

# Organic matter addition can prevent acidification during oxidation of sandy hypersulfidic and hyposulfidic material: Effect of application form, rate and C/N ratio



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## ABSTRACT

Adding organic matter (OM) could minimize acidification during oxidation of sulfidic soils. Three incubation experiments were carried out under oxidizing conditions for 6 weeks using two acid sulfate soils: one with hyposulfidic material (soil Hypo) and one with hypersulfidic material (soil Hyper). The organic materials used were dried and finely ground (<2 mm) plant shoots: mature wheat straws a (Wa, C/N ratio 54) and b (Wb, C/N ratio 137) and young kikuyu shoots (C/N ratio 15). In all experiments, acidification in the un-amended treatments was smaller in soil Hypo than soil Hyper. In Experiment 1, Wa was added at 30 g kg<sup>-1</sup> by mixing into the soil or placing as a layer on the soil surface. After 6 weeks in both soils the pH was lowest in the un-amended control; soil pH was higher when OM was added by mixing than as a layer. In Experiment 2, Wb was mixed into soils at 0–40 g kg<sup>-1</sup>. Acidification of soil Hypo was prevented when ≥30 g kg<sup>-1</sup> Wb was added. In soil Hyper, OM addition reduced acidification. In Experiment 3 residues were mixed into the soils at 30 g kg<sup>-1</sup>: Wa, Wb and kikuyu alone and different mixtures of Wb and kikuyu (with C/N ratio 107, 76 and 46, respectively) for soil Hypo, or only Wa and Wb for soil Hyper. After 6 weeks, in soil Hypo the pH was highest with kikuyu and higher with Wa than Wb. The pH increased with decreasing C/N ratio.

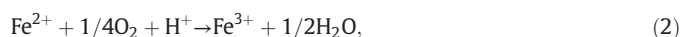
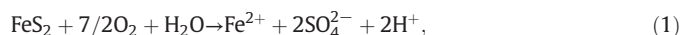
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## 1. Introduction

Acid sulfate soils (ASS) are soils containing iron sulfide minerals (predominantly pyrite) and/or their oxidation products (Van Breemen and Harmsen, 1975). ASS are widespread, e.g. 10<sup>5</sup>–10<sup>6</sup> km<sup>2</sup> coastal ASS world-wide (Andriess and van Mensvoort, 2007; Sullivan et al., 2012). In Australia, ASS occupy an estimated 215,000 km<sup>2</sup> of which 58,000 km<sup>2</sup> is coastal and 157,000 km<sup>2</sup> is inland non-tidal (Fitzpatrick et al., 2010). Generally, three soil materials are recognized in ASS (Fitzpatrick et al., 2009; Sullivan et al., 2010): (i) sulfuric material (pH <4) containing sulfuric acid, (ii) sulfidic material containing mainly pyrite (FeS<sub>2</sub>), and (iii) monosulfidic material with monosulfide minerals (FeS). In accordance with the World Reference Base for Soil Resources (IUSS Working Group WRB, 2014) and the Australian Soil Classification (Isbell, 2016), sulfidic material can be further classified into two types: hypersulfidic material and hyposulfidic material. The former is capable of severe soil acidification following oxidation of sulfides; the latter is not. The term hyposulfidic material does not exist in Soil Taxonomy, but the term hypersulfidic material equates to sulfidic material by Soil Taxonomy. In Australia, inland ASS environments are generally dry

and have already been impacted by drought (Mosley et al., 2014a; Mosley et al., 2014b), and more frequent droughts are expected in the future (Hobday and McDonald, 2014; Stokes et al., 2008). Therefore oxidation of ASS with sulfidic material is of great environmental concern.

