Science of the Total Environment 408 (2010) 3576–3583

Contents lists available at ScienceDirect

Science of the Total Environment

journal homepage: www.elsevier.com/locate/scitotenv

Inventory of aquatic contaminant flux arising from historical metal mining in England and Wales

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ARTICLE INFO

Article history:
Received 31 January 2010
Received in revised form 14 April 2010
Accepted 15 April 2010
Available online 21 May 2010

Keywords:
Metal mine
Pollution
Flux
Mine water
Inventory

ABSTRACT

The impact of discharges from abandoned metal and ironstone mines has been a much studied form of aquatic pollution in recent decades. Few attempts however, have been made to accurately determine the overall contaminant mass flux arising from abandoned mine sites at scales above catchment level. Such assessments are critical to determine the significance of former mining to national, regional and ultimately global trace metal flux. This paper presents the most comprehensive national survey to date across England and Wales of the total pollution burden discharged at source from abandoned non-coal mine sites. 338 discharges have been identified (from 4923 known abandoned metal mines) and while concurrent flow and contaminant concentration records are only available for around 30% of these, significant quantities of metals (and As) have been quantified to be discharged. A minimum of 193 tonnes of Zn, 18.5 tonnes of Pb, 0.64 tonnes Cd, 19.1 tonnes of Cu, 551 tonnes Fe, 72 tonnes Mn and 5.1 tonnes As are released in water discharges from abandoned non-coal mines to the surface water environment of England and Wales each year. Precautionary extrapolation of mass fluxes based on the frequency distribution of measured concentration and flow data, for discharges with absent data, suggests that the actual total mass flux for these contaminants could be up to 41% higher. The mass flux of Pb released from mines exceeds that of all currently permitted discharges (e.g. active industrial sites and wastewater treatment works) to surface waters across England and Wales, while those of As, Cd and Zn are of a similar magnitude. These data put into context the enduring legacy of historic mining on the water environment, highlighting its significance relative to more highly regulated polluting sites. Comparison of the figures with estimates of global trace metal flux suggests that the national total identified here is significant on a global scale.

1. Introduction

Anthropogenic perturbation to global trace metal cycles has long been identified since the pioneering atmospheric trace metal inventories of Nriagu (1979) and Lantzy and Mackenzie (1979). For the water environment, assessments of global trace metal flux and associated human impacts have been developed since the study of Martin and Meybeck (1979), which provided the first comprehensive overview of river particulate composition and associated global river fluxes. Over the past three decades the average elemental composition (both particulate and dissolved components) of many important global rivers has become better characterised and efforts to develop and update global databases (e.g. Poulton and Raiswell, 2000; Carey et al., 2002, Gaillardet et al., 2003; Viers et al., 2009) have consistently alluded to the major human perturbation to the mass flux of trace metals in river systems. The mining and associated processing of metalliferous ores have long been documented to cause acute and persistent damage to the water quality of receiving water courses over scales from individual streams (e.g. Kimball et al., 2002; Nuttall and Younger, 1999), to entire river basins (e.g. Sarmiento et al., 2009), up to national level (e.g. Mayes et al., 2009). In their benchmark inventory of global trace metal cycles, Nriagu and Pacyna (1988) made the first and most comprehensive estimate to date of the total mass flux arising from the base metal mining and refining sector for a suite of metals and metalloids. Few attempts have been made since then to provide a detailed regional or global flux estimate for the mining sector on emissions to the water environment due to (a) the lack of suitably comprehensive datasets of source discharge flow and quality on which to base such assessments and (b) the short half lives of trace metal contaminants in the aqueous phase as they rapidly transfer from water column to fluvial sediments (Nriagu and Pacyna, 1988). The latter point renders assessments of metal discharge flux at source from ambient water quality monitoring stations problematic since they usually lie some distance downstream from polluting sources. The resultant contamination of sediments that arises due to instream attenuation processes of aqueous contaminants has been

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doi:10.1016/j.scitotenv.2010.04.021

well characterised in many mining-impacted streams (Macklin et al., 2006) and the longevity of the sediment-borne metal pollution equally well-described (e.g. Coulthard and Macklin, 2003; Dennis et al., 2009). However, assessments of trace contaminant release at source are essential for fully appreciating the ongoing pollution inputs from abandoned mines into the surface water environment which ultimately contribute to the secondary sources of contaminated sediments in mining-affected river systems.

Regional studies of one major mining region and comparison with global trace metal flux recently undertaken by Sarmiento et al. (2009), have suggested that the heavily mining-impacted Odiel river basin, which drains the Iberian Pyrite Belt in Spain contributes up to 15% of the global dissolved Zn flux to the oceans as quantified by Gaillardet et al. (2003). Similar perturbations to global sulphur cycles have also recently been suggested to occur due to coal mining activity (Raymond and Oh, 2009). While acknowledging the large uncertainty surrounding extrapolations from two long-term datasets from two large catchments in Pennsylvania, Raymond and Oh (2009) suggest that sulphur released through the generation of acidic coal mine drainage could account for between 28 and 40% of the global riverine sulphate flux produced through pyrite oxidation.

