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Hydrogeochemistry in a relatively unmodified subtropical catchment: Insights regarding the health and aesthetic risks of manganese

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ABSTRACT

Study region: Metal and nutrient concentrations were analysed from freshwater and soil collected from a relatively unmodified ephemeral subtropical catchment in Central Queensland, Australia. Samples were collected quarterly between December 2012 and December 2014.

Study focus: There is increasing concern about the health effects of manganese in drinking water, prompting interest in understanding manganese dynamics. This study investigated the temporal and spatial patterns of metals and nutrients, as influenced by summer rainfall; and the impacts of those variables on drinking water, associated infrastructure and concomitant health risks.

New hydrological insights for region: Results show that the hydrogeochemistry of Baffle Creek was predominantly driven by rain events, with differing responses to varying magnitudes of rain and discharge. Following a flushing event, dissolved aluminium and nitrate increased in the surface and drinking water, whereas manganese and ammonia were undetectable in the drinking water and occurred only in small concentrations in the surface waters. In contrast, when rainfall events occurred without an associated flush, manganese, iron and ammonia concentrations increased in the drinking water. The hydrochemistry of manganese and iron in the supply chain infrastructure was strongly positively correlated with ammonia, and aluminium correlated with nitrate concentrations. Currently the drinking water supply does not pose a threat to chronic manganese exposure for humans, however elevated concentrations are experienced periodically; and may become more problematic under climate change scenarios.

1. Introduction

Manganese (Mn) is found naturally in ground and surface waters, with approximately 97% of surface water sites in the USA known to contain Mn (USEPA 2002). The primary source of Mn in natural catchments is the surrounding soils, bedrock and organic matter washed into the water bodies. In surface waters, Mn occurs in both dissolved and suspended forms, with the prevalence of form depending on factors such as pH, anions present and oxidation–reduction potential (ATSDR, 2000). In most waters, the divalent form Mn^{2+} dominates at pH 4–7, however, more highly oxidised forms can arise at higher pH values, or result from microbial oxidation (ATSDR, 2000). Furthermore, Mn can be adsorbed on soil; however the extent of adsorption depends on the organic content and cation exchange capacity of the soil (WHO, 2011).

Traditionally Mn has been limited to an aesthetic concern (Sly et al., 1990), however, there has been growing concern

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internationally on the human health effects of Mn in drinking water, particularly with regard to impacts on pregnant women (Grazuleviciene et al., 2009), infant size and mortality (Hafeman et al., 2007; Rahman et al., 2015), and intellectual function in children (Wasserman et al., 2006; Ljung and Vahter 2007; Wasserman et al., 2011) as well as in the elderly (Kondakis et al., 1989). In humans, Mn is an essential trace element used in bone formation and in amino acid, lipid and carbohydrate metabolism (Trumbo et al., 2001). However, chronic exposure to Mn in drinking water at concentrations below the health-based guidelines used in many countries (e.g., 0.4 mg/L–0.5 mg/L), may result in neurological disorders similar to a Parkinson's-like disease called "Manganism". Along with neurological disorders, modest Mn concentrations in drinking water have also been linked with increased dental caries (Tsanidou et al., 2015), behavioural problems (Khan et al., 2011), reduced intelligence quotient (IQ) scores (Bouchard et al., 2011), difficulty with mathematics (Khan et al., 2012) and reduced motor skills in children (Dion et al., 2016).

According the Australian Drinking Water Guidelines (ADWG), uncontaminated rivers and creeks have Mn concentrations ranging from 0.001 mg/L to 0.6 mg/L, with greater concentrations occurring when polluted or anoxic conditions prevail (NHMRC 2011). At concentrations as small as 0.02 mg/L, Mn has been reported to form films on water pipes (Sly et al., 1990); these films have the potential to slough as a black precipitate (Cerrato et al., 2006b), thus increasing Mn concentrations and causing dirty water problems for consumers (Khoe and Waite, 1989; Cerrato et al., 2006a; Gerke et al., 2016). Microorganisms can also influence the Mn cycle and biofilm within supply chain pipes (Cerrato et al., 2010; Ginige et al., 2011). In Australia, Mn odour and discolouration is the source of most complaints relating to potable water supplies, with 86% being due to dirty water problems associated with increased metals (Mn and Iron (Fe)) (Ginige et al., 2011). This places considerable pressure on water managers to find effective ways to treat water supplies where a greater probability of elevated Mn concentrations exists.

