



Impact of mitigation strategies on acid sulfate soil chemistry and microbial community



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HIGHLIGHTS

- Acid sulfate soils cause environmental damage by releasing acid and metals.
- Acid sulfate soils treated with CaCO₃ and Ca(OH)₂ released solutions with higher pH.
- The microbial populations associated with treated and untreated soils were similar.
- The data can be used to design mitigation strategies to treat acid sulfate soils.

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ABSTRACT

Potential acid sulfate soils contain reduced iron sulfides that if oxidized, can cause significant environmental damage by releasing large amounts of acid and metals. This study examines metal and acid release as well as the microbial community capable of catalyzing metal sulfide oxidation after treating acid sulfate soil with calcium carbonate (CaCO₃) or calcium hydroxide (Ca(OH)₂). Leaching tests of acid sulfate soil samples were carried out in the laboratory. The pH of the leachate during the initial flushing with water lay between 3.8 and 4.4 suggesting that the jarosite/schwertmannite equilibrium controls the solution chemistry. However, the pH increased to *circa* 6 after treatment with CaCO₃ suspension and *circa* 12 after introducing Ca(OH)₂ solution. 16S rRNA gene sequences amplified from community DNA extracted from the untreated and both CaCO₃ and Ca(OH)₂ treated acid sulfate soils were most similar to bacteria (69.1% to 85.7%) and archaea (95.4% to 100%) previously identified from acid and metal contaminated environments. These species included a *Thiomonas cuprina*-like and an *Acidocella*-like bacteria as well as a *Ferroplasma acidiphilum*-like archeon. Although the CaCO₃ and Ca(OH)₂ treatments did not decrease the proportion of microorganisms capable of accelerating acid and metal release, the chemical effects of the treatments suggested their reduced activity.

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1. Introduction

Sulfide rich sediments are widespread in many low-lying coastal areas worldwide and are especially prevalent in Australia, Asia, and Europe (Andriessse and van Mensvoort, 2006). These sediments are referred to as potential acid sulfate soil (PASS) material when they

have not been oxidized (Rabenhorst et al., 2006). Natural phenomena (e.g., land uplift) or artificial drainage (e.g., for agricultural use) can expose the sediments to atmospheric oxygen and result in metal sulfide oxidation and the subsequent formation of acid sulfate soil (ASS) (Sundström et al., 2002). These soils typically have a pH of 2.5–4.0 (Åström, 2001). Finland has approximately 3000 km² of ASS in coastal farmland developed from sulfide rich sediments (Beucher et al., 2013) that constitutes the largest ASS area in Europe (Andriessse and van Mensvoort, 2006). As a result of the oxidation of iron sulfides, large amounts of acid and high level of metals (e.g., Al, Cd, Ni, and Mn) are released (Åström, 2001; Boman et al., 2010; Nordmyr et al., 2008; Sundström et al., 2002) that leads to severe environmental damage. For instance, massive acidic drainage from ASS has caused large fish kills in western coastal areas of Finland as recently as 2006.

Abbreviations: PASS, potential acid sulfate soil; ASS, acid sulfate soil; RFLP, restriction fragment length polymorphism.

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Sulfur- and iron-oxidizing microorganisms are widely found in many acid environments such as acid mine drainage (Hallberg, 2010), sulfide mine tailings (Rzhepishevska et al., 2005), and ASS (Wu et al., 2013). The role of extreme acidophiles in catalyzing the iron and/or inorganic sulfur compound oxidation leading to the release of acid and metals from metal sulfides is well established (Vera et al., 2013). These microorganisms need to be able to survive in the low pH and high metal concentrations generated by sulfide mineral dissolution and their pH homeostasis (Slonczewski et al., 2009) and metal resistance (Dopson et al., 2003, 2014; Dopson and Holmes, 2014) strategies have been reviewed. A microbiological study of ASS in boreal environments identified several 16S rRNA gene sequences similar to known acidophilic iron and inorganic sulfur oxidizing microorganisms, suggesting that the acid and metal releases are accelerated by these microorganisms (Wu et al., 2013). In addition, 16S rRNA gene sequence analysis suggested the presence of acidophiles adapted to the low temperatures found in Finland. These included the iron- and sulfur-oxidizing species *Acidithiobacillus ferrivorans* and 16S rRNA gene sequences related to clones detected in Arctic soil where rocks had been weathered by chemolithoautotrophic microorganisms (Borin et al., 2010). A second study of boreal ASS identified the microbial populations, including methanotrophs, in ASS and non-ASS and demonstrated the capacity for greenhouse gas emission if PASSs are drained (Šimek et al., 2014).

Due to the significant damage caused by acid and metal release from acid mine drainage, various remediation techniques have been developed including chemical treatment with lime, calcium carbonate, sodium hydroxide, etc., to neutralize the total acidity of the acid mine drainage liquors (Johnson and Hallberg, 2005). Several strategies have also been studied to prevent pyrite oxidation or to remediate the acidic groundwater caused by ASS (Golab et al., 2006; Indraratna et al., 2005). Techniques such as controlled drainage, surface liming, and lime-filter drainage have also been tested on ASS in Finland (Åström et al., 2007). However, the applied methods only appear to have had a minor effect on the release of acid and metals.

