Evaluating metal behaviour and mine water treatment benefits in abandoned mine catchments with variable pollutant load inputs

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Abstract

The results of three synoptic survey investigations of the importance of diffuse mining pollution are reported. The three watersheds range in size from 10 – 3 000 km². In all three of them diffuse pollution – from a combination of groundwater inputs, waste rock runoff and sediment remobilisation – becomes increasingly important at higher river flow-rates. Whilst point source remediation is predicted to result in substantial improvements in downstream water quality during low flow conditions, at higher flows reductions in downstream metal concentrations may be less than 30%.

Introduction

Abandoned metal mines are a major source of freshwater pollution in the UK. In excess of half of the entire mass flux of zinc and cadmium to the streams and rivers of England and Wales arise form abandoned metal mines (Mayes et al., 2010), with less than half coming from all other sources of pollution combined. Consequently there is an ongoing programme of UK government investment in designing and building treatment systems to lower the burden of metals entering freshwaters in abandoned metal mine districts. In most cases the treatment initiatives are targeted at point sources of pollution i.e. from abandoned mine entrances, since it is comparatively straightforward to (a) characterise such discharges in terms of flow-rate and water quality and (b) engineer a structure to capture such discharges and direct them into a treatment system.

However, there is a growing body of evidence that point source pollution is not the only cause of degradation of downstream water bodies (e.g. Mighanetara et al., 2009). Diffuse sources of mine water pollution – such as arise due to surface runoff from mine waste rock, direct inputs of groundwater to surface waters, and remobilisation of metals from riverine sediments - may be a variably important source of pollution. The measurement, interception and treatment of such sources of pollution is far more challenging than equivalent interventions for point source pollution. Properly quantifying diffuse source pollution is important though, as without such understanding it is not possible to calculate the true benefits of – potentially costly - point source pollution remediation schemes in the same watersheds. Ultimately, as more and more point source treatment systems are installed, remediation of diffuse pollution sources will become the limiting factor to further improvements in river water quality in mined watersheds.

Over the last 5 – 10 years we have undertaken long-term synoptic monitoring in contrasting abandoned mine watersheds to fully characterise the nature and extent of diffuse mining pollution, and therefore to understand the benefits of point source remediation. Particular challenges in executing such monitoring programmes are (a) accurately measuring flow-rate and (b) monitoring across the full range of hydrological conditions and (c) monitoring in sufficient locations to derive the importance of diffuse pollution sources during any single set of hydrological conditions. Here we report on the results of those monitoring programmes and the implications of the work.

Study areas

Three mining-impacted rivers were the focus of the investigation:

1) The Coledale Beck is an upland river in the English Lake District National Park (Cumbria), with a watershed area of approximately 10 km^2. At an elevation of 280 m above sea level (a.s.l) is the abandoned Force Crag lead / zinc / barytes mine (54°35'00"N 3°14'23"W), which was finally abandoned in 1991. In addition to a major point source of mine water pollution, and several smaller ones, there are extensive areas of waste rock which are obvious candidate sources of diffuse pollution. The Coledale Beck drops steeply, from an elevation of 550 m to 100 m above sea level over a horizontal distance of 4 km. Further details on the geology and mining in the area can be found in Postlethwaite (1913).
2) The River Nent is also in Cumbria, and it too is an upland river. It is a tributary of the River Tyne, which drains east to the North Sea. The River Nent watershed lies in the upper reaches of the River Tyne system, and has an area of approximately 100 km2. The watershed was extensively mined for lead and zinc. There are four major point source discharges to the River Nent, two in the vicinity of the village of Nenthead (450 m a.s.l; 54°47'09"N 2°20'30"W), and the other two some 2 km downstream. Upstream of these point discharges there are extensive tracts of exposed waste rock.
3) The River Tyne is one of the largest river systems in northern England, with a watershed area of nearly 3 000 km^2. It has two main branches: the River North Tyne and River South Tyne (the River Nent is a tributary of the latter), which meet 2 km upstream of the town of Hexham, at 54°59'19"N 2°07'50"W. Within the watershed of the River South Tyne is the North Pennine orefield, which was one of the most productive mining areas (for Pb and Zn) in the UK during the 18th and 19th centuries. The Environment Agency (England and Wales) has flow gauging facilities at several locations down the length of the River Tyne, which were critical during this investigation.

