The Effectiveness of Using Sewage Sludge as a Sealing Layer on Sulphide-rich Mine Tailings: A Pilot-scale Experiment, Northern Sweden

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Abstract An evaluation of the effectiveness of using sewage sludge as a sealing layer to prevent oxygen diffusion to underlying tailings was conducted from 2001—2009 using pilot-scale cells in northern Sweden. The leachate drainage of an uncovered tailings control cell and a cell with a 0.25m sealing layer of sewage sludge was analysed from 2003—2009. An initial high concentration flush of dissolved elements occurred in both cells. Sulphur and heavy metal concentrations gradually declined in both cells but the differences in concentrations were several orders of magnitude lower for the sludge remediated cell. Cd-Fe-Zn concentrations in the control increased over time due to sulphide oxidation. The consumption of oxygen in the sewage sludge created reduced conditions in the underlying tailings. Elevated metal concentrations leached from the sludge into the tailings were subsequently retained in the tailings and were not elevated in the effluent leachate solution.

Key Words mine tailings, sulphide oxidation, pilot-scale experiment, sewage sludge, sealing layer

Introduction

A long-term, low maintenance remediation solution is desired to cover sulphide-bearing mine waste in order to prevent oxygen diffusion and any subsequent sulphide oxidation, mitigating against the formation of acid rock drainage (ARD). The construction of a composite cover design was first proposed for use in Sweden by New Boliden AB in the 1980's (Lundgren 1997). It comprises of a sealing layer that prevents oxygen diffusion to underlying waste. An overlying un-reactive protective layer protects against frost penetration, water runoff and erosion, which retains the sealing layer integrity. Barrier materials for the sealing layer contain low permeable materials such as high-clay containing till that are sourced from deposits in proximity to mine sites, and are often expensive to extract. In recent years there have been advances to assess possibilities in extending the barrier material range to include low cost waste alternatives. An estimated 210 000 tonnes of sewage sludge is produced each year in Sweden (Statistics 2008). Disposal of this waste is problematic. Using sewage sludge as a sealing layer on sulphide-bearing mine tailings is proposed. Co-disposal of this material with mine tailings solves two problems. This creates an attractive and low cost alternative for the industry. Sewage sludge has an advantage over conventional barriers as it is an organic cover that can consume oxygen (eq. 1) and reduce sulphate (eq. 2) which may promote metal sulphide precipitation (eq. 3) (Peppas, et al. 2000). However, reduction of the cover integrity over time has been identified (Ahlberg 2006) due to the oxidation and degradation of organic matter. It can contain readily leachable trace elements (Al, Mn, Cu, Ni, Zn) (Stehouwer, et al. 2006) and nitrate concentrations which exceed EU and Swedish EPA limits (Eriksson 2001). This study aims to evaluate the long-term effectiveness of using sewage sludge as a sealing layer to prevent oxygen diffusion to underlying tailings.

$$CH_2O + O_2 = H_2O + CO_2 \qquad (1)$$
$$2CH_2O + SO_4^{2-} + 2H^+ = 2H_2CO_3 + H_2S \qquad (2)$$
$$H_2S + Me^{2+} = MeS_2 + 2H^+ \qquad (3)$$
$$FeS_2 + {}^{15}/_{4}O_2 + {}^{7}/_{2}H_2O = Fe(OH)_3 + 2SO_4^{2-} + 4H^+ \qquad (4)$$
$$Ca(OH)_2 + 2H^+ = Ca^{2+} + 2H_2O \qquad (5)$$
$$Ca^{2+} + SO_4^{2-} + 2H_2O = CaSO_4 \cdot 2H_2O \qquad (6)$$
$$CaCO_3 + 2H^+ = Ca^{2+} + H_2CO_3 \qquad (7)$$

Equations 1—3 (Peppas, et al. 2000); Equations 4—7 (Lottermoser 2003)

Materials and Methods

The Georange Environmental Test Site at the Kristineberg Mine, northern Sweden was completed in 2002. Two insulated concrete cells ($5\times5\times3$ m^3) are filled with a 1m (Cell 3) and a 1.2m (Cell 6) layer of high sulphide-bearing (c. 50% pyrite, 2.5% pyrrhotite) tailings with calcite contents at 2.5%. They are sourced from the Kristineberg Mine and processed at the Boliden concentrator.

