Advances in using sewage sludge to remediate sulfidic mine tailings – An overview from pilot- and field-scale experiments, northern Sweden

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Abstract Sewage sludge can and has been used effectively as an organic-rich cover for sulfidic mine waste remediation. However, the optimum use of the material as a layer in engineered covers remains unconfirmed. Results obtained from four different sewage sludge applications are presented and discussed in context to their success and for their applicability for being used at full-scale sulfidic-mine waste remediation projects. The experiments evaluated sewage sludge when used as a vegetation substrate on bare tailings, a water-saturated cover, a composite cover, and lastly when used as a sealing-layer barrier material, which was deemed the most successful technique.

Keywords Sewage sludge, geochemistry, remediation

Introduction

In Sweden, 59 Mt of mine waste are produced annually (Statistics-Sweden 2008b). The weathering of un-remediated sulfidic tailings may produce acid rock drainage (ARD). ARD mitigation in Sweden is largely directed towards constructing engineered dry covers to cap and reduce oxygen diffusion to the underlying sulfide tailings (Höglund *et al.* 2005).

Traditional cover materials have consisted of durable, unreactive natural materials such as glacial overburden or natural soil. Sourcing and excavating large volumes of these is often problematic and of further environmental concern. Replacing natural soils with an alternative such as an organic waste generated from another industry has become an inexpensive and attractive solution for the mining industry, providing the co-disposal of two separate wastes together. One such organic waste is sewage sludge (SS), a solid by-product material generated during the treatment of domestic waste-water.

Approximately 210 Kt of SS are produced annually (Statistics-Sweden 2008a) from more than 2100 waste water treatment plants in Sweden (Marklund 1997). It has been investigated as a vegetation substrate on the surface of tailing impoundments and waste rock dumps (Forsberg 2008), and has been proven to be a suitable substrate for supporting and sustaining long-term vegetation establishment (Pichtel *et al.* 1994). SS is not traditionally utilised as a sealing layer material for mine waste, but it may have the potential for such usage because of its favourable physical and geochemical suitability, such as a low hydraulic conductivity of 1×10^{-9} m/s (Mácsik *et al.* 2003). In addition, fresh SS contains a high organic matter content that may allow it to function as an organic reactive barrier (Peppas *et al.* 2000).

However, limitations surround the use of SS as it is chemically unstable. It may contain readily-leachable concentrations of metals (Cd, Cu, Ni, Pb, Zn; Eriksson 2001) which may migrate as soluble organo-metallic complexes (Andres and Francisco 2008). The release of nitrate may exceed vegetation requirements and leach into underlying sulfide-mine tailings (Cravotta 1998) where it may be a primary terminal electron acceptor to pyrite where groundwater is devoid of oxygen (Appelo and Postma 2005).

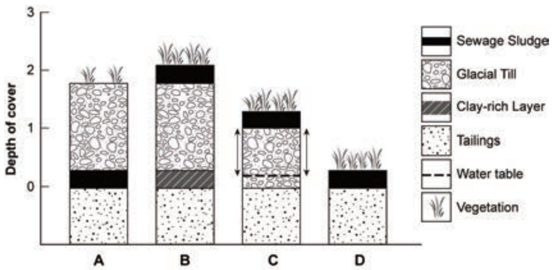

Fig. 1 *Schematic diagram of the four different sewage sludge applications*

The use of SS may therefore be problematic. This paper evaluates four different SS applications at pilot- and field-scales in northern Sweden. The experiments were conducted over the course of 0 to 8 years at 2 mine sites run by the Swedish mining company New Boliden AB. The results are summarized in this paper. It is the aim of this paper to identify the optimum use for SS so that it may be utilised by the mining companies on successful sulfide-tailings remediation projects in the future.