Location maps are not shown here for reasons of space. However, the results reported below are exclusively comparing the metal flux of an individual point source, or sum of point sources, compared to metal flux at an in-stream location (usually downstream), across hydrological conditions. It is the comparison between point source flux and in-stream flux that is key, rather than the exact locations of the various points.

Methods

The key to deriving the importance of diffuse pollution sources is the synchronous measurement of flow-rate and water quality of both point sources and in-stream locations. In-stream locations includes both upstream and downstream of the point sources, and upstream and downstream of suspected / potential diffuse sources. In order to be able to compare metal flux (mass per unit time) at different locations sampling at all locations had to be undertaken within a single day, and whilst hydrological conditions were constant. To understand the importance of diffuse pollution under different hydrological conditions it was necessary to sample on different days when hydrological conditions were different. In order to capture as wide range of hydrological conditions as possible monitoring was undertaken over at least a period of 12 months.

On the Coledale Beck one Environment Agency flow gauging structure was available for use. This was located immediately downstream of the Force Crag mine site, and therefore was used routinely. Sharp-crested V-notch weirs have been installed on many of the smaller streams and discharges in the watershed. At other locations on the river salt gulp dilution gauging was undertaken periodically to measure flow-rates at key locations. In brief, field methods involved injection of up to 7 kg of lab-grade NaCl into the river, with downstream semi-continuous conductivity logged using a YSI ProPlus conductivity meter. 2 × 1 L river water samples, and 2 × 25 mL salt solution samples, were collected for the subsequent calibration required for flow calculation (Herschy, 1998). On the River Nent there are no flow gauging structures, and therefore salt gulp dilution gauging was used for determining flow at all locations. On the River Tyne use was made of flow gauging infrastructure installed and operated by the Environment Agency (England and Wales). In particular, use was made of two gauges on the River South Tyne, at Featherstone and Haydon Bridge, and a third on the River Tyne at Bywell.

Water samples were collected simultaneous to flow measurement. Samples were collected in 30 mL plastic viles, with those for subsequent metals analysis acidified with lab-grade concentrated nitric acid. Samples for filtered metals analysis were filtered through a 0.45 µm cellulose nitrate filter. pH, conductivitiy and temperature were measured on site using a pre-calibrated Myron L 6P Ultrameter. Total alkalinity was determined at the time of sample collection using a Hach Digital Titrator with 1.6N sulphuric acid or 0.16N sulphuric acid with a Bromcresol-Green Methyl-Red indicator. Analysis of total and filtered metals was subsequently undertaken using a Varian Vista-MPX ICP-OES or Agilent 770 Series ICP-MS, as appropriate for the metal concentration. Blanks and standards were used throughout and triplicate samples analysed periodically. Anion analysis was conducted using a Dionex DX320 ion chromatograph.

Results and Discussion

Coledale Beck: There is a single major point source of pollution from the Force Crag mine to the Coledale Beck (referred to as Level 1). It is a groundwater discharge from the abandoned underground workings, and consequently has a rather consistent flow-rate, as indicated on Figure 1. It also has a consistent Zn flux, varying from 2.9 to 4.3 kg/d. During low flow conditions this point source of pollution accounts for the majority of the Zn in the Coledale Beck downstream of the mine site (Site ID #4; Figure 1). However, as flow-rate in the Beck increases the Zn flux in the river increases sharply also, and these increases are not accounted for by the Level 1 point source (flow of the Coledale Beck during monitoring event 5, on Figure 1, was 390 L/s). Under the highest flow conditions monitored during this work Zn flux in the Coledale Beck reached 13.9 kg/d, with the Level 1 point source representing only 26% of this total (3.65 kg/d).

