



Abandoned metal mines and their impact on receiving waters: A case study from Southwest England



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HIGHLIGHTS

- Systematic assessment of mine contamination for soil, spoil, sediment and biota.
- Bioavailability estimated for metals in water, spoil, soil and sediment.
- Chemical speciation linked to quality standards and ecological impacts.
- Contamination identified in all media split between adit discharge and diffuse runoff.
- Systematic approach to mine site assessments identified for general use.

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ABSTRACT

Historic mine sites are a major source of contamination to terrestrial and river environments. To demonstrate the importance of determining the significance of point and diffuse metal contamination and the related bioavailability of the metals present from abandoned mines a case study has been carried out. The study provides a quantitative assessment of a historic mine site, Wheal Betsy, southwest England, and its contribution to non-compliance with Water Framework Directive (WFD) Environmental Quality Standards (EQS) for Cd, Cu, Pb and Zn. Surface water and sediment samples showed significant negative environmental impacts even taking account of the bioavailability of the metal present, with lead concentration in the stream sediment up to 76 times higher than the Canadian sediment guidelines 'Probable Effect Level'. Benthic invertebrates showed a decline in species richness adjacent to the mine site with lead and cadmium the main cause. The main mine drainage adit was the single most significant source of metal (typically 50% of metal load from the area, but 88% for Ni) but the mine spoil tips north and south of the adit input added together discharged roughly an equivalent loading of metal with the exception of Ni. The bioavailability of metal in the spoil tips exhibited differing spatial patterns owing to varying ambient soil physico-chemistry. The data collected is essential to provide a clear understanding of the contamination present as well as its mobility and bioavailability, in order to direct the decision making process regarding remediation options and their likely effectiveness.

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1. Introduction

Historic mining for metals in Europe dates back to pre-Roman times, but with notable exceptions most have ceased operations. These abandoned sites are an important source of environmental contamination with elevated levels of toxic elements often recorded in soils and adjacent river systems (e.g. Pirrie et al., 2003;

Rieuwerts et al., 2014; Hudson-Edwards et al., 1996). For example, in many areas of the UK, such as southwest England and other parts of Europe, evidence of uncontrolled historic mining activities has shown to have a large and lasting impact (Galána et al., 2003; Nieto et al., 2006; Rieuwerts et al., 2009). Discharge of metal rich water from abandoned mines to surface and groundwater, and contamination of soils and sediments through associated industrial activity are among the highest recorded in the UK. For example, sediments in the regions Camel, Erme, Fal, Fowey, Gannel and Tamar estuaries are amongst the most contaminated in the UK for cadmium (Cd), lead (Pb), zinc (Zn) and copper (Cu) (Environment Agency, 2008a).

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As a consequence, the legacy of historic mining in Europe poses a significant management issue and a potential barrier to achieving new Environmental Quality Standards (EQS) set under the EU Water Framework Directive (WFD - 2000/60/EC) for metals such as Cu, Cd, Cr, Hg, Ni and Pb. For example, 72% of failures to achieve the Cd quality standard in UK freshwaters are found in mined areas (Environment Agency, 2008b) and for the Specific Pollutants (UKTAG, 2008) with EQS set by the UK (Cu, Zn, manganese (Mn), iron (Fe) and chromium (Cr)) mine impacted catchments contribute an estimated 9% of rivers at risk in England and Wales and 2% in Scotland (Environment Agency, 2008b).

Dissolved metals and metalloids may enter the surface waters from point sources such as mine adits and from diffuse sources; mainly rainwater which has percolated through spoil heaps and leached metals and metalloids therein (Galána et al., 2003; Nieto et al., 2006; Rieuwerts et al., 2009). Metals in runoff from spoil heaps may enter receiving waters as dissolved minerals or adsorbed to particulates, which are transported downstream and deposited by river processes (Jarvis et al., 2006). Over time, suspended sediments will settle in the river or estuary, leading to a gradual accumulation of metals in sediments. Metals within river systems are subject to varying physico-chemical conditions, transferring between the dissolved and solid phases of the aquatic environment, and depending on conditions, may move from a relatively refractory phase into phases with greater mobility and bioavailability, thus impacting on the ecology present (Klerks and Levinton, 1989).

Sediments acting as reservoirs for contaminants in the aquatic environment have been widely documented (e.g. Hartl, 2002; Pirrie et al., 2003; Sasaki et al., 2005 and Rainbow et al., 2011) and can as a result cause negative impacts to benthic ecology. Subsequently, macroinvertebrate biological indices have become a fundamental component of ecological monitoring in the UK and Europe (Metcalf, 1989; Hering et al., 2004). To meet the UK's obligations under the WFD, the UK has developed the River Invertebrate Classification Tool (RICT) which runs the RIVPACS IV software (Wright et al., 2000; SEPA, 2015).

