



Iron and sulfur cycling in acid sulfate soil wetlands under dynamic redox conditions: A review

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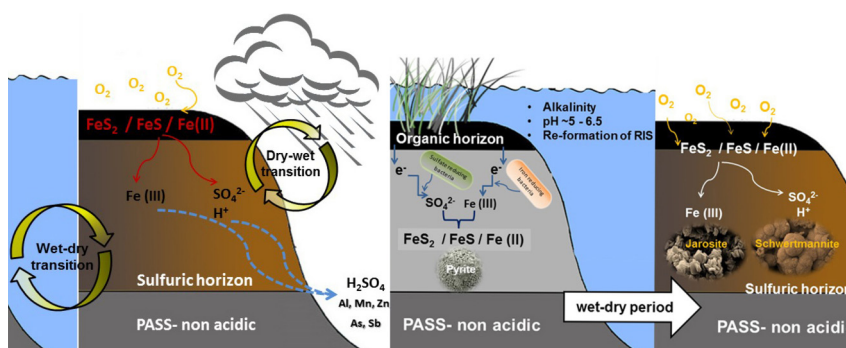
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HIGHLIGHTS

- Potential acid sulfate soils (PASS) contain substantial quantities of sulfidic minerals.
- Oxidation of sulfidic minerals in ASS environments generates acidity and secondary Fe(III) minerals.
- Seasonal redox oscillations can affect Fe and S speciation in ASS environments.
- Transformation of Fe and S mineral phases may regulate metalloids mobility in ASS environments.
- Dynamic redox can drive complex biogeochemical processes in fresh-water re-flooded ASS.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 14 November 2017
Received in revised form
19 January 2018
Accepted 21 January 2018

Keywords:

Acid sulfate soils
Iron
Sulfur
Dynamic redox
Arsenic
Antimony

ABSTRACT

Acid sulfate soils (ASS) contain substantial quantities of iron sulfide minerals or the oxidation reaction products of these sulfidic minerals. Transformation of iron (Fe) and sulfur (S) bearing minerals is an important process in ASS wetlands with fluctuating redox conditions. A range of potentially toxic metals and metalloids can either be adsorbed on or incorporated into the structure of Fe and S bearing minerals. Therefore, transformation of these minerals as affected by dynamic redox conditions may regulate the mobility and bioavailability of associated metals/metalloids. Better understanding of the interaction between Fe/S biogeochemistry and trace metal/metalloid mobility under fluctuating redox conditions is important for assessing contaminant risk to the environment. This review paper provides an overview of current knowledge regarding cycling of Fe, S and selected trace metal/metalloids in ASS wetlands under fluctuating redox conditions and outlines future research challenges and directions on this subject.

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1. Introduction

A complex interplay between hydrology, redox conditions and iron (Fe) and sulfur (S) mineralogy leads to diverse reactions that

determine contaminant fate and general water quality in acid sulfate soil (ASS) wetlands. While many of the Fe and S-bearing minerals that commonly occur in ASS-affected environments are capable of being potent scavengers for potentially-toxic trace metals and metalloids under oxidising acidic conditions, many of these minerals are also metastable and prone to change under reducing conditions. Some trace metals and metalloids such as arsenic (As) may have a large impact on the environment.

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Therefore, a sound understanding of metastable host-mineral phases and the corresponding behaviour of associated trace metal/metalloid contaminants under fluctuating redox conditions is essential for ASS wetlands management.

This review focuses on the formation, characteristics and remediation of ASS wetlands with a particular emphasis on key Fe and S minerals and their behaviour under both oxidising and reducing conditions and the consequences for potentially toxic trace metals and metalloids behaviour under dynamic redox conditions.

1.1. Overview of acid sulfate soils

Acid sulfate soils generally contain substantial quantities of Fe sulfide minerals or the oxidation reaction products of these sulfidic minerals (e.g. acidity and secondary Fe(III)-containing minerals) (Dent, 1986; Dent and Pons, 1995; Fanning et al., 2002).