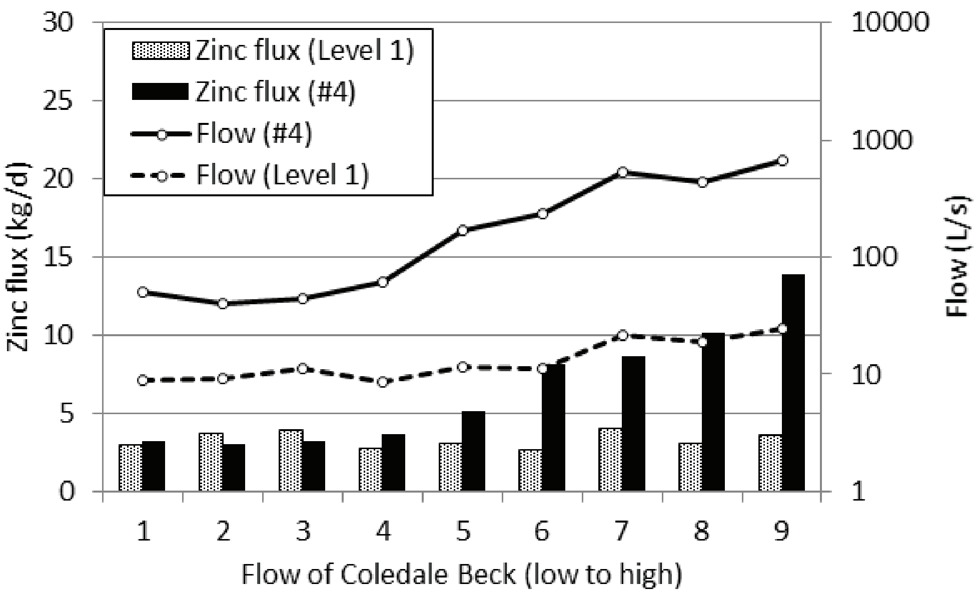

__Figure 1__ Zinc flux of the major point source (Level 1) of pollution to the Coledale Beck and the Coledale Beck itself (site ID #4), as a function of flow-rate in the Beck (Flow on monitoring event 1 was 111 L/s and on monitoring event 9 it was 1 487 L/s).

From these data it is possible to calculate the improvements in water quality arising from treatment of the major point source of pollution, Level 1. Under low flow conditions in the Coledale Beck more a substantial reduction in Zn concentration in the Beck should be feasible (over 90% reduction). However, as flow-rate in the Beck increases, and diffuse pollution sources such as waste rock runoff and groundwater inputs increase in importance, the benefits of treatment diminish; at the highest flow rates point source treatment is predicted to result in Zn concentration reductions in the river of less than 20%.

River Nent: There are four major point source mine water discharges to the upper River Nent: Rampgill, Capelcleugh, Haggs and Croft. The Haggs and Capelcleugh discharges are the most important point sources of pollution to the River Nent in terms of Zn pollution (the Haggs flux is shown on Figure 2). The Capelcleugh discharge is the most important source of Cd and Ni, and the Haggs discharge is the second most important. Point mine water discharges are of limited significance as sources of Pb pollution to the River Nent.

For the range of flow conditions monitored during this investigation the point sources of mine water pollution were responsible for between 37% (at higher flow conditions) and 85% (at lowest flow conditions) of the filtered Zn flux of the River Nent downstream (Zn is predominantly present in its filtered form; Figure 2). The Haggs and Capelcleugh discharges are the most important contributors of Zn (and Cd and Ni); the Haggs discharge accounted for between 14% (at higher flow conditions) and 33% (at lowest flow conditions) of the Zn flux of the River Nent during this investigation, and equivalent figures for the Capelcleugh discharge were 14% and 34% for higher and lower flow conditions respectively (Figure 2). Mass balance calculations suggest that treatment of either of these discharges (to remove 90% of the Zn) would lower Zn concentrations in the River Nent by between approximately 15 – 30%. Equivalent calculations for the Rampgill and Croft discharges suggest that Zn concentrations in the River Nent would be lowered by

