



Amount of organic matter required to induce sulfate reduction in sulfuric material after re-flooding is affected by soil nitrate concentration



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ABSTRACT

Acid sulfate soils (ASS) with sulfuric material can be remediated through microbial sulfate reduction stimulated by adding organic matter (OM) and increasing the soil pH to >4.5, but the effectiveness of this treatment is influenced by soil properties. Two experiments were conducted using ASS with sulfuric material. In the first experiment with four ASS, OM (finely ground mature wheat straw) was added at 2–6% (w/w) and the pH adjusted to 5.5. After 36 weeks under flooded conditions, the concentration of reduced inorganic sulfur (RIS) and pore water pH were greater in all treatments with added OM than in the control without OM addition. The RIS concentration increased with OM addition rate. The increase in RIS concentration between 4% and 6% OM was significant but smaller than that between 2% and 4%, suggesting other factors limited sulfate reduction. In the second experiment, the effect of nitrate addition on sulfate reduction at different OM addition rates was investigated in one ASS. Organic matter was added at 2 and 4% and nitrate at 0, 100, and 200 mg nitrate-N kg⁻¹. After 2 weeks under flooded conditions, soil pH and the concentration of FeS measured as acid volatile sulfur (AVS) were lower with nitrate added at both OM addition rates. At a given nitrate addition rate, pH and AVS concentration were higher at 4% OM than at 2%. It can be concluded that sulfate reduction in ASS at pH 5.5 can be limited by low OM availability and high nitrate concentrations. Further, the inhibitory effect of nitrate can be overcome by high OM addition rates.

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1. Introduction

Acid sulfate soils (ASS) have a large distribution in the world, with a total estimated area of coastal ASS of 10⁷–10⁸ ha (Macdonald et al., 2011; Wim and Mensvoort, 2005). Upon oxidation, ASS with sulfidic material (pH > 4.0) transform to sulfuric material (pH < 4.0) (Isbell, 2002) and release acidity and toxic metals, which can have severe negative impacts on the environment (Fitzpatrick et al., 2009). Stimulation of microbial sulfate reduction has been proposed as an efficient strategy for remediating ASS with sulfuric material as the process generates alkalinity (Baldwin and Fraser, 2009; Fitzpatrick et al., 2009). This could be particularly useful when chemical amelioration, such as liming, is costly or ineffective

(Dear et al., 2002). For effective management of sulfuric ASS it is important to understand the factors that may limit or enhance sulfate reduction.

Following re-flooding and soil submersion, soil oxygen is quickly depleted and other oxidized components used as electron acceptors in anaerobic respiration are reduced according to thermodynamics in the sequence: NO₃⁻, Mn(IV), Fe(III), SO₄²⁻, CO₂ (Borch et al., 2009; Ponnampetuma, 1972). The presence of electron acceptors ranked higher in the sequence retards the reduction of electron acceptors ranked lower (Ponnampetuma, 1972) because the former can: (i) outcompete the latter for electron donors (organic carbon) (Hubert and Voordouw, 2007; Lovley and Phillips, 1987), and (ii) oxidize the reduction products of the latter (Canfield et al., 1993; Carlson et al., 2013; Hubert and Voordouw, 2007; Myers and Nealson, 1988; Zhang et al., 2009). Additionally, nitrate inhibits sulfate reduction because the product of nitrate reduction, nitrite,

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inhibits the dissimilatory sulfite reductase (Haveman et al., 2004; Kaster et al., 2007). The inhibitory influence of Mn(IV) and Fe(III) oxides on sulfate reduction (Canfield et al., 1993; Lovley and Phillips, 1987; Myers and Nealson, 1988; Zhang et al., 2009) is weaker because of (i) their low solubility, (ii) lower standard reduction potential of these redox couples than the NO_3^-/N_2 couple, and/or (iii) the limited number of microbes using these electron acceptors (Ponnamperuma, 1972).

Large-scale exposure and oxidation of ASS occurred in the Lower Murray region of South Australia during a recent prolonged and severe drought (Mosley et al., 2014b). Despite several years of re-submergence after the end of the drought, neutralization of the acidity in these soils via sulfate reduction does not appear to be occurring (Mosley et al., 2014a). In our previous study (Yuan et al., unpublished) we showed that low soil pH and availability of organic matter (OM) may limit sulfate reduction in these ASS with sulfuric material after re-flooding. But the ameliorative effect of the treatment with increased pH and OM addition on sulfate reduction differed among soils (Yuan et al., unpublished). Possible reasons for a small increase of sulfate reduction compared to the other soils could be high concentrations of competing electron acceptors, particularly nitrate as a large proportion of ASS in the LMRIA are under intensive pasture with high nitrogen fertilizer use. High nitrate concentrations have also been reported for other ASS (Macdonald et al., 2010). The influence of competing electron acceptors, such as nitrate has not been adequately studied in a systematic manner and is currently not taken into account for ASS management. The aim of the present study was to investigate the effects on sulfate reduction in sulfuric material after pH adjustment to 5.5 and re-flooding of: (i) addition rates of OM (Experiment 1) and (ii) combinations of amendment with nitrate and OM (Experiment 2). Our hypotheses were: (i) sulfate reduction increases with OM addition rate, and (ii) nitrate can inhibit sulfate reduction but this can be overcome by high OM addition rates.