After exposure to oxygen, pyrite can be oxidized according to the following reactions (Aherm et al., 2004; Chandra and Gerson, 2010):

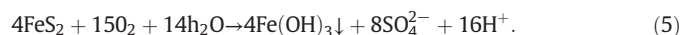


Oxygen initiates pyrite oxidation (Eqn. 1) but in most situations dissolved ferric iron (Fe<sup>3+</sup>) is the primary oxidant (Eqn. 4), and the transformation of dissolved ferrous iron (Fe<sup>2+</sup>) to ferric iron (Eqn. 2), mainly catalyzed by chemolithotrophic acidophilic iron-oxidizing bacteria, is considered as the rate-limiting step in pyrite oxidation (e.g., Emerson et al., 2010; Johnson and Hallberg, 2005).

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The overall equation for pyrite oxidation and hydrolysis of  $\text{Fe}^{3+}$  is:



This reaction leads to significant release of acid ( $\text{H}^+$ ) and resulting dissolved metals (Ahern et al., 2004; Johnson and Hallberg, 2005; Neculita et al., 2007), which can have detrimental effects on soil and water quality and thus ecosystem services (Boman et al., 2010; Bronswijk et al., 1993; Faltmarsch et al., 2008; Mosley et al., 2014a; Mosley et al., 2014b).

Conventional management to prevent oxidation or acidification of sulfidic material includes covering with water or non-ASS soil, or addition of neutralizing agents such as limestone, but this can be costly or not practically feasible (Astrom et al., 2007; Baldwin and Fraser, 2009; Dear et al., 2002). A more economical and environmentally friendly potential option is the application of organic matter (OM). OM can buffer pH change via binding protons to organic anions (e.g. carboxylate ion,  $\text{R-COO}^-$ ) or proton consumption during the decarboxylation of organic anions (Helyar, 1976; Paul and Ulf, 2011; Ritchie and Dolling, 1985; Rukshana et al., 2011; Yan and Schubert, 2000; Yan et al., 1996):



Regarding ASS, OM can also retard pyrite oxidation through oxygen consumption by decomposing microbes, complexation of dissolved ferric iron and coating of pyrite (Bronswijk et al., 1993; Bush and Sullivan, 1999; Cook et al., 2004; Rigby et al., 2006; Ward et al., 2004). Mitigation of oxidation of pyritic fines by OM was observed in a reaction vessel (Rigby et al., 2006), and mulching with OM has been suggested as a method to prevent oxidation or acidification of sulfidic material by maintaining anoxia and providing pH buffer capacity (Baldwin and Fraser, 2009; Fitzpatrick et al., 2009). In a field experiment, Fanning et al. (2004) reduced acidification in acid sulfate soils by addition of lime-stabilized biosolids (sewage sludge) at high application rates. The acid neutralization in this case was likely mainly because of the lime in the amendment, but the organic matter in the amendment may have improved effectiveness of the remediation. Systematic studies are required to assess and optimize the use of OM to prevent or minimize the acidification of sulfidic material. In particular, the influence of factors that regulate OM decomposition rate, for example OM properties, requires further attention to maximize amelioration outcome. Important OM properties in this context are C/N ratio and concentration of complex carbon compounds such as cellulose and lignin. OM with low C/N ratio (<20) can be decomposed rapidly because it contains sufficient N to satisfy the N requirements of microbes (Chapin et al., 2011; White, 2009). This could be beneficial in the context of preventing pyrite oxidation because this could mean greater competition for oxygen. High concentrations of cellulose and lignin retard decomposition because only a subset of the microbial population is able to break them down (Vanlauwe et al., 1996).

Three experiments were carried out to determine the effects on acidification of sulfidic material under oxidizing conditions of (i) OM application form [mixed into the soil or placed as layer on the soil surface (Experiment 1)], (ii) OM application rate [0–40 g  $\text{kg}^{-1}$  mixed

into the soil (Experiment 2)], and (iii) C/N ratio of OM [OM with C/N ratios 15–137 mixed into the soil (Experiment 3)]. We hypothesized that the ameliorative effect of OM on acidification during oxidation of sulfidic material will (i) depend on the acidification potential (pyrite concentration) of the sulfidic material (i.e. hyposulfidic or hypersulfidic material), (ii) be greater with OM mixed into the soil than if placed as a layer on the soil surface, (iii) when mixed into the soil will increase with OM addition rate, and (iv) be greater when OM added has a lower C/N ratio.