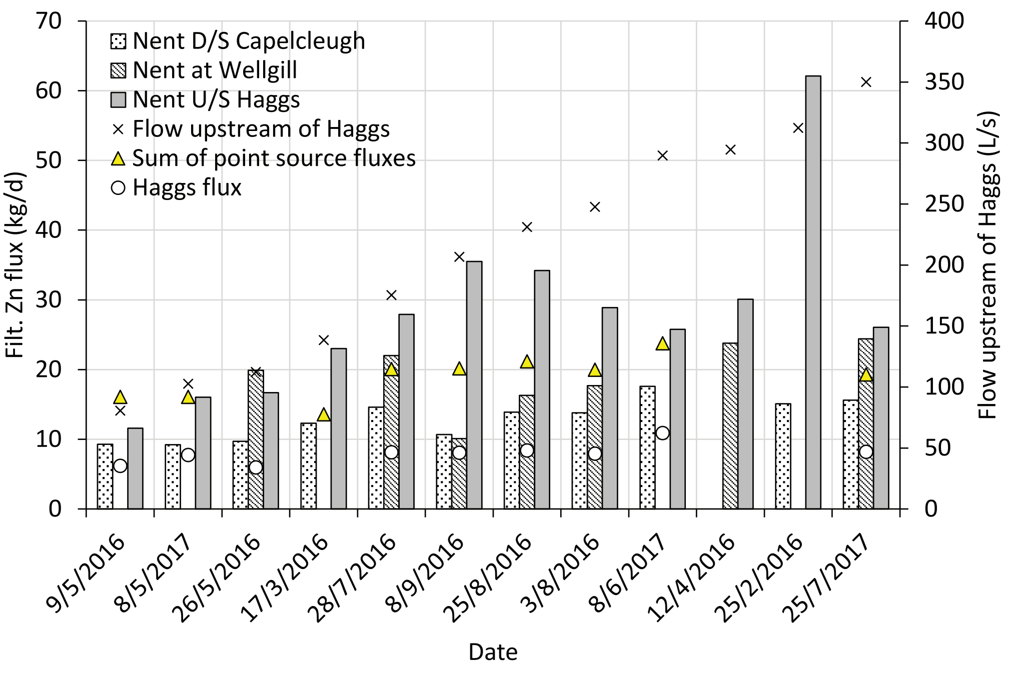

Figure 2 *Filtered zinc (< 0.45 μm) flux of point sources and in-stream locations on the River Nent, as a function of flow-rate in the river*

no more than 10% and 5% respectively. These figures are likely to be best case scenario. This is because the flow conditions in the River Nent during the investigation were not particularly high; at higher River Nent flow-rates the benefits of point source treatment are likely to be less, due to the increasing importance of diffuse source pollution during such hydrological conditions.

Even under the relatively low flow conditions during this investigation diffuse metals pollution has been shown to be significant in the River Nent catchment. As has been found elsewhere, diffuse pollution becomes more important at higher flows. It is difficult to be certain, but it appears as though direct inputs of polluted groundwater may be the most important source of diffuse metals pollution in the study reach of the River Nent.

River Tyne: The Zn flux from the River Nent (above) discharges to the River Tyne. For the data presented here, at its highest the flux from the River Nent is a little over 60 kg Zn/d (Figure 2). However, even though the River Nent is the most important source of metals to the River Tyne, the zinc flux from it is minor compared to that in the lower reaches of the River Tyne, which at the highest flows recorded exceed 15 000 kg/d (Figure 3). What is notable from Figure 3 is that above a flow-rate of around 300 L/s particulate Zn (i.e. associated with suspended sediment) begins to dominate the total flux of metal. Metal-polluted waste rock and sediments are distributed throughout the River Tyne catchment, in the river itself, in river bank materials, and on floodplains. Although filtered Zn concentrations increase somewhat as flows increase, the very highest fluxes are a consequence of resuspension and transport of the finer, metal-polluted, sediments in the lower-lying reaches of this mature river. This is in contrast to the Coledale Beck and River Nent, in which the Zn flux is dominated by filterable metal in all but the very highest flow conditions (not recorded here). This point is shown in Figure 4, which shows that on all but one occasion (n = 17) more than 90% of Zn was present in its filterable (< 0.45 µm) form. Thus, the form of the diffuse Zn in the lower lying River Tyne contrasts with its form in the more juvenile, upland rivers which have far less fine particulate matter (silts and sands).

