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A method for separation of heavy metal sources in urban groundwater using multiple lines of evidence[☆]

Emily Hepburn [a], Anne Northway [b], Dawit Bekele [c, d], Gang-Jun Liu [e], Matthew Currell [a, *]

[a] School of Engineering, RMIT University, Melbourne, Australia
[b] Environment Protection Authority Victoria, Australia
[c] Global Centre for Environmental Remediation, University of Newcastle, Australia
[d] Cooperative Research Centre for Contamination Assessment and Remediation of the Environment (CRC CARE), Australia
[e] School of Science, RMIT University, Melbourne, Australia

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ABSTRACT

Determining sources of heavy metals in soils, sediments and groundwater is important for understanding their fate and transport and mitigating human and environmental exposures. Artificially imported fill, natural sediments and groundwater from 240 ha of reclaimed land at Fishermans Bend in Australia, were analysed for heavy metals and other parameters to determine the relative contributions from different possible sources. Fishermans Bend is Australia's largest urban re-development project, however, complicated land-use history, geology, and multiple contamination sources pose challenges to successful re-development. We developed a method for heavy metal source separation in groundwater using statistical categorisation of the data, analysis of soil leaching values and fill/sediment XRF profiling. The method identified two major sources of heavy metals in groundwater: 1. Point sources from local or up-gradient groundwater contaminated by industrial activities and/or legacy landfills; and 2. contaminated fill, where leaching of Cu, Mn, Pb and Zn was observed. Across the precinct, metals were most commonly sourced from a combination of these sources; however, eight locations indicated at least one metal sourced solely from fill leaching, and 23 locations indicated at least one metal sourced solely from impacted groundwater. Concentrations of heavy metals in groundwater ranged from 0.0001 to 0.003 mg/L (Cd), 0.001–0.1 mg/L (Cr), 0.001–0.2 mg/L (Cu), 0.001–0.5 mg/L (Ni), 0.001–0.01 mg/L (Pb), and 0.005–1.2 mg/L (Zn). Our method can determine the likely contribution of different metal sources to groundwater, helping inform more detailed contamination assessments and precinct-wide management and remediation strategies.

1. Introduction

In recent decades, the shift towards de-industrialisation of the world's major cities has resulted in rapid re-development of brownfield land (Chen and Jiao, 2008). The reuse of brownfields for housing, parks and recreation has emerged as a key planning strategy for governments to address growing urban populations (De Sousa and Ghoshal, 2012; Bartke, 2013; Atkinson et al., 2014; UN, 2015). However, historical industrial districts often contain heavy metals in soils, sediment and groundwater (Critto et al., 2006; Carlon et al., 2007, 2008; Relić et al., 2010). Mobilisation of heavy metals can impact ecological systems via discharge of contaminated groundwater to surface water and threaten plant and animal health if bioavailable (Moore, 1999; Alloway, 2013a,b). Human exposure pathways include dermal contact, inhalation of particulates or direct ingestion of impacted soil, and contact with affected surface waters (NEPM, 2013a,b,c). Consequently, environmental regulators are attempting to better understand public health and environmental risks associated with brownfields prior to their re-development (Ramsden, 2010; Xie and Li, 2010).

Understanding sources of heavy metals and their environmental fate is essential to successful risk assessment, remediation/management and ultimately, re-development; particularly in coastal areas, where dynamic geochemical and hydrogeological conditions can influence their behaviour (Du Laing et al., 2009a; Lions et al., 2010; Wang et al., 2016). Mobilisation of heavy metals in surface

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* Corresponding author. GPO Box 2476, Melbourne VIC 3001 Australia.
E-mail address: Matthew.currell@rmit.edu.au (M. Currell).

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water and groundwater is typically described in terms of how they distribute between dissolved and solid phases; the rates of adsorption, desorption and dissolution can be influenced by geochemical parameters and sediment composition (Weiner, 2013). Field and laboratory-based simulations show heavy metals may desorb due to changes in physico-chemical conditions such as salinity, pH and redox potential (Clemente et al., 2008; Acosta et al., 2011; Fdez-Ortiz de Vallejuelo et al., 2014; Hafeznezami et al., 2016). Sequential leaching experiments also demonstrate a strong pH dependency and the importance of Fe-Mn oxides, clays and organic matter on metal mobility in soils (Dijkstra et al., 2004; Markiewicz-Patkowska et al., 2005; Relić et al., 2010; Kumar et al., 2015). However, to determine the predominant source(s) of metals in groundwater where there are multiple natural and/or anthropogenic inputs, multiple lines of geological, hydrogeological and geochemical data are required.

Many studies have sought to identify heavy metal sources in coastal aquifers and sediments using statistical and empirical methods (Chen and Jiao, 2008; Škrbić; Đurišić-Mladenović, 2010; Syakti et al., 2015; Chen et al., 2016a; Chen et al., 2016b; Sindern et al., 2016; Wu et al., 2016). However, limited studies have approached metal source categorisation by including detailed profiling of metal concentrations in soil, sediment and groundwater, using techniques such as X-ray fluorescence (XRF) (Carr et al., 2008; Weindorf et al., 2013). Also, few studies have examined metal sources and behaviour through soils/sediment and groundwater in artificial, highly modified systems, instead focusing on contamination to relatively undisturbed areas (Liaghati et al., 2003; Davis et al., 2009; Luo et al., 2015; Chen et al., 2016a; Chen et al., 2016b). Detailed characterisation of heavy metal sources, fate and transport in urban areas is typically conducted on a 'site by site' basis, as part of environmental site assessment guidelines and policies. However, in regions such as Fishermans Bend, where large parcels of land containing many sites and complicated land-use history are being redeveloped as precincts, a more regional approach is needed to characterise heavy metal contamination and distinguish point source effects from broader regional-scale processes.