## 2. Materials and methods

### 2.1. Soils and plant residues

Two sandy sulfidic materials from South Australia (Table 1) with different pyrite concentrations were used. In accordance with the World Reference Base for Soil Resources (IUSS Working Group WRB, 2014) and the Australian Soil Classification (Isbell, 2016) the materials are classified as hypersulfidic and hyposulfidic. The hyposulfidic material (soil Hypo) with low pyrite concentration was collected from a wetland adjacent to the River Murray at Banrock Station (34°12'S, 140°20'E) and the hypersulfidic material (soil Hyper) with higher pyrite concentration from Point Sturt in Lake Alexandrina (35°29'S, 139°2'E) (Table 1). Information about sampling sites as well as soil profile and material classification is given in Table S1. Before the experiments, the soils were dried in a fan-forced oven at 40 °C for 48 h, ground, and sieved to <2 mm. OM was added as plant residues differing in C/N ratio were two mature wheat (*Triticum aestivum* L.) straws (Wa and Wb) and shoots of young kikuyu (*Pennisetum clandestinum*). The residues were finely ground and sieved to <2 mm (for C/N ratios see Table 2).

### 2.2. Experimental procedure

#### 2.2.1. Experiment 1: OM application form

To investigate the effect of the OM addition form in Experiment 1, there were three treatments (with three replicates): control (Ctrl), OM mixed into the soil (OMM), and OM placed as a layer on the soil surface (OML). Dry soil was filled into 70 ml polypropylene containers with 35 g for soil Hypo and 25 g for soil Hyper. Different amounts of soil were used to give approximately the same height of the soil layer in the containers for the two soils (around 2 cm after treatment). Reverse osmosis (RO) water was added to and mixed with soil to adjust the soil water content to 100% of water holding capacity (WHC) (i.e., approximately field capacity, which is considered as the optimum moisture for oxidation of sulfidic material (Jayalath et al., 2016). For treatments OMM and OML, wheat straw Wa was added at 30 g  $\text{kg}^{-1}$  (oven dry basis). For treatment OMM, slightly more water was added than to the control due to the higher WHC in the soil mixed with OM. For treatment OML, the soil water content was adjusted to 100% WHC and dry wheat straw was placed on the soil surface, then RO water was added to the wheat straw layer so that the total amount of water added in treatment OML was the same as in OMM. The height of the moist OM layer was approximately 0.5 cm.

The containers were incubated for 6 weeks in the dark at room temperature (around 25 °C), covered loosely with the lids. Soil moisture was kept constant by adding RO water to maintain the weight every

**Table 1**

Texture, WHC (water holding capacity), pH measured with 1:5 soil to water ratio or at 100% WHC, and concentrations of TOC (total organic carbon),  $S_{\text{KCl}}$  (1 M KCl extractable sulfur), and Scr (chromium reducible sulfur) of soils.

Soil	Depth (cm)	Sand (%)	Clay (%)	Silt (%)	WHC (g $\text{g}^{-1}$ )	pH (1:5)	pH (100% WHC)	TOC (g $\text{kg}^{-1}$ )	$S_{\text{KCl}}$ (mg $\text{kg}^{-1}$ )	Scr (%)
Hypo <sup>a</sup>	5–15	77	13	10	0.18	5.45	5.02	4.8	81	0.02
Hyper <sup>b</sup>	25–40	93	5	2	0.09	6.00	5.65	4.4	153	0.14

<sup>a</sup> Classified as soil (see Table S1) with hyposulfidic material (i.e. soil buffering capacity maintains pH >4).

<sup>b</sup> Classified as soil (see Table S1) with hypersulfidic material (i.e. potential to produce acidic soils with pH <4).

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