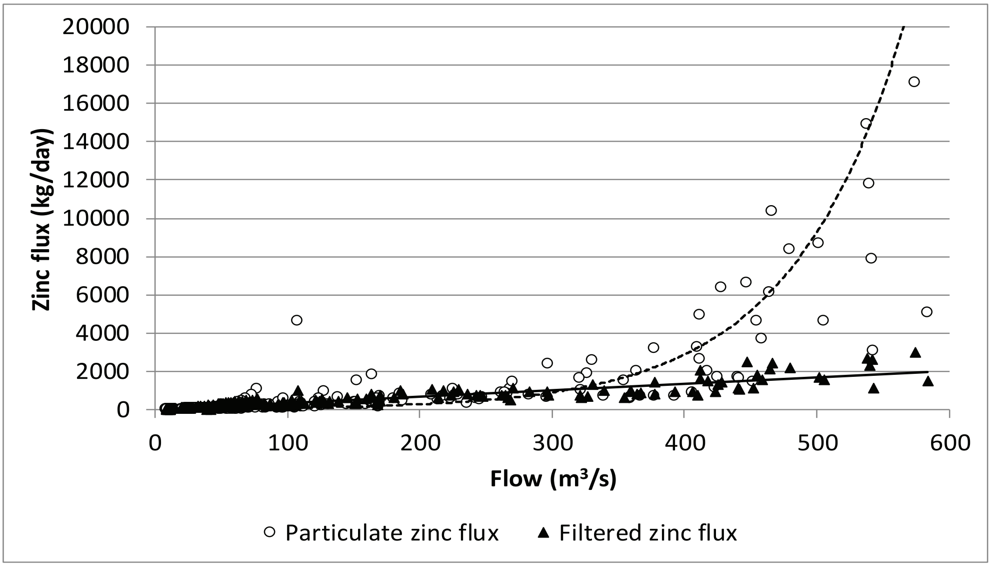

Figure 3 *Particulate and filtered zinc flux in the River Tyne under varying hydrological conditions*

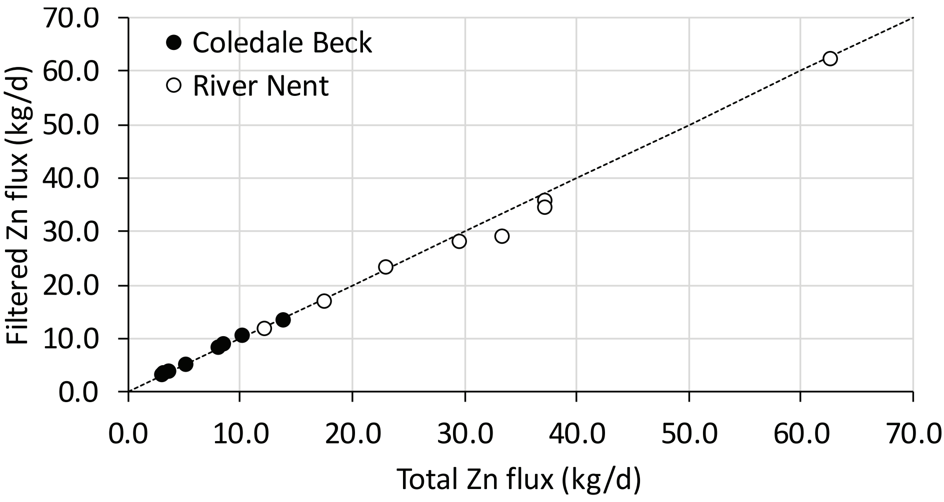

Figure 4 Total and filtered zinc concentrations in the Coledale Beck and River Nent, Cumbria, illustrating that zinc is primarily present in filterable (< 0.45 μm) form

Data in Figures 3 and 4 suggest that point source remedial interventions in the River Nent are unlikely to result in any substantive reduction of Zn flux in the lower reaches of the River Tyne. However, two points need to be borne in mind:

1) Zn in point sources of pollution on the River Nent may be held in transient storage in finer sediments downstream (in the River Tyne), and therefore effectively act as a diffuse source when they are remobilised during higher flow events i.e contribute to the particulate Zn flux shown in Figure 4.
2) Monitoring exercises such as those described here fail to quantify metal flux transported as bed load (via traction and saltation), which may become important sources of pollutants in the water column following erosion.

Therefore remedial interventions for point sources of pollution may in fact result in somewhat greater benefits than implied by Figures 3 and 4 alone.

Conclusions

The results of investigations of the importance of diffuse abandoned mine pollution in three contrasting watersheds are considered. In the upland catchments of the Coledale Beck and River Nent point source pollution is key to in-stream metal flux at low flow conditions. However, at higher river flows diffuse pollution sources become increasingly important. Under such conditions diffuse sources, such as groundwater inputs and waste rock runoff, start to dominate metal flux. Consequently the potential benefits of point source remediation diminish substantially under such hydrological conditions. In the large, lower-lying, Rive Tyne catchment (downstream form the River Nent) very high zinc fluxes are evident at higher flows. In contrast to the upland river systems, here it is particulate zinc that is a key contributor to the absolute flux, which in essence reflects the long term impacts of widespread historic mining pollution. As point source pollution remediation initiatives become more widespread in the UK, diffuse sources of abandoned metal mine pollution will increasingly become the limiting factor to further improvements in water quality.

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Environment Agency, or any other organisation mentioned herein.

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