



Addition of organic material to sulfuric soil can reduce leaching of protons, iron and aluminium



Tan Dang^a, Luke M. Mosley^b, Rob Fitzpatrick^b, Petra Marschner^{a,*}

^a School of Agriculture, Food and Wine, The University of Adelaide, Adelaide, SA 5005, Australia

^b Acid Sulfate Soils Centre, The University of Adelaide, Adelaide, SA 5005, Australia

ARTICLE INFO

Article history:

Received 24 November 2015

Received in revised form 4 February 2016

Accepted 9 February 2016

Available online 23 February 2016

Keywords:

Acid sulfate soil

Aluminium

Biochar

Iron

Protons

ABSTRACT

Oxidised acid sulfate soils (ASS) with sulfuric horizons (sulfuric soils) can release large amounts of acid and metals, which can be rapidly mobilised when the soils are rewetted or re-flooded. Under flooded conditions, addition of organic materials to ASS has been shown to stimulate microbial sulfate reduction, which results in pH increase. But little is known about the potential of organic materials to reduce leaching of protons and metals from sulfuric soils and how this is influenced by properties of the organic materials and amendment form. Sulfuric material (pH 3.5) was collected from a coastal oxidised ASS at Gillman in the Barker Inlet, South Australia. Eight organic materials (compost, two plant residues and five biochars differing in feed stock and pyrolysis temperature) were applied at 15 g C kg^{-1} in two forms: mixed into or placed as a layer under the sulfuric soil. Twenty grammes of soil (dry weight equivalent) was placed in PVC cores, and the cores were leached four times with 45 ml of reverse osmosis water. In the unamended soil, 70–90% of the total leached protons, Fe and Al were released in the first leaching event with only small amounts being released in the three subsequent leachings. In amended treatments release of protons Fe and Al was lower than in unamended soil, the decrease is referred to as retention. The amount of protons, Fe and Al retained in the amended soil compared to the control was highest in the first leaching. Cumulative retention of protons Fe and Al was highest in the soil amended with eucalypt biochar produced at 550 °C and wheat biochar produced at 450 °C, but low in wheat straw and compost. Leachate pH of the unamended soil was 3.5–4, but up to 6.4 in amended soils. In amended soils, proton retention was positively correlated with C concentration of the materials, while Fe and Al retention was positively correlated with percentage of Aryl and O-Aryl groups and negatively correlated with percentage O-Alkyl, Di-O-Alkyl groups. Generally retention of Fe and Al was greater when organic materials were mixed in the soil than if placed as a layer underneath the soil, but there was little difference between amendment forms in proton retention. We conclude that mixing some biochars, particularly eucalypt biochar produced at 550 °C and wheat biochar produced at 450 °C, can strongly reduce leaching of protons and metals in sulfuric soils whereas wheat straw or compost are less effective.

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1. Introduction

Acid sulfate soils (ASS) are wide-spread and occur globally in over 500,000 km², mainly in coastal zones (Sullivan et al., 2012). ASS with sulfidic material (Soil Survey Staff, 2014) form under waterlogged or sub-aqueous conditions and are characterised by sediments or soils rich in iron sulfides, mainly pyrite (Ljung et al., 2009). When exposed to air, e.g. due to natural or artificial drainage, pyrite oxidation leads to release of sulfuric acid (Attanandana and Vacharotayan, 1986; Dent, 1986; Pons, 1973) and formation of ASS with sulfuric horizons (Soil Survey Staff, 2014). In sediments or soils with low pH buffering capacity, the pH can fall below 4 or even 2 (Attanandana and Vacharotayan, 1986; Dent, 1986; Dent and Pons, 1995; Harbison, 1986), which induces release

of metals to groundwater systems (Pavelic and Dillon, 1993). Acidity and high metal concentrations reduce plant growth (Bronswijk et al., 1995; Shamshuddin et al., 2014; Yampracha et al., 2005). The acidic leachate from sulfuric ASS (pH < 4) can have detrimental effects on ground and surface water quality and damage infrastructure (Baldwin and Fraser, 2009; Ljung et al., 2009). The dominant acid-generating metals in ASS leachates are Fe and Al, but protons are released when the metals hydrolyse (Mosley et al. 2014a).

