



Metal enrichment in estuarine sediments proximal to acid sulfate soils as a novel palaeodrought proxy



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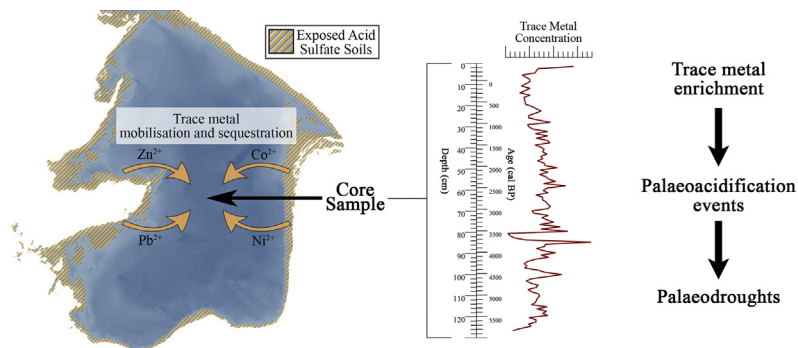
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HIGHLIGHTS

- Correlation to regional climate proxies suggests drought as a major driver of metal flux into the system.
- Trace elements sequestered following acidification events established to be a novel palaeodrought proxy.
- Recent unprecedented acidification likely to be a result of regional water management decisions

GRAPHICAL ABSTRACT



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ABSTRACT

Persistent drought over eastern Australia at the turn of the last millennium reduced stream flow in Australia's largest and most economically important drainage basin. Low water levels in the basin's terminal lakes triggered widespread pyrite oxidation, which altered surface water chemistry and released metals into the environment. The frequency of these events, and the links between drought and acid sulfate soil activation, are not known because the historical and instrumental records are short. Here, we present a Holocene-aged record of trace metal enrichment from Lake Albert—part of the terminal Lower Lakes system of the Murray-Darling Basin, Australia—that demonstrates the potential of trace elements mobilised during acidification events for palaeodrought reconstructions.

Symptomatic metals were measured from a core of clayey sediment to form a multi-element assemblage that reveals acidification events in the geological past. Correlation with regional climate proxies suggests that climate forcing is significant in driving metal flux to estuarine sinks in acid sulfate soil rich landscapes, although the intensity of a drought is not the only variable responsible for acidification intensity.

The constructed record indicates that regional climate moved from a generally humid climate phase with intermittent droughts, to a more arid climate at ~5.2 ka which prevailed until ~1.7 ka. Following conditions were relatively wet with low climatic variability through till European Settlement. Enrichment is observed coincident with both the 1982–83 drought and Millennium Drought, the latter of which reaching an intensity unsurpassed in the last ~4.8 ka, likely a product of anthropogenic changes to the Lower Murray-Darling Basin system.

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1. Introduction

Acid sulfate soils (ASS) are primarily distinguished by a significant natural concentration of sulfidic minerals, frequently pyrite (FeS_2),

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which have the potential to be oxidized in aerobic conditions to form sulfuric acid creating severely acidic conditions; and are subsequently classified in relation to the chemical nature of the sulfidic material and degree of oxidation experienced (Dent, 1986; Isbell, 2016). Acid sulfate soils form in anaerobic marine environments with high concentrations of sulfate, iron oxyhydroxides and organic matter, and are abundant in saline or brackish coastal environments such as estuaries, mangroves, marshes and peat lands (Berner, 1984; Hall et al., 2006).

The initial exposure of sulfidic material to oxygen can be attributed to anthropogenic or natural processes, most commonly relating to the lowering of surface water levels or exhumation. This commonly occurs naturally as a result of reduced stream flow or lowered groundwater levels associated with periods of low precipitation (Dent and Pons, 1995), but may also be a result of geomorphological processes (e.g. river channel migration) or, as observed in Finland, isostatic uplift (Boman et al., 2010). Anthropogenic drivers of sulfide oxidation are primarily due to abstraction of surface and ground waters for agricultural irrigation, exhumation of soils during construction, and the suppression of peak river flows by upstream weirs, locks and barrages (Baldwin, 2011; Sammut et al., 1996).

The oxidation of sulfidic material results in the formation of an assortment of minerals in situ, as well as facilitating the release of various elements into proximal environments (Åström and Åström, 1997; Åström and Deng, 2003; Bigham and Nordstrom, 2000). Metals bound to pyrite are mobilised during the breakdown of pyrite during oxidation, along with the inherent Fe^{2+} . It is however identified that this contribution to mobilised metal content is relatively insignificant in comparison to mobilised elements from full/partial dissolution of aluminosilicates, sulfides, carbonates and organic material as a result of acidified conditions (Österholm and Åström, 2004; Sohlenius and Öborn, 2004).