Study Sites and Experimental Set-up

Pilot-scale Experiment: Sewage Sludge Sealing Layer

The Georange Environmental Test Site is located at the Kristineberg Mine in northern Sweden (Fig. 2). A SS sealing layer was evaluated to understand if it was able to prevent oxygen diffusion and ARD formation to underlying tailings. Data from pore gas, leachate and sediment geochemistry were derived over an 8 year time-span. The engineered dry cover arrangement was applied above a 1.0 m thick fresh unoxidized tailings layer sourced from the Kristineberg Pb-Zn-Cu mine (Fig. 1: A). It consisted of a 0.25-0.30 m thick compacted anaerobically-digested SS sealing layer and an overlying 1.5 m thick protective layer consisting of locally-derived glacial till. For reference, the geochemistry was compared to a neighbouring cell consisting of 1.4 m thick unoxidized tailings identical in origin and experimental set-up.

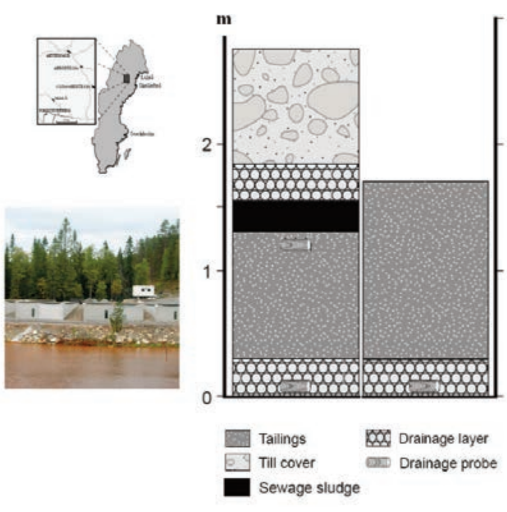

Fig. 2 *Experiment set-up and location of the pilot-scale test cells*

Field-scale experiment: Sewage sludge vegetation substrate above engineered dry composite cover and water saturated cover

Field trials were conducted at a formally remediated sulfide-tailings facility, Impoundment 1 (Fig. 3), at the Kristineberg Mine. After 50 years of natural weathering, sulfide oxidation and ARD formation, the impoundment was remediated in 1996 by applying an engineered composite dry cover (1.5 m protective layer of glacial till and 0.3 m clay-rich till sealing layer) to the formerly raised tailings dam area, and by applying a simple 1 m thick water-saturated till cover in the west of the impoundment. In August 2009, 10.8 Kt of anaerobically-digested SS sourced from Stockholm Municipality was applied onto the dry covered area of the impoundment, and 1.2 Kt onto the water saturated cover areas to a depth of 0.3 m. It was applied to provide a substrate and nutrients to areas of the impoundment that had had poor vegetation establishment. A profile of the two cover applications is illustrated in Fig. 1: B/C.

Groundwater monitoring took place a few years after remediation in 1998–2006 and from June 2009-October 2011 after the SS application. Samples were collected from five installed groundwater wells from: the uncontaminated inflowing groundwater (Well P); the

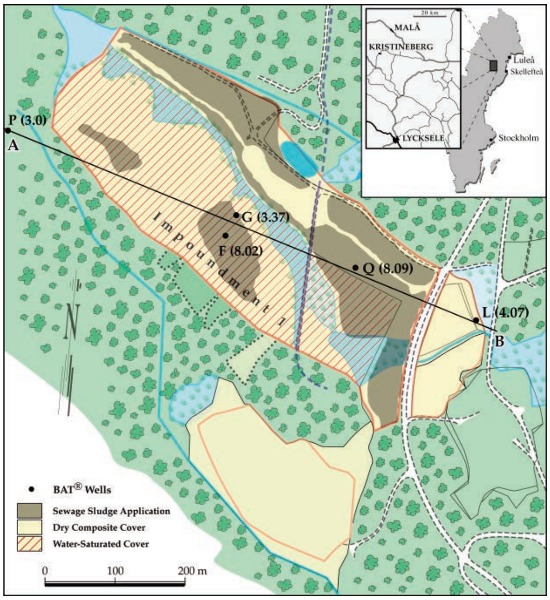

***Fig. 3** Map of Impoundment 1 showing location and cover types with sewage sludge application. Line A-B indicates dominant groundwater flow direction*

water-saturated areas of the impoundment (Well F and G); the dry-covered areas of the impoundment (Well Q); and the outflow at the impoundment toe (Well L). Solid SS samples were collected before application, and after 2 years. The objective of the study was to identify if sludge-borne metals and nitrate were released to the underlying groundwater system, and if so, to delineate their magnitude, duration and fate.

