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Alternative waste residue materials for passive in situ prevention of sulfide-mine tailings oxidation: A field evaluation

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HIGHLIGHTS

- Sewage sludge and fly ash were evaluated in novel field-scale dry cover amendments.
- Sewage sludge was ineffective at mitigating ARD formation in underlying tailings.
- Sludge-borne metals (Cu, Ni, Fe, Zn) accumulated in underlying tailings.
- The fly ash application under the sewage sludge cover layer aided oxygen diffusion mitigation.
- Sewage sludge hampered the application, providing nitrate as oxidant to pyrite.

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ABSTRACT

Novel solutions for sulfide-mine tailings remediation were evaluated in field-scale experiments on a former tailings repository in northern Sweden. Uncovered sulfide-tailings were compared to sewage-sludge biosolid amended tailings over 2 years. An application of a 0.2 m single-layer sewage-sludge amendment was unsuccessful at preventing oxygen ingress to underlying tailings. It merely slowed the sulfide-oxidation rate by 20%. In addition, sludge-derived metals (Cu, Ni, Fe, and Zn) migrated and precipitated at the tailings-to-sludge interface. By using an additional 0.6 m thick fly-ash sealing layer underlying the sewage sludge layer, a solution to mitigate oxygen transport to the underlying tailings and minimize sulfide-oxidation was found. The fly-ash acted as a hardened physical barrier that prevented oxygen diffusion and provided a trap for sludge-borne metals. Nevertheless, the biosolid application hampered the application, despite the advances in the effectiveness of the fly-ash layer, as sludge-borne nitrate leached through the cover system into the underlying tailings, oxidizing pyrite. This created a 0.3 m deep oxidized zone in 6-years. This study highlights that using sewage sludge in unconventional cover systems is not always a practical solution for the remediation of sulfide-bearing mine tailings to mitigate against sulfide weathering and acid rock drainage formation.

1. Introduction

Mining and processing of sulfide ores may produce significant quantities of waste residues, such as sulfide-bearing tailings, that are disposed of in large-scale repositories. Without remedial efforts, atmospheric weathering by oxygen and water, in the presence of bacteria, will create the onset of pyrite and other gangue metal-sulfide mineral oxidation, with the consequential formation of acid rock drainage (ARD) [1]. ARD is characterized by elevated dissolved sulfate (>1000 mg/L), acidity (<pH 6) and metal loadings [2]. Passive treatment techniques such as permeable reactive barriers (PRB's) treat pre-existing ARD-contaminated water and have been studied extensively in the literature [3,4]. Passive in situ (PIS) prevention techniques, function oppositely to PRB's, in that they prevent ARD formation. Flooding, capping, and/or organic carbon amendments aim at reducing oxygen transport to sulfide-tailings, thereby eliminating sulfide mineral oxidation and the formation of ARD [2].

ARD mitigation by PIS prevention requires forward thinking. Remediation must commence directly during and/or after tailings deposition. The application of engineered dry covers using solid materials is an alternative to flooding sulfide-mine tailings [5]. Dry

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cover materials must consist of durable, un-reactive natural materials such as glacial overburden, natural soil or clay. Sourcing and extracting large volumes of suitable natural materials may pose further environmental impacts related to the mining operation. Replacing natural materials with readily-available waste residues generated from other industries has become an inexpensive and novel solution alternative, providing the co-disposal of two residual wastes together. By-products from the pulp and paper industries, as well as from municipal waste-water treatment plants are currently being utilized globally [6–8].

Sewage sludge (SS) is a biosolid residue generated during the treatment of domestic waste-water [9]. Fly-ash (FA) is derived via the combustion of biofuels, such as peat, coal, oil or municipal solid waste. Both materials are attractive waste residues to use in novel dry cover solutions for sulfide-tailings remediation. Approximately 210 000 t of SS [10] and 435 000 t of FA [11] are produced annually in Sweden. These waste materials are now beginning to be used in PIS prevention techniques for the treatment of mine drainage [12].

FA residue composition generally consists of high levels of Si, Ca, Al, S, and heavy metals [13]. It possesses high alkalinity and may form a hardened layer due to the formation of gypsum and hydrated silicates upon contact with water [14]. FA has been used to decrease metal leaching from contaminated soils by adsorbing metals onto Fe-hydroxides [15,16]. FA also contains K, Ca and Mg, important as plant nutrients [17] that are helpful in establishing vegetation during tailings reclamation.