In this study, we demonstrate a method with which to differentiate heavy metal sources in groundwater in an area long subject to intensive anthropogenic activity. Numerous environmental audits have been completed for individual sites, documenting local-scale sources of heavy metal contamination in groundwater (e.g. Golder, 1993; URS, 2001, 2005, 2014). However, there has been no regional-scale study to assess the wider contribution of the extensive artificial fill layer to groundwater heavy metal loads, or contamination plumes from sites such as legacy landfills. Both sources may impact groundwater to varying degrees at a given locality. Through assessment of groundwater and XRF-derived profiles of heavy metal concentrations, we sought to categorise heavy metals in groundwater into different source types, based on their respective mean concentrations in fill. We hypothesised that incorporation of simple statistical techniques with multiple lines of evidence could provide an indication of different heavy metal sources in groundwater and that this method could be used as a guide by environmental practitioners when assessing the likelihood of such sources. Differentiation of heavy metal sources in former industrial precincts can help inform contamination management and remediation strategies. The method is designed to be applicable at a regional scale, relying on datasets that can be readily collected by practitioners. We stress that the method is not designed to replace more detailed studies of metal sources, fate and transport in particular locations, for example using solute transport modelling and more intensive field data collection.

2. Background and setting

2.1. Historical land use at Fishermans Bend

Fishermans Bend is located approximately 1 km southwest of Melbourne's Central Business District (CBD) in southeast Australia (Fig. 1). It has been a site of industrial and commercial activity since the mid-1800s, prior to which the area was characterised by shallow swamps, commonly used for dumping of industrial and domestic waste (Biosis, 2013). Sand quarrying occurred prior to the 1920s after which landfilling occurred at seven known locations (Fig. 1) (U'Ren and Turnbull, 1983). The land was later developed for industries including automotive manufacturing, metal fabrication, and transport and logistics (Biosis, 2014). The area is currently used for heavy and light commercial and industrial purposes (Golder, 2012). Urban renewal began in 2012 when the Victorian State Government re-zoned 240 ha for residential use. Now the largest urban re-development project in Australian history, Fishermans Bend has an anticipated development timeframe of over 40 years (DELWP, 2017). Due to its industrial history, land reclamation activities and complex geology, successful re-development will benefit from careful analysis of soil/sediment and groundwater contamination, based on clear understandings of hydrogeochemical processes (Golder , 2012). Detailed reviews of historical land uses are available in Golder (2012) and AECOM (2015).

2.2. Geology and hydrogeology

Fishermans Bend is located within the Yarra River Delta, a sequence of unconsolidated Quaternary sediments deposited at the mouth of the Yarra and Maribyrnong rivers (Fig. 1). Holdgate and Norvick (2017) characterised the geological evolution of the delta showing that the uppermost units comprise Holocene transitional alluvial/marine sediments. The main geological units of relevance to this study are the Port Melbourne Sand (PMS) and the Coode Island Silt (CIS). Overlying these is a layer of artificially imported fill, comprising a mixture of industrial and construction waste, and quarried material from locations within or proximal to the region. Fill generally consists of grey-brown sand and gravel, with variable amounts of re-worked clay, dredged sediment and inclusions such as scrap metal, brick, plastic and concrete. The fill ranges from absent to approximately 5 m (average 2.5 m) thick.

The Port Melbourne Sand consists of pale grey-brown, fine-to medium-grained quartzose sands and acts as an unconfined high-yielding aquifer, typically 5–10 m thick, with a shallow water table between 1 and 3.5 m below ground surface (BGS), (Neilson, 1992; AECOM, 2016). A broad range of hydraulic conductivities between 0.86 and 43 m/day are reported in Cooney (1984, cited in Leonard, 2006) likely due to the presence of clay lenses within the unit. The Coode Island Silt consists of dark grey-brown, soft, silty clays (illite and kaolinite), marine shells and plant material (Smith and Milne, 1979). The unit acts as a low-yielding aquitard, 20–25 m thick, with a high porosity and low permeability (Hancock, 1992). Contact between the Port Melbourne Sand and the Coode Island Silt is typically sharp but can be gradational, particularly in the northeast of Fishermans Bend where the units are lateral equivalents (Holdgate and Norvick, 2017).

Groundwater elevations in the water table aquifer indicate flow predominantly converging in the southeast of the study area towards a redundant sewer, which appears to re-direct groundwater away from a topography-driven drainage path (Fig. 1, AECOM, 2016). Groundwater elevations in the west of the study area indicate potential flow towards Hobsons Bay. Tritium activities in the Port Melbourne Sand aquifer range from 1.75 to 2.45 TU (median = 1.94 TU; n = 11), and from 0.20 to 0.35 TU (median = 0.25