The treatment of water containing increased Mn concentrations can be difficult (Choo et al., 2005), as Mn takes much longer to oxidise and precipitate compared with other metals such as Fe (Khoe and Waite 1989). Manganese removal can also be impaired in the presence of dissolved organic matter (Carlson and Knocke, 1999), though the addition of potassium permanganate can be used to enhance the oxidation process (Wang et al., 2016). Reverse osmosis and ion exchange can be used to achieve greater Mn removal, whereas treatment using activated carbon and greensands has been reported with varying results (Barbeau et al., 2011b). The use of bio filters can be an efficient (Hasan et al., 2014) Mn removal method with up to 98% of Mn being removed with the pH as low as 6.3 (Hoyland et al., 2014).

Given the challenges of effective and efficient Mn treatment removal, understanding the temporal and spatial relationships of Mn will be invaluable in assisting water managers to refine or customise treatment options. For example, an early forewarning of increased Mn may signal the need to add additional steps in the treatment process, as a measure to effectively remove Mn from the water. This is particularly the case in the context of smaller treatment plants, where resources may be limited, and communities do not have ready access to alternate supplies in the event of water contamination with Mn and resulting discolouration.

The Baffle Creek Catchment is a unique modern day example of an ephemeral freshwater system in subtropical Australia pre European settlement. Unlike many other catchments throughout subtropical Australia, Baffle Creek is unique in its lack of structures obstructing flow (e.g. dams/weirs) (Gilbert and Brodie 2001), as well as its relatively unmodified condition with minimal residential and commercial development throughout the catchment. Historically, investigations into Mn in freshwater systems in Australia have taken place following the introduction of river regulation, including Mardi Dam in the Wyong Shire (Khoe and Waite, 1989) and numerous reservoirs in the South-East corner of Queensland (Bertone et al., 2016; Zaw and Chiswell, 1995, 1999; Grinham et al., 2012). This has resulted in a paucity of knowledge regarding hydrogeochemical processes in natural systems. As such, the Baffle Creek catchment represents an ideal system to ascertain the transport of metals in a natural/relatively unmodified catchment context, as well as potential impacts on human health and deterioration of the drinking water supply chain infrastructure.

The present study investigated the spatial and temporal variation of Mn and Fe in Baffle Creek. The hydrogeochemistry of Mn and Fe was compared between the surface waters and the supply chain, to establish relationships between these two water sources. The main objective was to evaluate the spatial and temporal hydrochemistry of Mn and Fe in surface waters and drinking water supply chain, as well as to ascertain any current or potential future health or aesthetic risks. The hypotheses were that (1) Increased water discharge during a flood or "flush" event will result in reduced Mn and Fe concentrations in the surface waters and drinking water supply chain; and (2) Smaller rain events that result in less or no discharge, such as "fill-shift" events, will result in increased Mn and Fe concentrations in the natural lagoon from which the drinking water is drawn and the drinking water supply chain.

2. Methods

2.1. Catchment

Baffle Creek is situated on the eastern seaboard of sub-tropical Australia (Fig. 1), and is the southernmost catchment to directly discharge into the Great Barrier Reef Marine Park. The catchment is void of man-made structures regulating flow, making it the Southern Great Barrier Reef's only remaining unregulated catchment. The catchment margins range from Tannum Sands in the north, to the mouth of the Kolan River in the south, covering an area of 4 085 km^2 (Department of Environment and Heritage Protection, 2016). The climate in the catchment is subtropical, with December, January and February traditionally being the wettest months (Department of Environment and Heritage Protection, 2016).

Untreated Baffle Creek water enters the Miriam Value Drinking Water Treatment Plant from Baffle Creek and subsequently this water serves as the potable supply for the rural township of Miriam Vale, which has a population of approximately 500 people (ABS, 2011). There is minimal private or commercial development in the catchment with the primary land uses including dry land cattle grazing, dairy farming and timber plantations.

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