



Acidification of lake water due to drought

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SUMMARY

Droughts are predicted to increase in many river systems due to increased demand on water resources and climate variability. A severe drought in the Murray-Darling Basin of Australia from 2007 to 2009 resulted in unprecedented declines in water levels in the Lower Lakes (Ramsar-listed ecosystem of international importance) at the end of the river system. The receding water exposed large areas (>200 km²) of sediments on the lake margins. The pyrite (FeS₂) in these sediments oxidised and generated high concentrations of acidity. Upon rewetting of the exposed sediments, by rainfall or lake refill, surface water acidification (pH 2–3) occurred in several locations (total area of 21.7 km²). High concentrations of dissolved metals (Al, As, Co, Cr, Cu, Fe, Mn, Ni, Zn), which greatly exceeded aquatic ecosystem protection guidelines, were mobilised in the acidic conditions. In many areas neutralisation of the surface water acidity occurred naturally during lake refill, but aerial limestone dosing was required in two areas to assist in restoring alkalinity. However acidity persists in the submerged lake sediment and groundwater several years after surface water neutralisation. The surface water acidification proved costly to manage and improved water management in the Murray-Darling Basin is required to prevent similar events occurring in the future.

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1. Introduction

Droughts are likely to increase in frequency and severity in many regions of the world due to climate change (IPCC, 2008; Talaksen and van Lanen, 2004). Such events can result in large deteriorations in water quality, particularly in regard to increased salinity, temperature and eutrophication (Caruso, 2002; Mosley et al., 2012; van Vliet and Zwolsman, 2008).

Low water levels during droughts also increase the likelihood of the exposure of sediments which have previously been submerged for long periods of time. Natural sulfate reduction processes in aquatic sediments can, where sufficient organic matter and iron minerals exist, result in an accumulation of sulfide minerals such as pyrite, FeS₂ (Holmer and Storkholm, 2001). Pyrite is quite stable under waterlogged conditions, however, upon exposure to air it can oxidise to produce sulfuric acid and dissolved ferrous (Fe²⁺) iron. The ferrous ion can subsequently be oxidised to ferric ion (Fe³⁺) which can hydrolyse and precipitate as solid ferric iron oxide (Fe(OH)₃) or at low pH (<4) oxidise further pyrite (Dent, 1986;

Stumm and Morgan, 1996). The overall pyrite oxidation reaction can be written:



The term acid sulfate soils is often used to classify materials which due to high pyrite contents and/or lack of acid neutralising carbonate materials, show severe acidification (pH < 4) upon exposure to air. The hydrogen ion (H⁺) acidity generated by the pyrite oxidation reaction can subsequently dissolve the sediment matrix, liberating other metals into solution such as aluminium and potentially leading to water body acidification (Nordmyr et al., 2008; Simpson et al., 2010). Exposing acid sulfate soils via drainage has resulted in acidification and metal contamination of groundwater in many locations (Bronswijk et al., 1993; Burton et al., 2008; Cook et al., 2000) and subsequent outflows of acidic drainage water have in some instance resulted in surface water acidification (Nordmyr et al., 2008; Sammut et al., 1996). Drying of a small freshwater wetland has also resulted in surface water acidification upon rewetting (McCarthy et al., 2006). Yan et al. (1996) reported lake acidification after a drought decreased lakewater levels, thus exposing sulfidic littoral sediments to the atmosphere. However, the sulfur present in these sediments was due to previous atmospheric deposition of industrial sulfur dioxide into the lake and its catchment.

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Australia's largest river system, the Murray-Darling Basin, has recently experienced the most severe climatic and drought conditions in over 100 years of records (MDBA, 2010; Timbal, 2009). Declining water levels in the lower reaches of the River Murray resulted in the extensive exposure, oxidation and acidification of pyrite-containing sediments in the large freshwater lakes at the end of the river system (Fitzpatrick et al., 2010). The aim of this paper is to report the causes and characteristics of the subsequent acidification of surface waters in these lakes. The emerging risks reported are also relevant to other systems that may experience increasing droughts in the future due to climatic changes or lowering water levels due to water management decisions.

2. Methods

2.1. Study area description

The study area comprised two lakes, Albert and Alexandrina, collectively known as the Lower Lakes (Fig. 1). These lakes and associated wetlands comprise the most downstream freshwater portion of the Murray-Darling Basin (1,061,469 km² catchment area). The Lower Lakes were separated from the coastal lagoon known as the Coorong and the river mouth in the early 1940s to satisfy irrigation demand for freshwater, prevent seawater intrusion and maintain navigation into the Lakes and river. The Lower Lakes are large and very shallow; Lake Alexandrina (650.2 km²), the larger and deeper of the two lakes has a maximum depth of approximately 4.1 m and a mean depth of 2.9 m at full capacity; Lake Albert (171.5 km²) is much shallower with a maximum depth of approximately 2.3 m and a mean depth of 1.4 m at full capacity (Mosley et al., 2012).

The Coorong and Lakes Alexandrina and Albert, are collectively recognised as one of Australia's most significant ecological assets and have been designated an ecosystem of international importance under the Ramsar convention (Phillips and Muller, 2006). The lakes also support several large irrigated agricultural areas, are important for recreational activities such as swimming, boating and fishing, and have high cultural importance for the local Ngarindjeri aboriginal people.