A cost-effective strategy to deal with the pollution from abandoned metal mines cannot be developed until the extent of the contamination is understood. The UK has prioritised 226 waterbodies in England and Wales where pollution from mines is the main cause of EQS failures under the WFD (Environment Agency, 2012; Defra, 2012). However, in few cases is there a clear quantitative understanding regarding the significance of the point and diffuse sources of mine inputs to receiving waters and their relative bioavailability (Banks and Palumbo-Roe, 2010; Mighanetara et al., 2009; Mayes et al., 2008). Speciation-based methods are available to characterise the form of metals within soils and sediment based on sequential extraction to determine which fractions including exchangeable, carbonate, reducible, oxidisable and residual phases the metals are associated with (Konradi et al., 2005; Passos et al., 2011; Zhong et al., 2011 and Rieuwerts et al., 2014). Weakly bound metal, in particular, will be more mobile and potentially bioavailable (and therefore toxic) and so determination of this fraction allows a more detailed site assessment to identify hotspots and risks to the terrestrial and aquatic ecology of the area. Furthermore, models are available which provide site specific predicted no effect concentrations for terrestrial organism exposed to potentially toxic elements including lead, nickel, copper and zinc in soils based on ambient conditions of cation exchange capacity, pH, clay content and organic carbon fraction (Arche, 2014).

Until recently, surface water EQS have been derived from hardness-based corrections as a surrogate for metal bioavailability. Metals related research has significantly added to the

understanding of physico-chemical influences on metal speciation (e.g. Pettersson et al., 1993; Vink, 2002) and the development of biotic ligand models (e.g. Dixon and Sprague, 1980; Meyer et al., 1999; Santore et al., 2002). These models enable the prediction of bioavailable concentrations based on a combination of the physico-chemical properties of water and ecotoxicological data (Comber et al., 2008). By accounting for bioavailability, it is possible to provide the most environmentally and ecologically relevant metric for metal risk. This approach has led to new aquatic EQS being derived at an EU levels for Pb and Ni and in the UK for Cu, Mn and Zn (Table A1 of supplementary data). Proposed Predicted No Effect Concentrations for soils have been developed taking account of a combination of pH, organic carbon, % clay and cation exchange capacity for Pb, Cu, Zn and Ni (Arche, 2014).

It is therefore now possible to estimate the chemical availability, and hence potential bioavailability, of metals in all relevant environmental media at a contaminated site. This potential has been tested here in combination for the first time using a contaminated mine site as a case study to demonstrate the benefits of using such an approach to identify hotspots of bioavailable metal most likely to cause negative impacts to biological receptors. Although not comprehensive from a temporal point of view, sufficient samples were taken over a 6 month period to provide excellent spatial distribution and to demonstrate the benefits of the approach when considering contaminated land remediation.

The main objective of the research was to identify and propose a risk assessment framework utilising available methods (chemical fractionation and modelling) capable of estimating the potential bioavailability of metals in soil, spoil, sediment and water at a contaminated site and demonstrate its benefits via a case study. The case study was based at Wheal Betsy an abandoned silver-lead mine which has been shown to be contaminating Cholwell Brook, a tributary of the Tavy (Fig. 1). Specific objectives to achieve these aims were to: (1) Utilise chemical and model-based methods to determine the mobility and potential bioavailability of key metals in soil, spoil, sediment and water (2) identify the major sources and pathways of heavy metal contamination into surface waters using spot samples and apportioning loads where possible; (3) demonstrate how impacts on receptors may be measured by using benthic-macroinvertebrates as biological indices.

2. Materials and methods

2.1. Study area

The study was conducted at Wheal Betsy, a former Pb-Ag mine on a north-south lode of the Culm Measures (shales and thin sandstones) located on the north-west edge of Dartmoor, Devon, UK (Ordnance Survey grid reference SX 51012 81385). Records indicate that over its operation lifetime (1806–1877), 400 t of Pb and 113 kg of Ag were mined and processed on site (Booker, 1967). Mineralogy can be divided into three areas covering 59,300 m³ Turner (2011); (1) the northern slopes dominated by steeply sloping spoil tips, comprising of coarse gravels, pebbles and cobbles; (2) the southern slopes which are a collection of finely grained spoil tips, varied in colour (yellow clays, orange sands and grey slates) and typical of mineral processing and; (3) the stream valley bottom. Cholwell Brook flows south down a steep valley through the highly contaminated areas of mine waste, and then into the River Tavy 3 km downstream, a main tributary of the River Tamar which flows into the English Channel at Plymouth. The mine's main adit and spoil tips at Wheal Betsy are an important source of Cd, Cu, Pb and Zn.

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