2. Materials and methods

2.1. Soils

Five soils with sulfuric material (soils 1–5; Table 1) collected in the Lower Murray Reclaimed Irrigation Area (LMRIA), South Australia ($35^\circ 7' 28.05''\text{S}$, $139^\circ 17' 55.17''\text{E}$) were used. They come from two profiles described in Fitzpatrick et al. (2013): DSa01A (soils 1–4) and DSa01B (soil 5). The soil is a sulfuric clay soil (Fitzpatrick et al., 2010; Fitzpatrick, 2013), Typic Sulfaquert or Sulfic Sulfaquert (Soil Survey Staff, 2014). The area is used for intensive cattle grazing with flood irrigation (for further information see Mosley et al., 2014a). Selected soil properties were determined (Table 1).

2.2. Experimental procedure

Soils 1–4 were used in Experiment 1. In our previous study (Yuan et al., unpublished), these clayey soils had a smaller increase in sulfate reduction compared with other soils after pH adjustment to 5.5 and OM addition. Twenty grams of moist soil (12–15 g oven dry soil) was mixed with 20 ml reverse osmosis (RO) water in 70 ml polypropylene containers. Organic matter (ground and sieved mature wheat straw to <2 mm with 313 g kg^{-1} total organic carbon and 7 g kg^{-1} total nitrogen) was then added to the soil paste at three rates: 2, 4 or 6% (w/w on oven dry basis). Controls were soils without OM addition. For soil 1 only, addition rates of 0.5 and 1% were also included, because in our previous study (Yuan et al., unpublished) OM was added at 2% only and more black material (likely monosulfidic, FeS) was observed in this soil compared to soils 2–4. It was unclear if this apparent sulfate reduction would occur with lower OM additions. After adjusting the soil pH in all treatments to 5.5 ± 0.1 by adding 1 M NaOH, the soils were incubated under 40 ml RO water for 36 weeks. There were three replicates per treatment. During incubation, pH of the overlying water and soil pore water were measured weekly from weeks 0–12 and every two weeks from weeks 12–36 (except in week 34) (Figs. S1 and S2). The concentration of O_2 in the overlying water was determined at the end of the experiment.

Only one soil (Soil 5) was used in Experiment 2. This soil was selected because it had low initial nitrate concentration and in our previous study with 2% OM added and pH increased to 5.5 black (likely monosulfidic) material occurred early when incubated under water (Yuan et al. unpublished). There were three factors: organic matter addition (OM), nitrate addition (Nitrate), and time (Time). Twenty grams of moist soil (15 g oven dry soil) was mixed with 20 ml RO water in 70 ml polypropylene containers. Ground and sieved mature wheat straw (similar as in Experiment 1) was then added at two rates: 2% and 4% (w/w on oven dry basis). After 1.1, and 2.1 ml 0.1 M KNO_3 was added to achieve 100, and 200 mg Nitrate-N kg^{-1} . The control was without nitrate addition. The lower nitrate addition rate represents approximately the highest nitrate-N concentration in the soils used (Table 1) and the higher addition rate twice that concentration. To compensate the change of salinity caused by KNO_3 addition, 2.1, 1.1, and 0 ml 0.091 M KCl, which has the same salinity as 0.1 M KNO_3 (Weast et al., 1988), was added to the soil for control, 100, and 200 mg Nitrate-N kg^{-1} , respectively. Soil pH for all the treatments was then increased to 5.5 ± 0.1 by adding 1 M NaOH. The soil was incubated under 40 ml RO water for 2 weeks. This short incubation period was chosen firstly because nitrate can be reduced within a few days after submergence (Ponnamperuma, 1972), therefore the effect of nitrate as competing electron acceptor for sulfate reduction is likely to be greatest initially. Secondly, we observed that the difference in extent of black material among nitrate addition rates that developed in the

Table 1
Texture, pH and concentrations of TOC (total organic carbon), S_{HCl} (4 M HCl extractable sulfur), and S_{KCl} (1 M KCl extractable sulfur) of soils used in this study.

No.	Depth (cm)	Sand (%)	Clay (%)	Water content (g g^{-1})	pH (1:1)	TOC (%)	Nitrate-N (mg kg^{-1})	Ammonium-N (mg kg^{-1})	S_{HCl} (%)	S_{KCl} (%)
<i>Experiment 1^a</i>										
1	57–95	1.3	75.3	0.33	4.30	3.0	13.0	48.3	0.32	0.26
2	95–190	1.9	53.7	0.54	3.09	2.3	0.7	117.7	0.64	0.42
3	190–280	1.6	56.1	0.63	3.21	1.6	16.5	125.2	0.55	0.37
4	280–350	1.8	58.7	0.40	4.23	1.5	93.9	51.2	0.40	0.29
<i>Experiment 2^b</i>										
5	76–190	5.5	56.2	0.33	4.06	1.5	10.6	40.2	0.24	0.14

^a Profile: DSa01A (Fitzpatrick et al., 2013).

^b Profile: DSa01B.

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