



Source term characterisation using concentration trends and geochemical associations of Pb and Zn in river sediments in the vicinity of a disused mine site: Implications for contaminant metal dispersion processes

Ian D. Pulford^{a,*}, Angus B. MacKenzie^b, Shane Donatello^{a,1}, Laura Hastings^{a,2}

^a Environmental Chemistry, Chemistry Department, WestCHEM, University of Glasgow, Glasgow, G12 8QQ, UK

^b Scottish Universities Environmental Research Centre, Rankine Avenue, Scottish Enterprise Technology Park, East Kilbride, G75 0QF, UK

Metal concentrations and fractionation have defined the source characteristics of Pb and Zn dispersed from a disused mine and transported by fluvial processes.

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ABSTRACT

River sediment at a disused lead–zinc mine was analysed to provide an understanding of the chemical nature of the source term for contaminated sediment exported from the site. Changes in concentration and geochemical associations of Pb and Zn were measured using aqua regia digestion and the BCR sequential extraction procedure. Sediment in the immediate vicinity of the mine was highly contaminated with Pb (max. c. 11,000 mg kg⁻¹) and Zn (max. c. 30,000 mg kg⁻¹), but these values declined rapidly within 1 km of the mine due to dilution and hydraulic sorting. Lead fractionation changed from being predominantly in the reducible fraction to being in the acetic acid-extractable fraction, whereas Zn was predominantly in the residual fraction. This material is transported as fine sediment in the river system.

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1. Introduction

Contamination of fluvial sediments with heavy metals derived from mine waste dumps is a global problem, with examples of recent research coming from Europe (Bertin and Bourg, 1995; Rowan et al., 1995; Hudson-Edwards et al., 1996; MacKenzie and Pulford, 2002; Brydie and Polya, 2003; Carter et al., 2006; Ettler et al., 2006; Kraft et al., 2006), North America (Brandvold et al., 1995; Hulshof and Macdonald, 1998; Miller et al., 1998; Hochella et al., 2005), South America (Segura et al., 2006; Villarroel et al., 2006), Asia (Song et al., 1999; Wen and Allen, 1999), Africa (Pettersson and Ingri, 2001; von der Heyden and New, 2004) and Australasia (Clark et al., 2001; Harrison et al., 2003; Ashley et al., 2004). These studies have been prompted by the need to have a better understanding of the processes involved in dispersal of heavy metals from such sites, and the resultant risks posed to both

human and environmental health. In this context, fluvial transport represents a major pathway in the pollution linkage model (source–pathway–receptor), which underlies current thinking in terms of contaminant dispersal and the risk posed by contaminants (Kibblewhite, 2001). Understanding such processes could also contribute to better management practices for rivers and their catchments (Carter et al., 2006; Macklin et al., 2006). Most of the focus in the above studies has been on sediment-associated transport, as this often accounts for >90% of heavy metal movement in streams and rivers (Gibbs, 1977; Miller, 1997), although understanding of this process is poor compared to understanding of transport of dissolved species (Hudson-Edwards, 2003).

Macklin et al. (2006) highlighted the difficulty of identifying the specific source, or sources, of heavy metals, and their relative contributions to contaminant loads, in a river system. With the introduction of more stringent controls on the discharge of heavy metals into streams and rivers, especially in North America and Western Europe, often accompanied by a decline in metal mining activities in those regions, the role of historically dumped waste at disused mine sites, or previously dispersed sediment in river channels and floodplains, has become more important over recent years (Macklin et al., 2006). MacKenzie and Pulford (2002) highlighted the importance of evaluating the impact of point sources of

* Corresponding author: Tel.: +44 0141 330 5950; fax: +44 0141 330 4888.
E-mail address: i.pulford@chem.gla.ac.uk (I.D. Pulford).

¹ Current address: Department of Civil and Environmental Engineering, Imperial College, London SW7 2AZ, UK.

² Current address: Wren & Bell, 11 Chester Street, Edinburgh EH3 7RF, UK.

contamination, such as waste dumps on disused mine sites, and distinguishing metal from such sources from regional background and global diffuse sources. Lead is an element that is particularly suited to such studies, as it is widespread in the environment as a result of dispersal due to anthropogenic practices, and the use of Pb isotope atom ratios is well-established as a diagnostic technique for characterising contaminant sources (Farmer et al., 2000).