In this study, a new strategy of sub-surface calcium carbonate and calcium hydroxide treatment on metal and acid generation at the Risöfladan experimental field, Finland was evaluated in laboratory-scale column experiments. Compared to previous studies, this approach specifically targeted the hydrologically active macropores in the critical soil horizon. In addition, the impact of the chemical treatments on the microbial community present in the ASS was evaluated. The results can be utilized for the long term mitigation of environmental damage caused by boreal ASS.

2. Methods

2.1. Study site and sampling

ASS samples for chemical treatments were collected at the Risöfladan experimental field in Vaasa, Finland (63.05°N, 21.71°E) by removing the plow layer (0–30 cm) and then pushing polyethylene tubes into the ground with an excavator. Details of the study area (Åström et al., 2007; Boman et al., 2010) and the physicochemical parameters of the Risöfladan experimental field (Wu et al., 2013) have been previously reported. In brief, the soil below the plow layer consisted of clay with a well-developed structure with an abundance of iron oxide coatings. The sulfur content was in the order of $\geq 0.2\%$ (wt/vol) and the pH was ~ 4 in all soil samples due to significant sulfide oxidation (Wu et al., 2013). In total, 24 cores were used in preliminary studies, method development, and the five cores for the experiments reported here. A new core was utilized for each experiment by slicing a soil sample from the appropriate soil horizon as described below. The tubes with the soil profiles were immediately sealed at both ends to prevent the ingress of air and delivered to the laboratory. A cylinder-shaped sample was obtained by cutting the 14.2 cm inner-diameter polyethylene tube into a 15 cm long piece and carefully removing the soil core.

The sample (from the soil horizon extending from 70 to 85 cm below the surface) was inserted into a cylindrical rubber membrane that was pressed against the soil during the laboratory experiment by submerging in water and applying an external pressure to prevent bypass flow between the membrane and the soil sample. Even though this soil horizon was designated as oxidized, it still contained significant amounts of pyrite (Boman et al., 2010).

2.2. Chemical methods and instrumentation

The experimental set-up for the column tests is shown in Fig. 1. The two chemical (CaCO_3 and $\text{Ca}(\text{OH})_2$) treatments were carried out in duplicate on separate soil samples. For each of the treatments, one representative experiment is presented and the data from the replicate experiments are in the Supplemental files. The experiment began with passing $\sim 10 \text{ dm}^3$ pure water through the column. During the experiments, pH, temperature-corrected (to 25 °C) conductivity, and oxidation-reduction potential (ORP) versus an Ag/AgCl (3.5 M KCl) reference electrode were continuously registered *in situ* at the outlet using an YSI Professional Plus instrument (YSI Inc., Yellow Springs, OH 45387, U.S.A.) with a flow-through cell (Quatro Cable Flow Cell) from the same manufacturer. The recorded ORP values were recalculated to potentials (E_{H}) versus the SHE by the addition of 205 mV, before being converted to pe values ($pe = E_{\text{H}} / (2.303RT)$). The column effluent was collected in a cylinder open to the atmosphere. Every few hours the volume of the outlet solution was measured and *ex situ* measurements of pH (Schott-Geräte CG822 or Radiometer PHM210) and conductivity (Radiometer CDM210) were performed. The concentrations of Cl^- , NO_3^- , and SO_4^{2-} were measured by ion chromatography (Dionex ICS 1100) and Al by UV spectrophotometry (Hach Lange DR 3900). Several soil samples were subjected to X-ray diffraction analysis using the PANalytical X'Pert Pro system with monochromatic $\text{Cu K}\alpha$ radiation. The phase composition of the samples was identified using the International Centre for Diffraction Data database. All experiments and measurements were performed at ambient temperatures (20–21 °C). Concentrations of selected metals were determined by Inductively Coupled Plasma Mass Spectrometry or by Inductively Coupled Plasma/Optical Emission Spectrometry. The treatment chemicals, CaCO_3 (trade name Nordkalk C2,

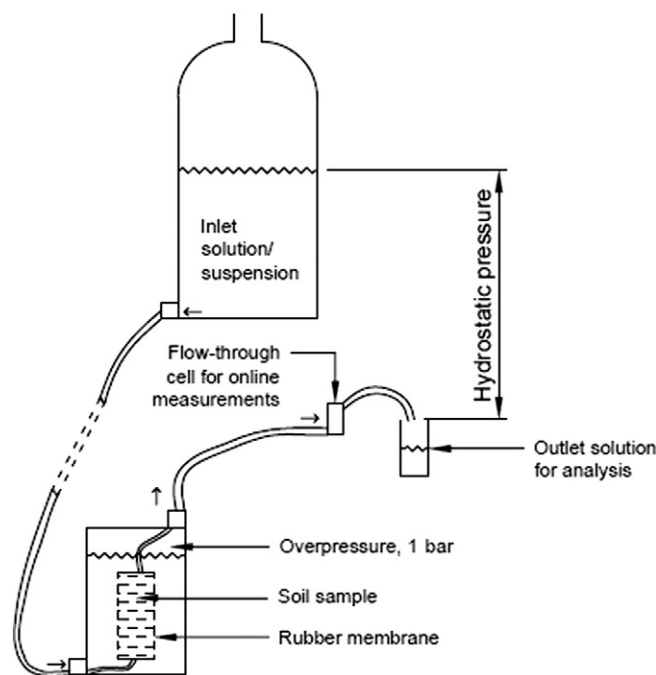


Fig. 1. Experimental set-up for the leaching tests. The maximum hydrostatic pressure employed corresponded to a water column of circa 3.7 m.

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