The utilization of SS as a dry cover material is diverse. Traditionally, SS has been used as a vegetation substrate directly above sulfide mine tailings [18] and waste rock [19]. SS may reduce soil bulk density [18], is a considerable source of macro-nutrients [20], and may be effective at increasing plant biomass in the long-term [21]. It has also been harnessed as an organic-rich substrate in PRB's [22]. Recent advances have been achieved by using it as a sealing layer barrier material [23] within an engineered composite cover, to prevent oxygen diffusion to underlying sulfide-tailings.

SS is usually disposed of via land filling [24]. It may contain readily-leachable elevated concentrations of metals (Cd, Cu, Ni, Pb, Zn) [25]. Agricultural SS applications have shown metals may accumulate in underlying soil horizons [9], or be transported to peripheral ground or surface waters due to organo-metallic complexation with elevated dissolved organic carbon (DOC) [26,27]. The release of nitrate from agricultural applications of SS is also prevalent [28,29] due to the oxidation of ammonium, present in high concentrations in fresh SS [14]. Nitrate may exceed vegetation requirements and leach into underlying sulfide-mine tailings [30] where it may be a primary terminal electron acceptor for pyrite oxidation where groundwater is devoid of oxygen [31].

SS, when applied as a single dry cover layer onto bare sulfide-tailings, with the primary function to prevent oxygen diffusion and ARD formation, is unconventional. Its use as a single layer may however be advantageous in that it may provide an inexpensive remedial method. In addition, surface applications of SS onto sulfide-tailings have been proven to chemically stabilize metals in situ [32]. Fresh SS contains a high organic matter (OM) content that may allow it to function as an organic reactive barrier [3], consuming oxygen by aerobic degradation [33].

The use of SS as a surface material directly above sulfide-mine tailings to prevent sulfide-oxidation is hypothetically problematic. This paper evaluates the geochemical influence this type of application may have on underlying sulfide-tailings and investigates if a combined SS–FA cover would improve the amendment. Field experiments were conducted on a tailings repository in northern Sweden. The solid, leachate and pore-water geochemistry were collected from four experimental plots over time periods ranging from 0 to 6 years, to identify the release, transport and attenuation of sludge-borne metals and constituents to the underlying sulfide-tailings.

2. Study area

The Boliden base-metal concentrator site is located in the Skellefte Ore District in northern Sweden, 30 km north-west from the city of Skellefteå (Fig. 1). The concentrator processed Cu, Pb, Zn, Au and Ag ores from over 30-proximal mines since 1953, depositing the waste residues into the proximal Gillervattnet tailings impoundment. The climate of the area is classified as sub-Arctic with sub-zero temperatures between October and April, and a mean annual temperature of 0.7 °C [34]. Field trials were conducted on four plots situated on the Gillervattnet tailings impoundment (Fig. 1).

2.1. Field Experiment A: sewage sludge application

In July 2008, freshly deposited sulfide tailings were divided into three plots to investigate the application of SS directly onto mine tailings:

1. SS0: uncovered tailings left to weather for 2-years.
2. SS1: uncovered tailings left to weather for 1-year and covered with 0.2 m of SS in July 2009.
3. SS2: 0.2 m of SS applied onto fresh tailings immediately in July 2008.

Sampling for all plots was conducted after 2-years in July 2010.

2.2. Field Experiment B: combined sewage sludge–fly ash application

In July 2004, a combined engineered SS–FA cover was completed above tailings that were deposited in 2002. The experiment was located 200 m to the east of Field Experiment A (Fig. 1). The application consisted of a 0.25 m thick SS layer that was applied in July 2004 above a 0.6 m thick compacted FA layer that was applied in July 2003. After seeding, the plot was successfully vegetated by 2005 and sampling occurred in July 2010.

3. Materials and methods

3.1. Source materials

3.1.1. Field Experiment A

The SS used in the SS1 and SS2 plots was derived from the nearby municipality of Skellefteå (Fig. 1). A fresh SS spot sample (SSP) was collected in 2010, derived from the same source. The SS consisted of an 80:20 vol% ratio mix of two SS's sourced from a wastewater treatment plant, and a biogas plant. The wastewater treatment plant had anaerobically digested the biosolid at 38 °C for 15 days; whereas the biogas plant had anaerobically digested it at 53 °C for 50 days. Both were dewatered to 22 % dry weight before mixing in the field.

3.1.2. Field Experiment B

SS used in this study was derived from Stockholm Vatten wastewater treatment plant where it had been anaerobically digested. The FA used was sourced from the nearby Skellefteå Kraft in Hedensbyn. The FA was derived from a circulating fluidized bed furnace that burnt biofuel and peat (80:20). The full original tailings, SS and FA solid geochemical analyses from the SS–FA plot are described in Neuschütz and Greger [35] and have been used as a reference for the data collected in the results.