Sewage sludge was sourced from Lycksele municipal water treatment plant. Table 1 shows the chemical composition of the original sewage sludge and tailings. The sludge contained higher values of Al, P, Ba and Cr compared to the tailings, with elevated values of As, Fe, K, Mn, Cd, Cu, Ni and Zn. Profiles of the covers are seen in fig.1. Cell 3 has a 0.25m sealing layer of sewage sludge which is covered by a 0.3m drainage layer and a 1.2m protective layer of unspecified till. The tailings in cell 6 are uncovered and are used as a control. Both cells contain a 0.3m drainage layer at the base where a drainage probe collects the leachate from each cell (Cell 3: KC1; Cell 6: KC6). A separate lysimeter (KC3) in cell 3 collects the leachate 0.1m below the sewage sludge sealing layer (fig. 1). The leachate from the base drainage layer probes was sampled in the summer months of 2003—2006, and 2009. The lysimeter (KC3) was sampled in 2009. Analysis of dissolved (<0.22µm) elemental fractions occurred using 0.22µm nitrocellulose membrane filters. NO_3^-, SO_4^{2-}, and alkalinity were only sampled in 2009. All equipment was washed acid washed and rinsed in Milli-Q$^{©}$ water, except bottles used for anions and alkalinity. Blank analysis was conducted using Milli-Q$^{©}$ water. Measurements were conducted using a Mettler Toledo SevengoPro dissolved oxygen metre and a Mettler-Toledo pH electrode calibrated to pH4.01 and pH7.00 respectively. Analysis was made at ALS Scandinavia in Luleå. Elements Ca, K, Mg, Na, S, Si, Mn and Sr were analysed using ICP-AES. Hg was analysed using AFS. All other elements were analysed using ICP-SFMS. Precision was calculated to within 10%. Anions were determined using ion chromatography.

Results and Discussion

Dissolved Iron and sulphur

Sulphur decreases in both the sludge remediated cell and the control from 800 to 59mg/L (fig. 1A) and 800 to 489mg/L (fig.2A) respectively between 2003—2009. The initial elevated concentrations represent a flush effect from the tailings due to sulphate contained in the residues from mineral processing. However, there is a difference in magnitude of declining concentrations between the two cells. In the control, the sustained elevated sulphur concentrations exist in the form of sulphate from the oxidation of pyrite (Table 2) and pH is negatively correlated with Fe and sulphur release indicating sulphide oxidation occurrence (eq. 4). In the sludge remediated cell, the low concentrations of sulphur may occur from the reduction of mineral processing sourced sulphate due to the addition of organic matter (eq. 2) or because sulphide oxidation has

	Tailings (ppm)	Sewage Sludge (ppm)
Major Elements	**TS**	**TS**
Si	166878	-
Al	21435	52200
Ca	34091	10300
Fe	94423	10600
K	5197	1350
Mg	22011	1400
Mn	767	232
Na	2070	-
P	168	18900
Ti	1103	-
Minor Elements	**TS**	**TS**
As	3610	145
Ba	128	219
Cd	8.19	0.97
Co	81.6	<2
Cr	25.1	34
Cu	1480	219
Hg	2.84	0.51
Ni	12.8	9.13
Pb	1270	48.5
S	209000	-
Zn	5330	377

***Table 1** Original chemical composition of tailings and sludge in 2001 (- indicate no analysis)*

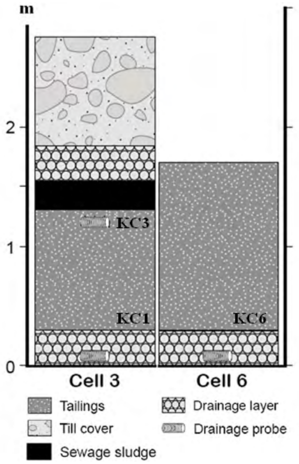

***Figure 1** Profiles of the two cell cover pilot-scale experiments with reference to leachate probes*

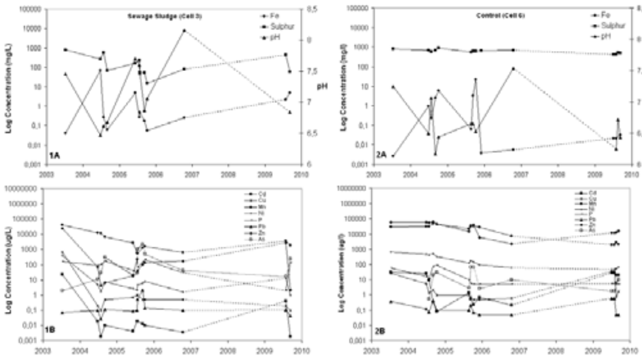

***Figure 2** Time-series graphs of the base cell leachate evolutions between 2003—2009 (dotted line indicates absence of sampling between 2006—2009); 1A: Dissolved Fe, S and pH in sewage sludge remediated cell (KC1) leachate; 1B: Dissolved elements in sewage sludge remediated cell (KC1) leachate; 2A: Dissolved Fe, S and pH in control cell (KC6) leachate; 2B: Dissolved elements in control cell (KC6) leachate*

