



Fate and dynamics of metal precipitates arising from acid drainage discharges to a river system

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HIGHLIGHTS

- Neutralisation of acid drainage produces metal-rich oxide precipitates.
- The fate of these in a river system under oxic and anoxic conditions was assessed.
- Contamination was observed in a localised area near the drainage discharge.
- Ratios of reactive to total recoverable metal were elevated in this zone.
- Deoxygenation of overlying surface water enhanced metal release.

GRAPHICAL ABSTRACT



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ABSTRACT

Neutralisation of acid drainage creates metal-rich precipitates that may impact receiving water bodies. This study assessed the fate of over seven years of acid drainage discharges on the sediments of the lower River Murray (Australia), including the potential for periodic water anoxia to enhance risk via reductive dissolution of amorphous (Fe, Mn and co-precipitated and bound metal) oxide phases. With the exception of one site with restricted water exchange, elevated reducible/reactive metal(oid) (Fe, Ni, As, Co, Zn) concentrations were only observed in the localised wetland-riparian area within approximately 100 m of the discharges. Only a minor exceedance of national sediment quality guideline values occurred for Ni. In the main river channel, elevated reactive metal (Fe, Mn, Ni, Zn) concentrations were also only observed less than approximately 100 m from the drainage discharge point. This appears due to (a) rapid neutralisation of pH leading to metal precipitation and deposition in the localised discharge area, and/or (b) dilution of any metal precipitates entering the main channel with natural river sediments, and/or (c) flushing of precipitates downstream during higher flow conditions. The influence of deoxygenation on metal release was profound with large increases in the concentration of dissolved Fe, Mn, Zn, Ni, and As in the overlying water during laboratory experimental simulations. However, given *in situ* sediment metal contamination is very localised, it appears on a river reach scale that the acid drainage precipitates

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will not significantly contribute, over and above, the background release of these metals during these conditions.

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1. Introduction

Acid drainage from mine sites and acid sulfate soils can create severe impacts on receiving waterways and their ecosystem services (Chapman et al., 1983; Mosley et al., 2014a; Beane et al., 2016; Grande et al., 2018). As well as low pH (<5), these discharges typically contain high dissolved metal concentrations (in the $\mu\text{g L}^{-1}$ to mg L^{-1} range) due to acid-promoted dissolution of natural mineral phases. Aluminium (Al), Mn and Fe are often found at highly elevated concentrations due to their dominant presence in primary and secondary mineral phases in soils and rocks (e.g. Al, Fe, Mn oxides and pyrite, FeS_2). Trace metals (e.g. Cd, Cu, Ni, Zn) and metalloids (As) can also be found at elevated concentrations as they are commonly co-precipitated with minerals or bound to surface oxide phases that undergo acid dissolution.

If sufficient dissolved carbonate and bicarbonate alkalinity is present in receiving water bodies to neutralise the acid discharges, large amounts of Fe and Mn may precipitate as amorphous oxide solid phases and these also adsorb or co-precipitate trace metals such as As, Cu, Cd, and Zn (Balistrieri et al., 2007; Nystrand et al., 2012; Simpson et al., 2013; Mosley et al., 2014a). These metal-rich solid and colloidal phases can be transported downstream or deposited on sediments, often producing the characteristic brown “ochre” precipitate coating of stream beds and other surfaces affected by acid drainage (Chapman et al., 1983).

