



Unravelling metal mobility under complex contaminant signatures

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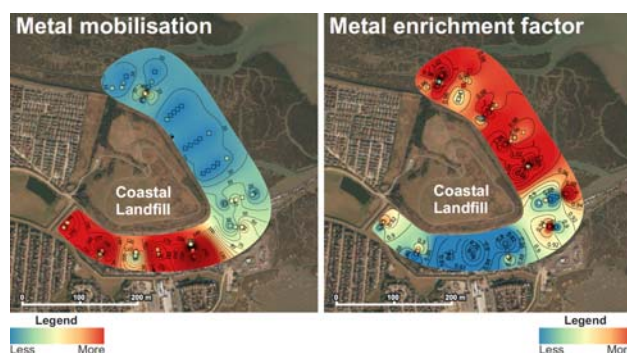
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HIGHLIGHTS

- Laboratory experiments and field survey were performed for pollution assessment.
- Contaminated sediments leached significant amounts of metals to overlying water.
- Metal mobilisation varied ~1000-fold depending on sediment pH, salinity, and redox.
- Metal mobility affected concentration in sediment and contamination signatures.
- Assessment of metal mobility in estuarine sediments is essential to infer environmental impacts.

GRAPHICAL ABSTRACT



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ABSTRACT

Metals are concerning pollutants in estuaries, where contamination can undergo significant remobilisation driven by physico-chemical forcing. Environmental concentrations of metals in estuarine sediments are often higher than natural backgrounds, but show no contiguity to potential sources. Thus, better understanding the metal mobility in estuaries is essential to improve identification of pollution sources and their accountability for environmental effects. This study aims to identify the key biogeochemical drivers of metal mobilisation on contaminated estuarine sediments through (1) evaluation of the potential mobilisation under controlled conditions, and (2) investigation of the relevance of metal mobilisation for in situ pollution levels in an area with multiple contaminant sources. Sediments from a saltmarsh adjacent to a coastal landfill, a marina, and a shipyard on the Thames Estuary (Essex, UK) were exposed in the laboratory (24 h, N = 96, 20 °C) to water under various salinity, pH, and redox potential. Major cations, Fe(II), and trace metal concentrations were analysed in the leachate and sediment. Salinity, pH and redox had a significant effect on metal mobilisation ($p < 0.001$), e.g. under certain conditions Fe(II) leaching was increased ~1000-fold. Measurements in situ of surface and subsurface sediment cores revealed that landfill proximity poorly explained metal spatial distribution. However, physico-chemical parameters explained up to 97% of geochemically normalized metal concentrations in sediments. Organic matter and pH were dominant factors for most of the metal concentrations at the sediment surface. At subsurface, major cations (Ca, Na, Mg and K) were determinant predictors of metal concentrations. Applying the empirical model obtained in the laboratory to geochemical conditions of the studied saltmarsh it was possible to demonstrate that Fe mobilisation regulates the fate of this (and other) metal in that area. Thus, present results highlight the importance of metal mobility to control sediment pollution and estuarine fate of metals.

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1. Introduction

Estuaries are transitional ecosystems between land and sea that provide a multitude of services (Millennium Ecosystem Assessment, 2005). These areas are amongst the most biologically productive of the planet, and therefore of extreme relevance for marine and freshwater biodiversity protection. Additionally, the provisioning of privileged access to marine, freshwater and continental resources made estuarine systems preferable places for urbanization and industrialization (Kennish, 1991). In fact, many important large metropolitan areas of current commercial and industrial centres, such as London, New York, Shanghai, Lagos, Istanbul and Tokyo are situated in estuarine systems. Therefore, estuarine environments have globally experienced both historical and current intense anthropogenic activities and consequent contamination (Chapman and Wang, 2001; Ridgway and Shimmield, 2002).

Metal contaminants have been traditionally problematic in estuaries because they tend to present non conservative behaviour, while still concentrating and accumulating in estuarine sediments (Machado et al., 2016). In fact, metal-contaminated sediments are commonly reported as of potential concern for the quality of estuarine waters and benthic organism worldwide (Bianchi, 2007). For instance, legacy contaminated sediments in estuaries from Southeast England might display up to 70% of bioavailable metals, many of which at levels that threaten wildlife and environmental services (Spencer and MacLeod, 2002). Many estuaries receive substantial inputs of metals such as Cd, Cu, Cr, Hg, Pb, Zn that concentrate in the sediment at levels up to one million-fold higher than the water concentrations (Förstner and Wittmann, 1979; Zwolsman et al., 1993; Attrill and Thomes, 1995).

