



Estuarine dynamics and acid sulfate soil discharge: Quantifying a conceptual model



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ARTICLE INFO

Keywords:

Wetland
Acidification
Floodplain restoration
Estuarine dynamics
Conceptual model

ABSTRACT

Over 170,000 km² of Acid Sulfate Soils (ASS) have been identified worldwide, with 65,000 km² in Asia; 45,000 km² in Africa; 30,000 km² in Australia; 30,000 km² in South America; and a combined total of 10,000 km² in Europe and North America. A state-of-the-science approach to remediate these soils is to encourage tidal buffering. This approach involves the removal of drainage infrastructure and the repurposing of agricultural land back to tidal wetlands. To date, remediation efforts have largely focused on individual locations at the farm plot scale. Remediation projects have commonly focused on onsite acidity levels, groundwater transport and impacts to agricultural infrastructure. This site-based approach provides detailed information on acid hotspots, but provides limited understanding of the overall issues affecting the broader coastal floodplain.

A catchment-wide estuarine dynamics approach has been developed and field tested to overcome current limitations. The catchment-wide approach combines onsite investigations with large-scale studies of: (i) the fate/transport of acidic plumes, (ii) the coastal floodplain response dynamics to rainfall or flooding events, and (iii) the objective prioritisation of impacted landscapes for remediation. This paper provides a detailed description of the catchment-wide approach supported by conceptual processes and a detailed case study. The 20 km² case study site highlights the importance of local and catchment processes in guiding rehabilitation plans.

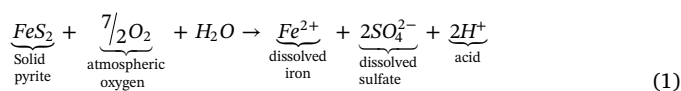
1. Introduction

1.1. Acid production

Early experiences with Acid Sulfate Soils (ASS), formerly known as ‘cat clays’, date back to the drainage schemes of the 17th century in the Netherlands, and the late-19th century in Australia. In fact, the long-term problem of ASS in the polders of the Netherlands provided a major resource for their identification worldwide (Rickard, 2015). To date, over 170,000 km² of ASS have been identified worldwide, with 65,000 km² in Asia; 45,000 km² in Africa; 30,000 km² in Australia; 30,000 km² in South America; and a combined total of 10,000 km² in Europe and North America (Rickard, 2015).

Soils that contain iron sulfides are commonly referred to as ASS (Department of Resources and Environment (DERM), 2009). Pyrite (FeS₂) is the most abundant iron sulfide found in the coastal regions of the world and formed during the last major sea level rise (approximately 6500 years ago) (Berner, 1984). Under most natural conditions, where these soils remain waterlogged, ASS remain innocuous, and are commonly referred to as Potential Acid Sulfate Soils (PASS) (Blunden

and Indraratna, 2000). When PASS are exposed to atmospheric oxygen, the pyrite reacts to produce large quantities of low pH (pH < 4.5) sulfuric acid (H₂SO₄) (Dent and Pons, 1995). The pyrite oxidation process is complex, however, in short, the reaction involves the conversion of pyrite to ferrous iron and sulfate in the presence of oxygen and moisture (White et al. 1997; Glamore, 2003), and is summarised by Eq. (1):



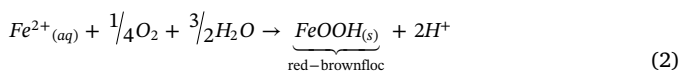
The production of acid by pyrite oxidation significantly alters the chemical composition of the soil matrix. As pH levels fall, acidified groundwater is transported above the pyritic layer due to capillary rise from evapotranspiration, or from an increase in the water table due to rainfall. In this way, the soil matrix above the pyritic layer can become acidified over time. The acidification process changes the solubility of many iron oxidation products and the dissolution of clay minerals, such as aluminium (Indraratna et al., 2002, 2010). In particular, at low pH levels, aluminosilicate clays release aluminium that remain soluble and

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form aluminium hydroxy ions through hydrolysis that also contribute to soil acidity (Glamore, 2003).

In acidic groundwater, Fe(II) produced in Eq. (1) can further oxidise to Fe(III), and in the process, remove one of the H^+ ions. The rapid conversion of Fe(II) to Fe(III) and the subsequent precipitation of iron oxyhydroxides can lead to the rapid acidification and deoxygenation of natural waters (Jones et al., 2014), as demonstrated in Eq. (2) below. This process is commonly known as ‘acid at a distance’ (Glamore, 2003), because it occurs downstream of the oxidation source and creates a further two moles of acid.



Construction of deep (> 0.5 m) drainage systems and flood mitigation infrastructure on coastal floodplains has increased the generation and export of acidity from ASS (Johnston et al., 2003). The release of acidic runoff is exacerbated by floodgates at the tidal junction of drainage networks, which prevent tidal waters from inundating low-lying areas of the floodplain (Glamore, 2003). By restricting the intrusion of tidal waters into the drainage system, the floodgates create a reservoir of acidic water. Floodgates also maintain drain water levels at the low tide mark and create a strong hydraulic gradient between the groundwater and the drain (Glamore and Indraratna, 2001). This results in the transport of acid from the groundwater to the drainage channel and onwards to the estuary, particularly following moderate to large rainfall events (Johnston et al., 2003; Rayner et al., 2015).

Worse than the habitat destruction from the loss of wetlands worldwide (White et al., 1997), the wide-scale drainage of coastal floodplains has exposed an insidious acid problem that severely impacts the wider estuary. The prolonged drying of the floodplain has allowed oxygen to penetrate into the sulphur-rich sediments creating acidified soils and groundwater. The subsequent acidification of waterways severely degrades whole estuarine ecosystems, and causes mass fish kills (Sammot et al., 1995, 1996; Pollard and Hannan 1994; Sullivan and Bush 2004; Lin et al., 2004; Kroon and Ansell 2006), attacks healthy oysters (Dove 2003; Smith et al., 1999) and covers the riverbed with toxic metals (Rickard 2015). Indeed, accelerated soil acidification globally poses unique land and water management challenges for floodplain managers to reduce the ongoing impacts of acid discharge from drained coastal ASS.