Remediation of ASS is difficult, particularly when sulfuric material occurs at depth. Liming or controlled flooding may be ineffective or have undesirable side effects (Dear et al., 2002; Mosley et al., 2014a; b).

Organic matter plays an important role in sulfate reduction in ASS by providing energy for sulfate reducers and stimulating pyrite formation (Jayalath et al., 2015; Jayalath et al., 2016; Yuan et al., 2015a; b). Organic materials may also be useful for remediation of sulfuric soils because organic soil amendments such as plant residues, manure, compost or

* Corresponding author.

E-mail address: petra.marschner@adelaide.edu.au (P. Marschner).

biochar can increase the pH of acidic soils (Xu et al., 2006; Yuan and Xu, 2011) and bind metals (Clemente et al., 2006; Dias et al., 2003; Fest et al., 2008; Karami et al., 2011; Walker et al., 2004; Rees et al., 2014) and protons (Pedra et al., 2008). Biochar and other organic materials have also been shown to reduce metal concentrations in leachate from mine tailings (Beesley et al., 2014; Fellet et al., 2011), and waste water (Bhatnagar and Sillanpää, 2010; Wan Ngah and Hanafiah, 2008; Zhou and Haynes, 2010). These studies showed that biochar and other organic amendments can increase the pH and bind metals. But little is known about (a) the potential of these materials to reduce leaching of protons and metals from sulfuric ASS, and (b) how leaching is influenced by properties of the organic materials and amendment form (e.g. mixed into the soil or as a layer under the soil).

Using synthetic drainage water based on drainage water of sulfuric ASS, we showed that passage of the acidic drainage water through various organic materials reduced proton, Fe and Al concentrations (Dang et al., 2015). Retention of protons, Fe and Al was greatest in two biochars and least in compost and wheat straw. The present study expands on these results by investigating if organic materials could also be used to reduce leaching of protons and metals in oxidised ASS; thus when they are in direct contact with the sites where protons and metals are released. A further difference to our previous study is that concentrations of protons and metals varied between leaching events, which is in contrast to the constant concentrations during the five leaching events with synthetic drainage water.

The aims of this experiment were to (i) compare the effect of different organic materials on leaching of protons, Fe and Al from sandy sulfuric soil, and (ii) assess how this effect is influenced by the properties of the organic materials and their placement in the soil. We hypothesised that (i) organic materials will reduce leaching of protons, Fe and Al, but the ability to retain protons and metals will differ among materials, and (ii) leaching of protons, Fe and Al will be reduced to a greater extent when organic materials are mixed in the soil than if placed as a layer under the soil.

2. Materials and methods

2.1. Experimental design

Sulfuric material was collected from between horizons E and B of an oxidised ASS at Gillman (34°49'47.25"S; 138°32'40.24"E) in the Barker Inlet, South Australia near site BG 15 (Thomas, 2010) at 20–80 cm depth (for soil properties see Table 1). The soil profile is classified as Typic Sulfaquept (Soil Survey Staff, 2014), Hyperthionic Gleysol (Drainic, Humic, Hypersulfidic) according to the World Reference Base for Soil Resources (IUSS Working Group WRB, 2015) and Sulfuric clayey peat soil in accordance with the Australian ASS classification key (Fitzpatrick, 2013). The loss of tidal inundation has caused a lowering

of the water table in the estuarine and mangrove swamp environment at Gillman, enabling oxygen to diffuse into sulfidic material, which caused pyrite oxidation and the formation of the peaty sulfuric soil (Thomas, 2010). The area has ponding basins for urban stormwater runoff, which is released to the Barker Inlet at low tides (Thomas, 2010). The sulfuric material was collected at the edge of an exposed drain, which was partly under water at the time of collection. The collected sulfuric material is sandy with white specks of gypsum and pale brown to pale yellow jarosite mottles (Fig. S1, Table S1). The material was air-dried and sieved to 0.5–2 mm.