These studies certainly suggest the stark impact that historic mining activity can have on global scale pollutant fluxes. Given that such sizeable contributions are determined from localised, albeit acutely polluted systems, and mining-derived water pollution has been well-documented in every continent of the world except Antarctica (e.g.; Kuma and Younger, 2004; Lottermoser et al. (1999); Kimball et al., 1999; Van Damme et al. (2008); He et al., 1997; Younger et al., 2002), it is likely that the mining-derived contribution of riverine metal fluxes is globally significant.

This paper presents results from a national assessment of polluting abandoned metal and ironstone mines in England and Wales. Related previous work has summarised the methodology for collating mine data and objectively assessing the impacts of abandoned non-coal mine sites on the aquatic environment nationally (Mayes et al., 2009). The data presented here provide an inventory of the mass loading of common contaminant metals (and As) discharged at source by base metal and ironstone mines into the surface waters of England and Wales. As such, it provides a case study for one region of globally important historic mining, which is used to highlight, (1) the significance of pollution from a relic industrial sector (the peak of base metal mining in the UK was between 100 and 200 years ago) compared to contemporary sources of aquatic pollution nationally and (2) consider the significance of the mining-derived mass fluxes in terms of global trace metal cycles.

2. Methods

As part of a national assessment of pollution arising from abandoned metal mines in England and Wales (see Mayes et al., 2009, for detail), specialists from the Environment Agency (the government environmental regulator for England and Wales) were asked to populate a database with details of known polluting discharges from abandoned mines. Such discharges comprise drainage adits, shafts and runoff from spoil heaps or other metal rich wastes. A range of information was requested from the EA respondents (who encompassed specialist environmental scientists, hydrogeologists, chemists and biologists) including the location and nature (e.g. point or diffuse discharge) of any discharges and their impacts on downstream ecology and water resources (see Mayes et al., 2009). Crucial to the assessment undertaken here however, were data detailing mean flow rates and mean metal (and As) concentrations for identified discharges. Total (unfiltered) concentration data were requested. Such information is collected during both ambient and campaign water quality monitoring undertaken by the Environment Agency and analysed in accredited laboratories in accordance with statutory water quality monitoring protocols. Concentration data were requested for As, Cd, Cu, Fe, Mn, Ni, Pb and Zn, which represent the most common (and most commonly measured) pollutants discharged from abandoned metal mines in England and Wales (Mayes et al., 2009). These concentration and flow data were used to compute an annual mass flux (in tonnes/year) for each contaminant at each site. Flow data for mine sites are sparse in many cases. Where flow data are available, it ranges between base-flow spot measurements usually undertaken during sampling campaigns for informing remediation feasibility assessments, to continuous flow measurement (e.g. calibrated weirs with stage loggers) at some of the larger discharges. In addition to the data return from the Environment Agency, a thorough review of published and grey literature (e.g. student theses and contract reports by environmental consultants) has been undertaken to collate currently available information detailing flow and/or chemical composition of metal mine discharges in England and Wales. Furthermore, manual collection of mine discharge flow and water quality data was undertaken at several sites known to the authors where data were absent in the return from the EA. Where manual data collation was undertaken (for 25 mine discharges), flow rates were measured using the velocity–area method (Shaw, 1994) and a Valeport 301 velocity meter with a flat electromagnetic sensor during base-flow conditions (July–August 2009). This device was particularly suited to measuring velocity in mine drainage adits given the typically shallow depth of water (generally <0.1 m) with smooth, almost laminar flow. Total metal (and As) concentrations in acidified (with 5 drops of concentrated trace element grade HNO_3) 150 mL samples collected in accordance with standard water quality sampling procedures (APHA, 2005) were analysed using a Varian Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES). Reliability of sample analyses was tested by comparison with certified reference materials.

3. Results and discussion

3.1. Contaminant flux at point of discharge

Fig. 1 illustrates the geographic distribution and relative magnitude of the contaminant flux from abandoned base metal and ironstone mines, while Table 1 provides the summary flux totals of the 338 mines. Sample data for some of the most polluting sites are given in Table 2. In terms of total elemental flux, that of iron exceeds the individual mass fluxes of any other monitored contaminants (Table 1). A significant portion of this Fe release arises from ironstone mine discharges in the Cleveland Ironstone Field in the Northumbria River Basin District (RBD) (see Younger, 2000a), accounting for 25% of the total Fe release. Two other single discharges, the Dyffryn Adda Adit draining the Parys Mountain copper mine complex on the Isle of Anglesey (Western Wales RBD: Fig. 1) and the County Adit (South West RBD), which underdrains a large portion of the Cornwall tin mining district account for 34% and 26% respectively of the total Fe release nationally. Fe concentration in the discharges shows a bi-modal distribution with 57% of the data clustering below total Fe concentrations of 0.5 mg/L and a second peak in Fe concentration in between 20 and 40 mg/L in which 12% of the data fall. This reflects the fact that relatively few base metal mines have significant quantities of pyrite, marcasite (FeS_2) and/or siderite ($FeCO_3$) associated with the extracted ores. Areas where there is appreciable Fe in base metal discharges include parts of the Western Wales RBD in Ceredigion, many of the discharges from the Sn–Cu mining district of Cornwall (South West RBD) and some discharges in the North Pennines which, along with the Cleveland ironstone mines (Northumbria RBD), account for the secondary, higher peak in Fe concentration.

Manganese is often associated with Fe in the mine discharges (Pearson's r on log-normalised data is 0.86, $P<0.001$) and as such, the