The present work focuses on processes involved in dispersal of lead and zinc from a disused mine at Tyndrum, Scotland (Fig. 1), where the ore is characterised by $^{206}\text{Pb}/^{207}\text{Pb}$ and $^{208}\text{Pb}/^{207}\text{Pb}$ atom ratios of 1.146 and 2.432, respectively (MacKenzie and Pulford, 2002). These ratios distinguish the Tyndrum ore from the general geological background for this area, with a $^{206}\text{Pb}/^{207}\text{Pb}$ ratio in the

range 1.16–1.18 (Bacon et al., 1992; Sugden et al., 1993). The major diffuse sources of Pb from anthropogenic origins were from the industrial use of Pb ore from the larger Leadhills deposit in southern Scotland ($^{206}\text{Pb}/^{207}\text{Pb}$ atom ratio 1.173 (Sugden et al., 1993)), burning of Scottish coals ($^{206}\text{Pb}/^{207}\text{Pb}$ atom ratio 1.181 (Farmer et al., 1999)), and petrol-derived Pb resulting from the use of tetra alkyl lead as an antiknock agent in petrol up until 1999, which had an average $^{206}\text{Pb}/^{207}\text{Pb}$ atom ratio 1.076 (Farmer et al., 2000). MacKenzie and Pulford (2002) utilised these characteristic differences in stable Pb atom ratios to demonstrate contamination of river bed sediments with Tyndrum-derived Pb extended to at least 6.5 km from the site, but the mechanism of contaminant dispersal was not defined.

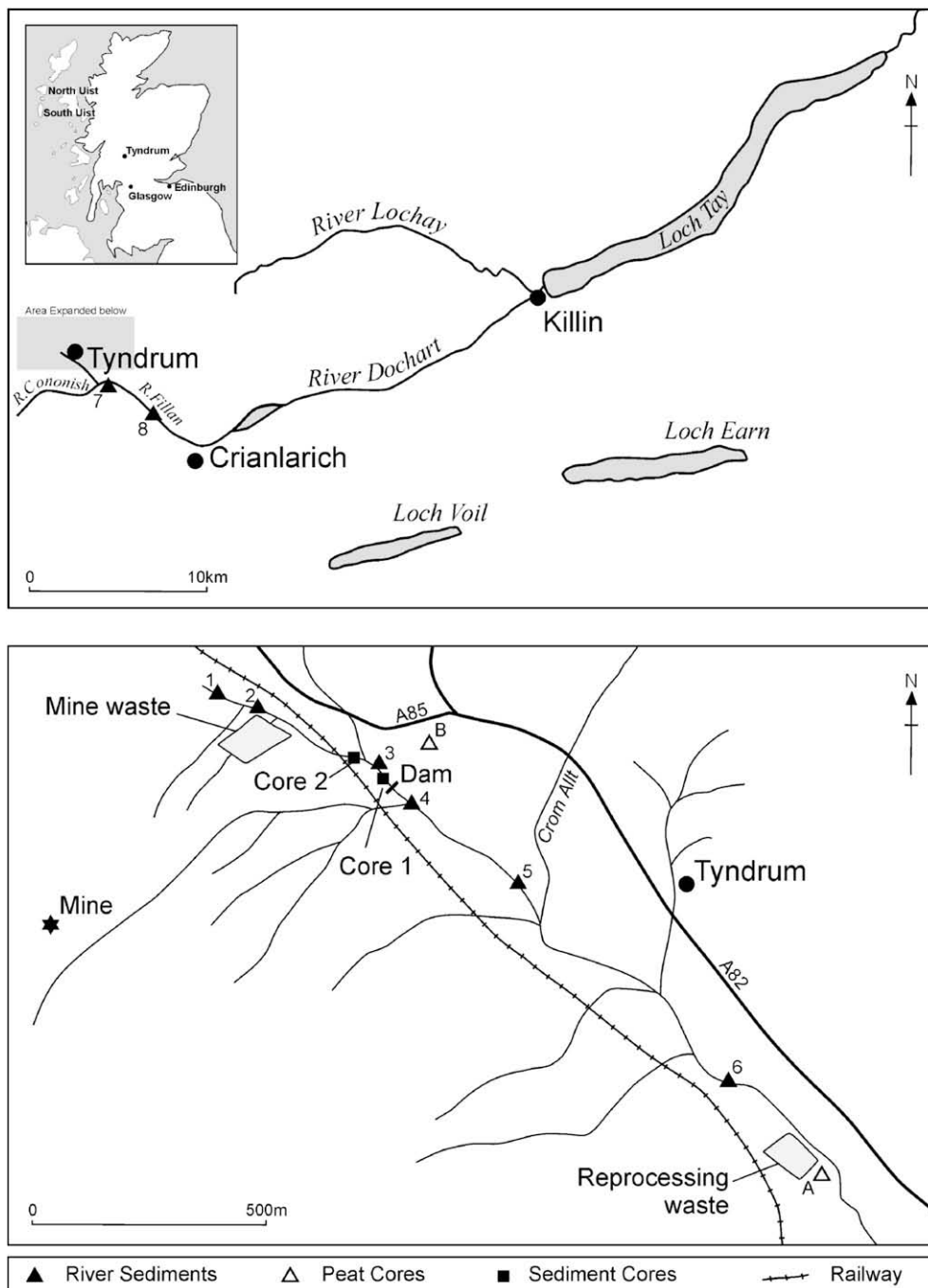


Fig. 1. Maps showing the location of the mine site, sampling areas 1–8 and the sites of cores 1 and 2.

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