Fe and Mn oxide solid phases are redox sensitive and anoxia may lead to the release of metals from the surface sediment through reductive dissolution reactions (e.g. $\text{Fe(III)}_{(s)}$ to $\text{Fe(II)}_{(aq)}$, Balistrieri et al., 1992). Reducing (sub-oxic to anoxic) conditions can often be found in sediments but can also form in the water column of lakes and rivers, in particular under low flows when water stratification may occur (Maier et al., 2001), or high flows when organic carbon is mobilised from floodplains (Whitworth et al., 2013). Anoxia in water columns and surface sediments could be of particular concern in the case of freshwater environments receiving acid drainage, due to the presence of larger amounts of Fe and Mn oxide precipitates (Simpson et al., 2013; Mosley et al., 2014a). Previous research in acid mine and acid sulfate soil drainage affected systems has focused mainly on metal concentrations and dynamics in surface waters and sediments under oxygenated (oxic) conditions (Chapman et al., 1983; Balistrieri et al., 2007; Mosley et al., 2014a). To improve assessment of the potential environmental risk arising from acid discharges in systems which undergo anoxia, an understanding of the redox-sensitive metal binding phases and potential for release of metal contaminants from these under anoxic conditions is required. Selective extractions can be used to target specific sediment phases (e.g. Fe and Mn oxides) to improve assessment of metal availability under varying environmental conditions (Linge, 2008), but these have received limited attention for assessing acid drainage impacts (Sarmiento et al., 2011; Nordmyr et al., 2008; Beane et al., 2016).

Soil acidification occurred on the irrigated floodplains of the lower River Murray in South Australia due to falling river and ground water levels during an extreme “Millennium” drought from 2007 to 2010 (Mosley et al., 2014a; b; Fitzpatrick et al., 2017a). This resulted in deep drying and cracking of clay soils and oxidation of sulfidic (pyrite, FeS_2) soil materials to form sulfuric ($\text{pH} < 4$)

materials. Following the end of the drought, drains in the farm fields that catch irrigation return flows and shallow groundwater, turned acidic ($\text{pH} 2\text{--}5$) and contained high dissolved metal (Al, Fe, Mn, Ni, Zn) concentrations (Mosley et al., 2014a). Discharges from these drains to the river (required practice for farm water management) created a significant challenge and risk to downstream water supplies and ecosystems (Frizenschaf et al., 2015). Following discharge of the acid drainage into the river system, the water pH is neutralised rapidly in the immediate mixing zone in the River Murray with resulting reduction in dissolved metal concentrations (Simpson et al., 2013; Mosley et al., 2014a, 2015). A recent study showed the soils in the region were still highly acidic 7 years after the drought ended with continued pumping of acid drainage during this time (Mosley et al., 2017). A key uncertainty in assessing the long term environmental risk posed by these discharges is the fate of metal precipitates formed during this neutralisation process in the river, including during anoxic conditions.

The aim of this study was to investigate the distribution and fate of reactive sediment metal phases under oxic and anoxic conditions in the Lower River Murray region. It was hypothesised that (1) the influence of the acid drainage will be able to be discerned via assessment of the relative amount of metal(loid)s associated with Fe and Mn oxide phases, and (2) these phases will dissolve to release metals under reducing conditions. The information will be used to assess the potential environmental risks, and plan mitigation and management options to reduce impacts on human health and the environment. The techniques and results have broader significance to other water systems that receive acid drainage and undergo temporary or permanent anoxia.

2. Methods

2.1. Study area description

The study area is at the terminal end of the River Murray system, and comprises about 5200 ha of historical floodplain that was reclaimed for agriculture in the early 1900s. The region is known as the Lower Murray Reclaimed Irrigation Area (LMRIA) and comprises 27 individual irrigation areas between Mannum and Wellington in South Australia (see Fig. 1). Of these areas about 14 had drainage waters that had turned acidic due to sulfidic soil exposure in the extreme drought period between 2007 and 2010 (Mosley et al., 2014a; b).

The drainage discharges are pumped into localised mixing zones, typically a shallow (<2 m deep) and narrow (<20 m radius) cleared channel in the riparian-wetland zone of the river. The shallow riparian-wetland zone extends along the river bank and contains large amounts of wetland vegetation. Following direct or diffuse passage through the riparian zone, the drainage water enters the main river channel of the River Murray. The River Murray in this region is a wide (approx. 200–300 m), moderately deep (typically 5–20 m), turbid and slow moving river system. There is no significant light penetration to the bottom of the river channel, which is typically devoid of aquatic vegetation.

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