On the other hand, the non-conservative behaviour of metals (Machado et al., 2016) driven by mobilisation hampers the assessment of pollution sources and misleads inference of pollution levels. For instance, the Newlands saltmarsh (Thames Estuary, UK) presents metal concentrations at concerning levels but with weak association to the main known sources (O'Shea, 2016 <https://qmro.qmul.ac.uk/xmlui/handle/123456789/12995>), which obstructs accountability for pollution. Indeed, O'Shea (2016) determined that a combination of proximity to contaminant source and post-depositional mobility accounted for spatial variability in metal concentrations in those intertidal sediments. Many of the physical, chemical and biological gradients present in estuaries affect the metal speciation, partitioning, transport, and consequently spatial distributions (Zwolsman et al., 1993). As an example, the decay of organic matter, periodic flooding, and air exposure of sediments on intertidal areas provoke a cyclic covariation of pH and redox (Machado et al., 2016), with consequent changes on chemical speciation and cycling of several metals (Du Laing et al., 2009c). The diagenetic remobilisation and the difference on sediment pH and salinization due to seawater inflow similarly affects metal speciation and fate (Zwolsman et al., 1993; Du Laing et al., 2009a,b; Johnston et al., 2011). As a result, complex contaminant signatures arising from multiple sources and high environmental mobility make identification of potential impacts particularly challenging.

Notwithstanding, metal pollution is still investigated in estuaries as in fully fresh or marine sediments, i.e. in terms of continuity of high contaminant levels to the contamination sources (Chapman and Wang, 2001). That is partially because it is still unclear how relevant the impacts of metal mobility are in estuarine sediments near contamination sources. Consequently, such lack of information obstructs a realistic understanding of the estuarine pollution and a quantification of the metal mobilised from the sediment to the bioavailable water fraction that mostly impacts aquatic biota.

The present study aimed to quantify the potential effects of the key biogeochemical drivers of metal mobilisation on the metal pollution levels in estuarine sediments. Therefore, laboratory experiments, field measurements and modeling methods were combined in a manner that could be easily implemented in environmental health assessments. The laboratory experiments aimed to provide an estimation of the

potential for metal leaching on surface sediments and served as basis for empirical models of geochemical metal mobilisation. Additionally, the in situ relationships between metal enrichment factors and sediment geochemistry were investigated in an intertidal area surrounded by multiple potential contamination sources to compare the influence of mobilisation and proximity to the emission source. The study site chosen here is under influence of a historical coastal landfill and multiple current metal sources (i.e. shipyard and docking area) in one of the estuaries most heavily impacted by metal pollution in the world (Thames Estuary, UK). Metal enrichment factors were used for the assessment of pollution level because they represent the most commonly employed proxy of metal contamination. Finally, an empirical model from laboratory experiments was combined with the in situ sediment geochemistry to compare pollution levels and metal mobilisation. This approach allowed inference of the importance of metal mobility for pollution levels and its interference on the identification of metal sources in estuarine systems.

2. Material & methods

For the purpose of the present study we define some terms that are broadly used in pollution science with potentially ambiguous meaning. The term “metal immobilisation” is defined as the collection of processes such as adsorption, absorption, precipitation and co-precipitation, crystallization that remove metal from the water column to a solid sedimentary phase (Förstner and Wittmann, 1979). The term “metal mobilisation” is then used here as the sum of physical, chemical and biological processes complementary to immobilisation that result into a transfer of metal as dissolved or particulate to the aqueous phase with potential to be transported by water hydrodynamics. The term “metal mobility” is thus defined as the interaction of immobilisation and mobilisation, in which low metal mobility implies high net immobilisation whereas high metal mobility implies high net mobilisation. Moreover, salinity is presented here as practical salinity units 1978 (Unesco/ICES/SCOR/IAPSO, 1981), which is proportional to the conductivity of the sample and its salt content. Therefore, the term salinization is used here as the increase in salinity (and salt contents) of water, slurry or sediments.

2.1. Study area

The Thames Estuary is a tidally dominated estuary with strong anthropogenic influence and noticeable metal contamination (Spencer and MacLeod, 2002; van der Wal and Pye, 2004; Vane et al., 2015). This estuary is amongst the first industrial centres in the world, which summed to the influence of the City of London are the main causes of a high metal pollution (Attrill and Thomes, 1995). Moreover, several landfills were constructed based on the attenuation principle in the Thames intertidal areas (O'Shea, 2016 <https://qmro.qmul.ac.uk/xmlui/handle/123456789/12995>), which today constitute potential additional contamination source.

The study area is situated on an intertidal saltmarsh of the Thames Estuary (south east England, 58° 21' 25" N and 18° 37' 37" E) receiving both current and legacy contamination from multiple sources (O'Shea, 2016 <https://qmro.qmul.ac.uk/xmlui/handle/123456789/12995>), including the Newlands historic landfill, and the shipyard and docking areas on the Oyster Creek (Fig. 1). The Newlands Landfill, hereafter referred as Newlands, is a historical coastal landfill situated on the northern bank of lower portion of the Thames Estuary (Essex, UK). As many other historic coastal landfills from western Europe, Newlands was constructed with no basal or side wall engineering, which allowed release of contaminated leachate, relying on attenuation by surrounding sediments (O'Shea, 2016 <https://qmro.qmul.ac.uk/xmlui/handle/123456789/12995>). The site was actively receiving waste from 1954 to 1989 when it was dumped approximately 1,000,000 m³ of diverse toxic waste including oil-contaminated material from beach clean ups as well as household and

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