filter section. This would lead to a reductive dissolution of the formed iron(hydr)oxides and a release of the adsorbed trace metals.

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References

Duff MC and Amrhein C (1996) Uranium(VI) adsorption on goethite and soil in carbonate solutions. Soil Sci Soc Am J 60: 1393—1400

Lee G, Bigham JM and Faure G (2002) Removal of trace metals by coprecipitation with Fe, Al and Mn from natural waters contaminated with acid mine drainage in the Ducktown Minig District, Tennessee. Appl Geochem 17: 569—581

Powell RM, Blowes DW, Gilham RW, Shcultz D, Sivavec T, Puls RW, Vogan JL, Powell PD and Landis R (1998) Permeable reactive barrier technologies for contaminant remediation. EPA/600/R-98/125

Smyth DJA, Blowes DW, Benner SG, and Ptacek CJ (2001) In situ treatment of metals-contaminated groundwater using permeable reactive barriers. In: International Conference on Mining and the Environment, Skellefteå, Sweden. Proceedings 2: 787—794