Sewage sludge vegetation substrate directly on tailings

Field trails were conducted on a large-scale unremediated sulfide-tailings facility, the Gillervattnet Impoundment, at the Boliden Mine. Fresh tailings were applied up until 2008. Three plots were sampled for solid inorganic geochemistry, and pore-water analysis: an uncovered reference tailings plot oxidized for 2 years; an uncovered reference tailings plot oxidized for 1 year and covered with SS for 1 year; a tailings plot covered with SS immediately after application 2 years ago. The SS in both applications was sourced from the Stockholm Municipality. It was applied to a depth of 0.2 m in both applications (Fig. 1: D). The three applications can be visualized in Fig. 4. It was the objective of this study to evaluate if the SS altered the underlying tailings geochemistry, if it was effective at allowing successful vegetation establishment and if it prevented sulfide oxidation.

Results

Sewage Sludge Sealing Layer

The SS sealing layer was effective at reducing oxygen diffusion to the underlying tailings. It was calculated that a mass transfer of 0.54 $mol^{-1}m^{-2}a^{-1}$ oxygen occurred during the period 2004–2005 (Nason *et al.* 2013), conforming to the limitations set out for a conventional sealing layer in Sweden of <1 $mol^{-1}m^{-2}a^{-1}$ (Carlsson 2002). Measured oxygen, carbon dioxide and methane indicated

***Fig. 4** Three field-scale plots at the Gillervattnet Impoundment: Uncovered tailings oxidized for 2 years; 1-year application of sewage sludge; 2-year application of sewage sludge*

that the SS may have functioned as an organic reactive barrier, consuming oxygen due to aerobic degradation of the organic matter, and producing methanagenesis and sulphate reduction.

It was additionally advantageous in that it created a reductive, alkaline environment in the underlying sulfidic-mine tailings, which promoted the precipitation of metals, improving the quality of the effluent drainage. The geochemical processes are illustrated by the schematic diagram in Fig. 5. However, up to -85 % depletion of the organic fraction of the SS occurred, accounting to a total volume loss of -20 % from the sealing layer. This is far less than surface applications of SS, though it should be considered a possible limiting factor when using SS as a sealing layer material in the long-term (>100 years). Nevertheless, in comparison the uncovered sulfide-bearing mine tailings received extensive sulfide oxidation characterised by the onset of ARD and a 0.35m deep acidic oxidation front formation.

Sewage sludge vegetation substrate above an engineered dry composite cover

Within the first year after SS application substantial vegetation establishment had occurred, and was a host to a vole population and an attractive bird-population such as grey wagtails, lapwings and rough-legged buzzards. The visual improvement of the SS application can be seen in Fig. 6. However, geochemically, the SS mass volume decreased in the two years after the application by -16.25 %. The mass lost was attributed to organic matter depletion (-22 %) and the leaching of sulfate, calcium and the metals, Cu, Ni, Pb and Zn. Nitrification of the SS also released excessive nitrate into the underlying cover materials. Fortunately, the results indicate that the function of the original engineered dry cover underlying the SS application focused the sludge-borne leachate laterally, where it was concentrated at the impoundment toe, without reaching or reacting with the tailings groundwater system.