The global extent of ASS has been estimated at ~50 million ha and includes both inland and coastal ASS in Asia, Australia, Scandinavia and west Africa (Fig. 1) (Ljung et al., 2009). Although pyrite (FeS_2) is the typically most prevalent Fe sulfide mineral in undisturbed ASS materials, other sulfide minerals such as greigite, marcasite and iron monosulfides such as mackinawite, may also be abundant in ASS environments (Bush et al., 2004; Dent, 1986; Van Breemen, 1973). Undisturbed, unoxidised sulfide-rich soils which have the potential to become acidic are commonly termed potential acid sulfate soils (PASS) and they are usually stable under anoxic and waterlogged conditions (Sullivan et al., 2009). However, a wide range of anthropogenic activities (e.g. drainage and excavation) and natural phenomena (e.g. drought) can cause exposure and oxidation of PASS and generate significant quantities of acid (Dent, 1986; Sullivan et al., 2008). The formation of actual acid sulfate soils (AASS) occurs when the magnitude of sulfuric acid surpasses the

buffering capacity of the sediments (Boman et al., 2010; Johnston et al., 2009a; Pons et al., 1982). Therefore, one of the foremost ASS management strategies is to identify the PASS materials to avoid exposure, oxidation and further environmental problems, and then to develop appropriate management options for these soils (Broughton, 2008; Kraal et al., 2013; Morgan et al., 2012; Sullivan et al., 2009).

1.2. Environmental hazards of ASS

Oxidation of reduced inorganic sulfur (RIS) species and formation of AASS has an enduring and complex effect on the soil and surrounding environment (Corfield, 2000; Rosicky et al., 2006). Some of the major environmental hazards associated with ASS that are related to the oxidation of RIS include acute acidification of soils and waters, aluminium (Al) toxicity, trace metal/metalloid mobilisation, de-vegetation of the soil, deoxygenation of water bodies and the formation of toxic sulfur gases (e.g. H_2S) (Huang et al., 2011).

The adverse environmental and ecological impacts of acute acidification and associated metals mobilisation on agricultural crops, terrestrial, and aquatic environments have been documented in many former studies (Callinan et al., 2005; Sammut et al., 1996; Wilson et al., 1999; Mosley et al., 2014). Severe acidification of ASS and resultant decrease in soil pH can significantly enhance the solubility and mobility of many metals following re-wetting of oxidised sediments [e.g. Al, Fe, zinc (Zn), nickel (Ni), copper (Cu) and vanadium (V)] (Åström, 2001; Macdonald et al., 2004; Simpson et al., 2010; Fitzpatrick et al., 2017; Michael et al., 2017).

For instance, Al solubility is highly pH-dependent, and potentially toxic amounts of dissolved Al are common in ASS drainage waters below pH 5.0. High aqueous concentrations of Al can disrupt plant root systems and can affect plant growth (Sammut et al.,

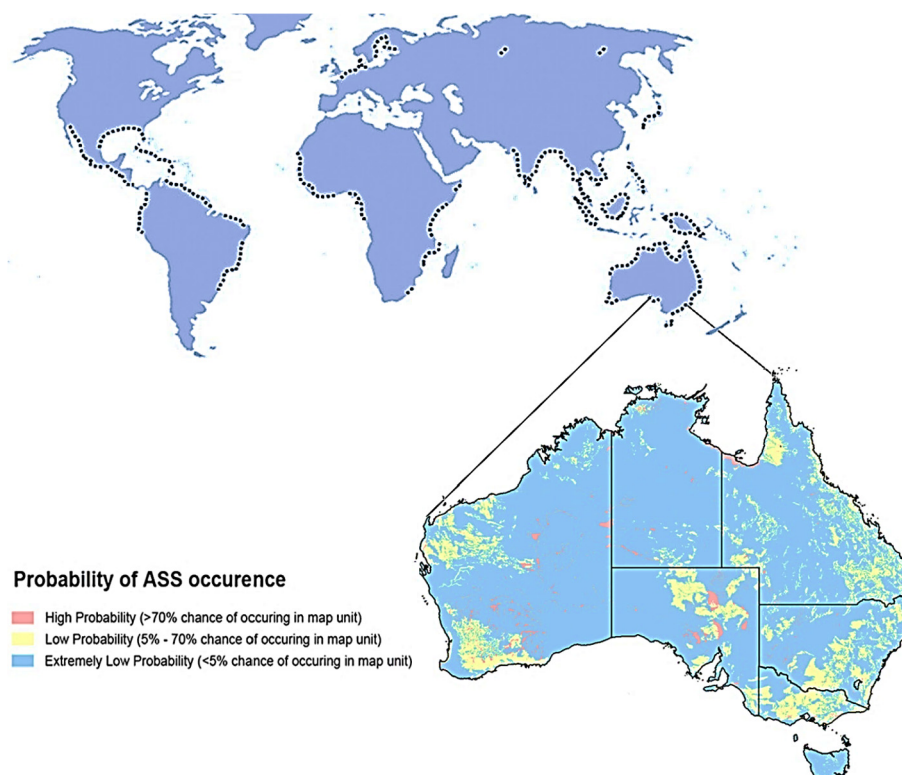


Fig. 1. Worldwide distribution of coastal ASS [after (Ljung et al., 2009)] and the probability of the ASS occurrence in Australia (Source: S. Marvanek, CSIRO).

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