The following organic materials were used: wheat straw, pea straw, compost (from municipal green waste), and five biochars from different feed stocks, produced at 450 °C or 550 °C: poultry biochar 450 °C, poultry biochar 550 °C, wheat biochar 450 °C, wheat biochar 550 °C and eucalypt biochar 550 °C (Table 2). These materials were selected to have a wide range of properties and particularly to test various biochars. The same materials were used in our previous study (Dang et al. 2015). The organic materials were dried, ground and sieved to 0.5 mm.

The organic materials were applied at 15 g C kg⁻¹ of soil (dry mass basis) in two different ways: mixed with soil or placed as a layer underneath the soil. The control was soil without addition of organic materials. Twenty grammes of soil (dry weight equivalent of unamended soil) was placed in PVC cores (3.7 cm width, 5.0 cm height) with a mesh base (0.75 µm; Australian Filter Specialist Pty Ltd., Huntingwood, NSW). Before adding the soil, Whatman # 42 filter paper was placed on the mesh to minimise loss of soil and organic material during leaching events. There were four replicates per treatment. The cores were leached four times with reverse osmosis (RO) water. At each leaching event a total of 45 ml of water was added per core, in nine aliquots of 5 ml with 5 min between each addition. The total amount of water (45 ml, corresponding to 40 mm water depth) per leaching event was chosen based on preliminary studies to ensure sufficient leachate for the analyses. The 5 min interval was used to minimise pooling of water on the soil surface. After addition of 45 ml, the leachate was collected, its total volume measured, and analysed as described below. Leachate volume was 40 ml in unamended soil and 20–30 ml in amended treatments. Leaching was carried out every seven days for 4 weeks. In the interval between leaching events the cores were kept covered at room temperature; the soil remained moist between leaching events.

Leachate data were expressed as protons (10^{-pH}), Fe and Al retained in the materials for each leaching event and total cumulative retention. Retention per g of soil was calculated as:

(Concentration of element in unamended soil per ml * amount of leachate in ml) – (Concentration of element in amended soil per ml * amount of leachate in ml). This value was divided by the amount of soil per core (20 g) to give retention per g of soil.

2.2. Analyses

Soil texture was determined by the hydrometer method as described by Gee and Or (2002). The pH of the soil and organic materials was measured in a material to water ratio of 1:1 (w/w). Total organic C and total N in the soil and organic materials were measured by dry combustion using a LECO Trumac CN analyser. Total Al and Fe in the soil and organic materials were determined after concentrated nitric acid dissolution (Zarcinas et al., 1996). The extracts were filtered through Whatman #42 filter paper and analysed for Al and Fe by ICP-MS. Extractable sulfur including S_{HCl} and S_{KCl} of the soil were measured by shaking soil with 4 M HCl or 1 M KCl solution at a 1:40 ratio overnight or 4 h on end-over-end shaker (McElnea and Ahern, 2004b). Then the suspension was centrifuged at 3000 ppm for 5 min and the supernatant was filtered through Whatman #42 filter paper. The sulfur concentration in extracts was determined by ICP-AES. Chromium reducible sulfur (Scr), titratable actual acidity, retained acidity and net acidity were measured as described in Sullivan et al. (2004) and McElnea and

Table 1

Selected properties of the sulfuric soil material collected at Gillman in the Baker Inlet, South Australia.

pH _w		3.3
Sand	%	91.2
Silt	%	8.0
Clay	%	0.8
Total organic C	mg g ⁻¹	18.9
Total N	mg g ⁻¹	0.9
HCl soluble sulfur	mg g ⁻¹	11.5
KCl soluble sulfur	mg g ⁻¹	1.1
Chromium reducible sulfur ^a	mg g ⁻¹	17
Titratable actual acidity ^a	mmol H ⁺ kg ⁻¹	220
Retained acidity ^a	mmol H ⁺ kg ⁻¹	1084
Net acidity ^a	mmol H ⁺ kg ⁻¹	1462
Acid neutralising capacity ^a	% CaCO ₃ ⁻¹	0
Total Fe	mg g ⁻¹	21.9
Total Al	mg g ⁻¹	9.1

^a From Thomas (2010).

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