2.2. Sampling and analysis

Surface water quality samples ($n = 1228$, approximately 300 per year) were collected from various locations (Fig. 1), approximately monthly in the open water body ($n = 663$) and daily-fortnightly (depending on the risk or presence of acidification) on the lake margins ($n = 565$) from 2008 to 2010. Grab samples were collected at all locations according to Standard Methods (APHA, 2005). pH was measured at the time of sample collection using a calibrated instrument (YSI Pro Plus). New polyethylene bottles (1 L), washed and rinsed with deionised water, were used to collect samples for laboratory analysis of conductivity, acidity, alkalinity, sulfate and chloride. Acid-cleaned 100 mL polyethylene bottles were used to collect samples for nutrient analysis. Acid-cleaned 250 mL polyethylene bottles were used to collect samples for metal analysis. Samples were stored at 4 °C and all analyses were performed within 1 week of sampling at the Australian Water Quality Centre's National Association of Testing Authorities (NATA) accredited laboratory using Standard Methods (APHA, 2005). NATA accreditation requires maintenance and documentation of strict quality control procedures. Total alkalinity was measured by Gran titration to a pH 4.5 end-point, acidity was measured by titration to pH 8.3 at 25 °C following hot peroxide digestion, dissolved (<0.4 µm filtered within 24 h of sampling and acidified to pH < 2 with HNO₃) metals (Al, As, Co, Cr, Cu, Fe, Mn, Ni, Zn) and selected major ions (sulfate by

ICP-AES assuming all dissolved S is present as sulfate, and chloride by a Ferricyanide method, APHA, 2005 method 4500-Cl⁻ E), turbidity by a nephelometer, and nutrients (ammonia, NH₄; total phosphorus, TP; total nitrogen, TN) by standard colorimetric methods (APHA, 2005). Salinity (mg/L) was calculated by measuring specific electrical conductivity (EC, µS/cm at 25 °C) using a calibrated conductivity meter and multiplying by a factor of 0.55 (local factor obtained by comparison of EC with salinity measured gravimetrically following evaporation). Dissolved oxygen was routinely measured in the field (YSI Pro Plus) and was close to saturation with atmospheric levels in the shallow lake waters.

Hydrological parameters, river flow and level (measured at Lock 1, Fig. 1), and lake level (average of 5 stations) were provided by the Department for Environment, Water and Natural Resources (South Australia). Lake volume and surface area were determined with ARC GIS 3D analyst™ using the measured water levels and surveyed bathymetry. Exposed sediment area was determined by the difference in lake surface area compared to the pre-drought shoreline location.

To assess the risk of groundwater acidification under the exposed lake beds in the drought (and risks to surface water arising from this), a series of shallow piezometers were installed on the shorelines of Lake Albert, Lake Alexandrina and Currency Creek in 2009. In this paper we report pH data from four piezometers at two locations (Cambell Park and Point Sturt, see Fig. 1 for general locations). For a detailed description of the piezometer sites, installation and testing refer to Earth Systems (2010). The piezometers have been sampled monthly-quarterly since installation in 2009 (sampled by Earth systems from 2009 to 2010 and South Australian EPA from 2010 to 2013). Prior to each sampling the piezometers were purged using a Solonist™ peristaltic pump connected to a 12 V battery. Approximately three well volumes were pumped from each piezometer. Pumping was continued through a flow-cell and multi-meter (YSI Pro Plus) and readings were taken when pH was stable (± 0.01 units).

3. Results and discussion

3.1. Hydrology and pyrite oxidation

The water levels from 2007 to 2009 in the Murray River below Lock 1 and in Lake Alexandrina were the lowest since records began in 1920 (Fig. 2). This was a result of very low river inflows from the Murray-Darling Basin, mostly as a consequence of autumn rainfall reductions (Timbal, 2009; Ummenhofer et al., 2010) and an underlying issue of water over-allocation for agriculture (MDBA, 2010). The average annual river flow in the drought period (2007–2009) was 537 GL/yr compared to 1588 GL/yr in the pre-drought period (2003–2005, Mosley et al., 2012). Summary hydrological statistics are presented for the Lower Lakes in Table 1. Prior to the drought period, regulated high water levels (historic pool level average of +0.7 m Australian Height Datum, AHD, where 0 m AHD corresponded to mean sea level in 1967), high organic matter production (highly productive eutrophic lakes), and presence of sulfate (coastal location) were likely conducive to the formation of pyrite in the sediment. The large (nearly 2 m, Table 1) water level declines from 2007 to 2009 resulted in large areas on the lake margins being exposed, including complete drying of many embayments (compare water levels in Fig. 2 with bathymetry in Fig. 1). The lowest daily water levels (−1.2 m AHD, in Lake Alexandrina and −0.5 m AHD in Lake Albert, a higher level maintained due to pumping water over a temporary regulating structure) were reached in April 2009 (Table 1). These water levels equate to a total sediment exposure of approximately 209 km² (161 km² Lake Alexandrina and 48 km² Lake Albert), which represents approximately 25% of the pre-drought surface area of both lakes (Table 1).

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