Sewage sludge vegetation substrate above an engineered water saturated cover

Similarly to the dry covered area, plant establishment occurred within 1 year of application, and the SS mass volume decreased in the two years since the application by -17.76 %. Copper, Pb and Zn were removed more so than from the dry covered area. This was due to a rapid mobilization of the metals by the interaction of the raised groundwater table with the SS. Consequently, unhindered due to the lack of a sealing layer, the sludge-borne constituents migrated into the underlying tailings groundwater system with concentrations of the metals Cu (188 µg/L), Ni (263 µg/L), Pb (95 µg/L) and Zn (2060 µg/L) peaking two years after the SS application occurred, up from 2009 pre-SS concentrations of Cu (0 µg/L), Ni (3.8 µg/L), Pb (0.4 µg/L) and Zn (1610 µg/L). The plume readily migrated laterally underneath the entire

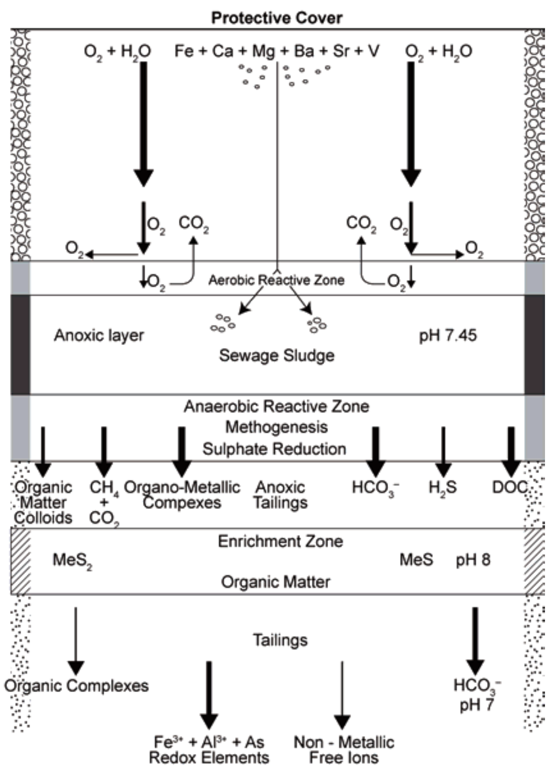

Fig. 5 *Schematic diagram of the dominant geochemical processes governing in the sewage sludge sealing layer experiment*

impoundment to the toe due to speciation linked with organo-complexes. It was accompanied by a nitrate pulse of 67 mg/L. Using SO_4^{2-}/Fe^{2+} molar ratios, pyrite oxidation was indicated in the tailings as the plume travelled laterally, caused by the introduction of nitrate as a primary terminal electron acceptor for pyrite, and as indicated by a decrease in pH and an increase in Fe concentrations. The plume was released by the SS over a 2-year period and ceased when vegetation establishment immobilized and retained the nitrate and metals, as has been prevalent in similar studies conducted in Sweden (Forsberg 2008). Concentrations subsided to within ± 10 % of pre-SS concentrations by 2011 in the area underlying the SS application in the water-saturated area. Modelling of the plume has indicated it will have exited the impoundment within 6 years after the SS application.

Sewage sludge vegetation substrate directly onto tailings

The unoxidized tailings plot had a 0.05 m deep oxidized zone characterized by active sulfide oxidation, indicated by a reduced pH (2.75) and elevated Cd and Pb pore water concentrations. In the two-year SS application, successful plant establishment (Fig. 4) had occurred. However, upon visual inspection, the plant roots from the application penetrated into the underlying tailings, creating oxygen and water pathways, and a 0.04 m oxidation zone. Concentrations of Cu, Fe and Zn, significantly increased in the underlying pore-water, up to two magnitudes higher than the reference, in the immediate 0.1 m of tailings. The pH was maintained at circum-neutral. Iron precipitated in the top 0.2 m of the underlying tailings yet the Cu and Zn did not, and continued to exhibit elevated pore-water concentrations up to 0.8 m in depth. It is likely that these metals were sludge-borne. In the 1 year sludge applied cover, no plant establishment had occurred, but cracks in the SS existed, causing a 0.02 m oxidation front in the underlying tailings. A similar pattern in the pore-water geochemistry to the 2-year application existed but to a lesser degree.