The behaviour and ultimate fate of released elements is dependent on both the chemical properties of individual elements as well as the specific chemical environment of the system, and is therefore complex and hard to predict. Most studies indicate that, as element-enriched acidified surface waters return to near neutral pH, mobilised elements are sequestered from solution as adsorbed or coprecipitated species with freshly precipitated iron and/or aluminium oxyhydroxides, or organic matter, or precipitated in reduced conditions as discrete sulfide minerals (Åström and Björklund, 1995; Morse and Luther, 1999; Nystrand et al., 2016; Simpson, 2008). Elements Al, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Cd, and Pb have all been observed to be enriched in acidified surface and pore waters, and in surficial sediments proximal to oxidized ASS (Broughton, 2008; Johnston et al., 2016; Macdonald et al., 2004; Nordmyr et al., 2008a; Nordmyr et al., 2008b).

The dynamic process of element dissolution, transportation, and sequestration from ASS is well known, due in part to the disastrous identified and experienced environmental impacts from ASS activation. In Australia, ASS have become recognised as a natural hazard (Fitzpatrick, 2003) following large scale acidification experienced during the 1995–2009 ‘Millennium Drought’ (Mosley et al., 2014; Ummenhofer et al., 2009). While reductions in regional precipitation, and by extension climatic variability, have been identified as an instigator of acidification (Fitzpatrick et al., 2008b; Simpson et al., 2010), just how frequently this drought-forcing of ASS oxidation occurs is poorly understood. Additionally, whether ASS systems demonstrate threshold behaviour in their response to drought forcing needs exploration. These data may be critical in terms of our ability to mitigate potential risk and to develop adaptive policy frameworks in the context of projected climate change, particularly in regions like Australia where freshwater resources are already under stress. Here, we hypothesise that elements mobilised from ASS can be used as a geochemical signal for past acidification events. This is internationally significant, as the ability to identify acidification events in sediment cores from ASS rich landscapes enables the observation of acidification events over hundreds or thousands of years, far beyond the historical or instrumental

record. Further, by comparing these long-term models of acidification response to existing palaeoclimatic data it may be possible to assess the response of ASS systems to climate forcing of various amplitudes and trajectories. More specifically this works objectives are to: (i) identify the conceptual basis for this novel proxy and its implementation; (ii) evaluate said proxy through high resolution geochemical analysis of trace elements in a sediment core, and comparison to regional palaeoclimate proxies; and (iii) discuss the record's significance for the Lower Lakes of the Murray-Darling Basin in light of the extreme Millennium Drought acidification event.

2. Materials and methods

2.1. Site selection

Historically low precipitation over the Murray-Darling Basin during the Millennium Drought (~1995–2009) caused water levels in the basin's terminal lakes, Lakes Albert and Alexandrina, to fall by approximately 2 m with a seven-fold increase in lake water salinity (Fig. 1; Leblanc et al., 2012). These lake levels were significantly below the range of historically observed seasonal and inter-seasonal variability, exposing formerly anaerobic sulfidic sediments around the lakes littoral fringe, mobilizing unusually large volumes of sulfuric acid and driving a greater overall system acidification (Fitzpatrick et al., 2008b). This culminated in the mobilisation of metals at a level unprecedented since European Settlement of the region in the 1880's (Baldwin, 2011; Hall et al., 2006). Acidification was particularly severe around Lake Albert, a large (171.5 km²) shallow freshwater lake (mean depth 1.5 m at pre-drought levels (2003–2005)), where 48 km² of pyritic sediment was exposed after lake level decline from +0.71 to –0.58 m above Australian Height Datum (mAHD) where it was maintained artificially via pumping during the Millennium Drought (Mosley et al., 2014).

2.2. Sampling

Sediment core (code LA1) was collected from near the centre of Lake Albert (coordinates (DD): –35.641217, 139.304283) in 2009 as detailed in Murdoch (2009), at a location approximately equidistant from the east and west banks of Lake Albert, where significant formation of sulfuric material was identified during the Millennium Drought (Fig. 2; Fitzpatrick et al., 2008b).

The core, 127 cm in length, was collected using an 80 mm polyvinyl chloride core barrel and was cut into three sections. All core sections were stored under identical conditions at 3 °C. As some shrinking of the core sections had occurred during storage, the sample depths were corrected with the assumption of uniform shrinkage across each section. Samples for dating were taken at 3, 6, 9, 12, 16, 17 and 20 cm depth with no remaining material available for geochemical analysis, and they are therefore omitted from subsequent analyses (Murdoch, 2009).

The core was split longitudinally, and sub-sampled into contiguous 1 cm slices cut at right angles to the angle of penetration (assumed to be 0°), and parallel to the assumed bedding plane. ~3 g of wet sediment was sub-sampled before being oven dried at 60 °C for 24 h. Oven dried samples were then powdered and homogenized in an agate mortar and pestle.

2.3. Analytical methods

In order to target a geochemical signal symptomatic of acidification the BCR (Community Bureau of Reference; a European standards, measurements and testing agency) Sequential Extraction Procedure was applied to minimise the impact of trace metals of lithogenic origin on overall signal, due to a targeting of elements in more mobile chemical phases, thus leaving primary silicate structures intact where trace

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