not occurred. Fe fluctuates with seasonal variations in both cells but concentrations decrease over time, peaking in early summer and decreasing in the autumn months. In the control this may be due to a flush-out effect as snow melt infiltrates through oxidation products that may have accumulated in the tailings profile during winter. In the sludge remediated cell elevated Fe may be sourced from the sewage sludge (Table 1), which leaches into the underlying tailings and remains as dissolved Fe^{2+} due to reduced conditions from the consumption of oxygen by the decomposition of organic matter in the sludge (eq. 1). This is indicated by the low dissolved oxygen concentrations (Table 2) both directly underneath the sludge layer (KC3: 4.06mg/L) and in the cell leachate (KC1: 2.64mg/L) when compared with the control (KC6: 6mg/L) and accounts for the high dissolved Fe concentrations that are on average, 10 times more elevated than in the control cell. The leachate from the sludge remediated cell contains positively correlated concentrations of sulphur, Fe and pH (fig. 1A) indicating that sulphide oxidation has not occurred.

Heavy Metal Concentrations

High concentrations of heavy metals occurred in the leachate of both cells in 2003 due to flushing. Elevated concentrations (fig. 2B) of Cd, Cu, Mn, Ni, Pb, Zn occurred in the control throughout 2003—2009. All concentrations decline gradually over time, except for Cd and Zn, which increase as they are more mobile at this pH range (pH6.88) in an oxidised environment (Palmqvist, et al. 1997). However, heavy metal concentrations in the control are up to 2 orders of magnitude higher than in the sludge remediated cell. Heavy metals in the sludge remediated cell leachate (fig. 1B) decrease rapidly in concentration over time after the initial flush stage and are lowest in 2009. The sources of metals in the control are due to sulphide oxidation products. The source of the heavy metals in the sludge remediated cell may be due to the release of elevated concentrations in the original sewage sludge composition (Table 1), and not as products from sulphide oxidation. However, Mn and As are elevated (fig. 1B), together with Fe discussed earlier, which may be leached from the sludge (Table 1). These are redox dependant elements and are hence mobile at near-neutral pH ranges within a reduced environment (Solaiman, et al. 2009). Peak concentrations of As occurred in 2005 (2180µg/L) and 2009 values were 246µg/L, higher than the 2003—2009 mean of 15µg/L in the control. Other heavy metals such as Ni-Cd-Zn may be adsorbed to pyrite in the reduced, near-neutral conditions in the tailings (Kornicker and Morse 1991), thus removed from solution and retained in the tailings.

pH, Alkalinity and Calcium

The pH in both cell leachates is above pH6.0 throughout 2003—2009 despite sulphide oxidation in the control. Ca concentrations are elevated in both cells. The sludge remediated cell leachate Ca concentration decreases over time from 774mg/L in 2003 to 220mg/L in 2009. In the control Ca concentrations remain constant at a mean value of 583mg/L. This is due to the dissolution of $Ca(OH)_2$ added during mineral processing (eq. 5) and calcite gangue minerals (eq. 7) which react with acidity and release calcium and HCO_3^- ions into solution. Because of the 2.5% calcite content in the original tailings, it is likely that the control cell pH will reduce over time due to continued

__Table 2__ Average anion and alkalinity concentrations in the leachate collected in summer 2009. KC1 and KC6 are collected from the drainage layer probes, and KC1 is collected from the lysimeter in the tailings profile 0.1m beneath sewage sludge layer

mg/L	NO_3^-	SO_4^{2-}	HCO_3^-	Dissolved O_2
KC1	13	142	750	2.64
KC6	0	1390	88	6.00
KC3	0.33	42	433	4.06

acidity release due to sulphide oxidation, exhausting the calcite. Ca concentrations decrease in the sludge remediated cell leachate because low acidity is produced. They remain relatively elevated however, due to the high Ca content in the original sludge (Table 1) which is leached into the tailings. If the Ca combines with the elevated SO_4^{2-} from the processing residue, gypsum (CaSO4•2H2O) may precipitate in the effluent drainage (eq. 6). Elevated alkalinity (HCO_3^-) in the sludge remediated cell (Table 2) may be due to sulphate reduction (eq. 2) with the subsequent release of H_2CO_3. Additionally, the release of H_2S gas (eq. 2) may combine with metals in solution (eq. 3) to precipitate metal sulphides in the reduced environment underneath the sewage sludge, trapping any metals released from the sewage sludge.

Conclusions

The sewage sludge sealing layer has been effective for reducing oxygen diffusion to underlying tailings for a period of 8 years. The consumption of oxygen and the reduction of sulphate by the decomposition of organic matter in the sludge have prevented sulphide oxidation and created a reduced environment in the tailings profile with a high buffering potential. Elevated heavy metal concentrations sourced from the sewage sludge have leached into the tailings and have been retained in the solid phase due to the reduced environment, accounting for low concentrations of dissolved heavy metals and sulphate in the effluent leachate. In contrast, the uncovered control cell leachate has exhibited high sulphate, heavy metal and dissolved oxygen concentrations in the leachate since 2003, evidential of continued sulphide oxidation of the tailings.

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