Optimum approaches to sewage sludge use in sulfide-mine tailings remediation

The best practices for the mining industry are ranked and summarized as follows:

1. The use of SS as a sealing layer offers a medium-to-long term-solution to the abatement of ARD, due to the successful mitigation the material has at decreasing oxygen diffusion to the underlying tailings. The primary limitation of SS in this application type regards the life-time of the cover due to the degradation of organic matter.
2. Using SS as a vegetation substrate onto an engineered dry composite cover is a successful remedial approach for promoting vegetation establishment. A temporary nitrate and metal-laden sludge-borne leachate may be released over a 2 year period. Though avoiding direct contact with tailings, it must be collected and treated

Fig. 6 *Impoundment 1: Before sewage sludge application; after sludge application of the dry covered area; after sludge application on the water-saturated area*

before entering peripheral environments.

3. Using SS on water-saturated cover types should be avoided. Though temporary, the nitrate and metal-laden sludge-borne leachate rapidly enters the tailings groundwater system, creating further oxidation of pyrite, and creating a contamination plume of elevated nitrate, Cu, Fe, Ni, Pb and Zn. Its residence time is dependent on the dominant hydrogeological regime.
4. Using SS directly onto fresh tailings should be avoided. Though vegetation is established rapidly, and it is aesthetically pleasing, cracking via drying and root penetration, promotes sulfide oxidation, and in addition the SS releases elevated sludge-borne metals (Cu, Fe, Zn) to the underlying tailings. This application merely slows the inevitable formation of ARD.

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References

Andres NF, Francisco MS (2008) Effects of sewage sludge application on heavy metal leaching from mine tailings impoundments. Bioresource Technology 99: 7521–7530

Appelo CAJ, Postma D (2005) Geochemistry, Groundwater and Pollution, 2nd Edition A.A. Balkema Publishers

Carlsson E (2002) Soil-cover remediation of sulphide-rich tailings. Technical Report, SBLO50: EC2002

Cravotta CA (1998) Effect of sewage sludge on formation of acidic ground water at a reclaimed coal mine. Ground Water 36: 9–19

Eriksson J (2001) Concentrations of 61 trace elements in sewage sludge, farmyard manure, mineral fertiliser, precipitation and in soil and crops: 5159–69

Forsberg SL (2008) Reclamation of Copper Mine Tailings Using Sewage Sludge. Doctoral Thesis, 88 pp

Höglund LO, Herbert RB, Lövgren L, Öhlander B, Neretnieks I, Moreno L, Malmström M, Elander P, Linvall

M, Lindström B (2005) MiMi – Performance Assessment – Main Report: MiMi Report 2003:3. In:

Höglund, LO and Herbert RB (Eds.), MiMi Print, Luleå, Stockholm

Mácsik J, Rogbeck Y, Svedberg B, Uhlander O, Mossakowska A (2003) Fly ash and sewage sludge as liner material – Preparation for a pilot test with fly-ash stabilized sewage sludge as landfill liner. Report 837. (In Swedish)

Marklund S (1997) Dewatering of Wastewater Sludge by Natural Air Drying. Licentiate Thesis, 7 pp.

Nason P, Alakangas L, Öhlander B (2013) Using sewage sludge as a sealing layer to remediate sulphidic mine tailings: a pilot-scale experiment, northern Sweden. Environmental Earth Sciences, DOI: 10.1007/s12665-013-2369-0 (Article in Press)

Peppas A, Komnitsas K, Halikia I (2000) Use of organic covers for acid mine drainage control. Minerals Engineering 13: 563–574

Pichtel JR, Dick WA, Sutton P (1994) Comparison of Amendments and Management-Practices for Long-Term Reclamation of Abandoned Mine Lands. Journal of Environmental Quality 23: 766–772

Statistics Sweden (2008a) Discharges to Water and Sewage Sludge Production in 2006. Published in Collaboration with the Swedish Environmental Protection Agency http://www.scb.se/templates/Publikation__232141.Asp

Statistics Sweden (2008b) www.scb.se Published in Collaboration with the Swedish Environmental Protection Agency