1.2. Remediation techniques

Research (Indraratna et al., 2002) has shown that it is very difficult to limit further acid production once the soil matrix is acidified. Since the majority of drained coastal ASS are already acidic, most remediation strategies focus on containment, dilution or neutralisation. Containment can lead to substantial acid build up and inhibit the movement of aquatic life, whilst the addition of agricultural lime can be costly (Luke 2016). However, several on-ground project examples using dilution or neutralisation as remediation techniques (Karimian et al., 2017; Lu et al., 2016; Johnston et al., 2011; Johnston et al., 2009; Golab and Indraratna 2009; Atkinson et al., 2003; Glamore 2003) have effectively reduced acid drainage at the source. A brief description of freshwater reflooding and tidal buffering are provided below for reference.

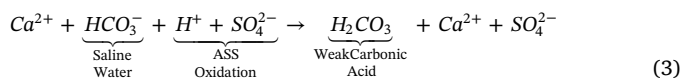
Remediation via dilution (or freshwater reflooding) involves reducing the concentration of a volume of acid (low pH) by adding a larger volume of freshwater (neutral pH). This strategy is based on the concept that the pH of water increases by one pH unit for every ninefold increase in the volume of freshwater, as pH is measured on a logarithmic scale (i.e. $pH = -\log_{10}[H^+]$). For example, a pH of 5 has a concentration of hydrogen (H^+) ions 10 times greater than a pH of 6. A flow dilution scenario can include re-instating wetting-drying patterns to increase soil moisture through dilution with freshwater flows. However, due to

the large volumes of acid generated by pyrite oxidation, available floodwaters may not be sufficient to dilute existing acid stored in the soil matrix (Glamore and Indraratna, 2004).

For example, in the Macleay River, on the eastern seaboard of New South Wales (NSW), Australia, it is not uncommon for the equivalent of 200 or more Olympic swimming pools of acidic water (e.g. concentrations of approximately 6.3×10^{-6} $\mu\text{mol/L}$) to be discharged into the river after large rainfall events. Furthermore, remediated wetlands by freshwater reflooding generates alkalinity and leads to the formation of reduced inorganic sulfur (RIS) and Fe(II) species in the surface soil layers, which are prone to rapid acidification during future droughts (Karimian et al., 2017). For these reasons, simply reflooding drained coastal ASS with freshwater may not resolve the problem of acid flux from surface waters (Glamore and Indraratna, 2004, 2009; Johnston et al., 2014; Karimian et al., 2017).

However, the risks of widespread acidic contamination to the estuary is often offset in highly flushed estuaries with a high tidal buffering potential (Glamore and Indraratna, 2001, 2004). This is based on the premise that seawater has an infinite natural supply of bicarbonate/carbonate ions, and incoming tides transport these acid buffering agents throughout an estuary (Stumm and Morgan, 1996). Indeed, Rayner et al. (2015) showed that the tidal buffering capacity of an estuary is directly proportional to the volume of buffering agents (i.e. alkalinity) within the system. Furthermore, the relationship between bicarbonate and salinity at a single site on the Shoalhaven River in south-eastern NSW, Australia, was documented by Glamore (2003) and shows a distinct correlation between the buffering capacity (HCO_3^-) and electrical conductivity (EC) in an estuary.

Tidal buffering occurs when acidic water containing hydrogen protons (H^+) reacts with bicarbonate (HCO_3^-) or carbonate (CO_3^{2-}) available in seawater. The key factor in tidal buffering is the reduction of H^+ ions to weak carbonic acid (Eq. (3)). While the focus here is on surface water quality and dynamics, it is worth noting that gypsum ($CaSO_4$) may form as a precipitate in Eq. (3), and this may need to be considered in terms of the potential environmental impacts associated with remediation.



Remediation via tidal buffering involves reflooding coastal ASS or adjacent drainage creeks, with brackish or saline water (Glamore and Indraratna, 2001; Johnston et al., 2004; Johnston et al., 2009). This typically involves the removal of drainage infrastructure and the repurposing of agricultural land back to tidal wetlands. For example, various studies (Glamore and Indraratna 2004; Johnston et al., 2009; Glamore and Indraratna 2009; Rayner et al., 2015) have shown that a tidal buffering strategy immediately improved drain water quality (i.e. pH, DO) and was most effective during prolonged dry periods when the incoming tide is super-saturated with bicarbonate, providing a large source of alkalinity to the affected areas. Luke (2016) has also trialled a lime-assisted tidal exchange (LATE) remediation strategy, whereby, agricultural lime was added to the incoming tide to support the natural remediation process, and also added to the out-going exit waters to prevent acid-flush into estuarine waters. However, following six years of gradually increasing tidal inundation, it was found that in-situ microbial and tidal exchange processes accounted for 99% of the change, whilst the addition of agricultural lime contributed less than 1% (Luke 2016).

Furthermore, recent studies (Johnston et al., 2011; Burton et al., 2011) highlight the large-scale changes to hydro-geochemical processes resulting from regular tidal inundation of remediated wetlands. These studies show that marine derived HCO_3^- can initiate landscape-scale shifts in the biogeochemical cycling of iron and sulfur, resulting in pyrite reformation. For example, reductive dissolution of Fe(III) minerals (e.g. jarosite ($KFe_3(SO_4)_2(OH)_6$) and goethite ($FeOOH$)) leads to

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