

Identifying diffuse sources of inorganic pollutants in post-industrial catchments

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ABSTRACT

The introduction of the EU Water Framework Directive (2000/60/EC) has put greater emphasis on addressing water pollution from a catchment management perspective. Diffuse inorganic pollutants derived from mining activity in post-industrial areas often represent a considerable obstacle in achieving compliance with the WFD. While there has been considerable progress in developing treatment systems for point sources of inorganic water pollution in recent years, there remains a gap in the characterisation and remediation of diffuse mine water sources.

This paper presents data from the River Gaunless catchment, a historically heavily-mined catchment in the north east of England, which as a consequence has a problem, with persistently high instream iron concentrations. Previous studies have highlighted the high contribution of diffuse mine water sources in the catchment, which may account for 45% of the instream iron loading in low flow conditions and up to 95 % of the instream loading in high flow conditions. However, the specific sources and locations of different diffuse contributors have not previously been clarified. These sources include surface runoff from exposed spoil, resuspension of ochre previously deposited on the riverbed and direct discharge of contaminated groundwaters to surface waters. In addition, there may be other iron-rich discharges within the catchment which supplement instream iron loadings, for example highways runoff and upland peat erosion. Analogous data from the River Allen catchment is also considered where similar issues have been encountered in a river basin impacted by lead and zinc-rich discharges from former metal-mining.

Some of the problems associated with characterisation and partitioning of diffuse sources at a catchment scale are considered. These include decisions on the nature and intensity of sampling regimes (which require extensive, synchronous flow and water quality data), and technical issues in identifying the provenance of instream metal loadings. Management options for the remediation of problematic diffuse sources are also considered.

INTRODUCTION

Compared to its European neighbours the UK has an unparalleled record in the construction and operation of mine water remediation schemes. Since 1997 the UK Coal Authority has been implementing a rolling programme of treatment initiatives to address metal-rich, and sometimes acidic, discharges from abandoned deep coal mines (see Younger *et al.*, (2002) for details of the causes and nature of mine water pollution). At the end of 2004 some 33 full-scale treatment schemes were in place across the former coal mining districts of England, Wales and Scotland, at a total capital cost of nearly £25 million (Jarvis *et al.*, 2005). These systems collectively treat some 100,000 m³/day of mine water, and retain in the order of 15,000 – 20,000 tonnes / year of iron-rich sludge which would otherwise have been discharged to the freshwater environment (disposal and / or re-use of this sludge is, in itself, a major issue) (Jarvis *et al.*, 2005).

Notwithstanding these successes the introduction of the EU Water Framework Directive (WFD) raises new challenges with respect to management of mining-related pollution. In undertaking its 'pressures and impacts assessment' exercise the Environment Agency determined that some 1,800 km of streams and rivers in England and Wales are "at risk" of

failing to meet WFD objectives due to mine water pollution, as are groundwater bodies with an extent of approximately 9,000 km² (www.environment-agency.gov.uk). As noted above, the UK has an effective approach to mine water remediation for deep coal mine discharges, but this rolling programme of initiatives is limited in that (1) the Coal Authority does not currently have a remit for addressing water pollution arising from either metal mines, or spoil heaps from coal or metal mines and (2) to date, with only one exception, all of the full-scale treatment systems in the UK remediate point sources of mine water pollution.

The purpose of this article is to illustrate the importance of diffuse sources of mining-related pollution to the overall quality of freshwaters in former mining districts. The paper draws on the outcomes of two ongoing investigations at catchments in County Durham and Northumberland. Direct observations, an understanding of hydrological pathways in abandoned mining facilities, and previous work, suggests that diffuse pollution may arise from a number of sources (Mayes *et al.*, 2005):

- 1) Diffuse seepages in the immediate vicinity of point discharges
- 2) Direct input of polluted groundwater to surface waters, via the hyporheic zone

- 3) Runoff from spoil heaps rich in sulphide minerals (especially pyrite)
- 4) Resuspension of metal-rich river bed and bank sediments

This paper reports the results of ongoing investigations of diffuse mining pollution in two catchments in the north-east of England. One of these catchments was predominantly a metal-mining catchment (the River Allen, Northumberland), and the other (the River Gaunless, County Durham) was principally mined for coal. By monitoring both the flow-rate and quality of all point sources of mine water pollution in these catchments, and also making equivalent measurements in the main river channels, it has been possible to determine that portion of the metal loading of the two main rivers which is attributable to diffuse inputs. The implications of the work, both for engineering interventions to address diffuse pollution, and for meeting the objectives of the WFD, are discussed.

STUDY SITES

The River Gaunless catchment covers an area of 93 km². The river itself drains east for a distance of 32 km before its confluence with the River Wear (Figure 1) at the town of Bishop Auckland, County Durham (National Grid Reference NZ 214307). In its mid- to upper-reaches the catchment is principally rural (mainly livestock farming) and the catchment is entirely underlain by Coal Measures strata. The area was extensively deep mined for 150 years up until 1976. Following cessation of mine pumping operations, groundwater levels in the catchment had recovered by 1979. Since then a number of uncontrolled discharges of mine

water, contaminated principally with iron, have caused contamination of the River Gaunless. As a consequence iron pollution is a persistent problem in much of the River, with total concentrations rarely less than 0.5 mg/L (Mayes *et al.*, 2005).

The River Allen is a major tributary of the upper River Tyne, occupying a 190 km² catchment approximately 30 km to the east of Newcastle upon Tyne (Figure 1). The Rivers East and West Allen meet at National Grid Reference NY 801588, approximately 6 km south of the confluence of the River Allen with the River South Tyne. The River Allen catchment was extensively mined for lead and zinc from the 17th Century until the early 1970s. The hydrology of the catchment is heavily influenced by mining features, most notably the 7 km-long Blackett Level, which discharges to the River East Allen in the town of Allenheads. Previous studies of the rivers, and in particular data reported here, show that zinc contamination is a particular issue in this catchment, with concentrations several orders of magnitude higher than current legislative standards in some reaches.

METHODS AND MATERIALS

Previous attempts have been made to quantify the proportion of diffuse and point iron loadings in the River Gaunless catchment (Younger, 2000), by utilising Environment Agency public archive data and derived flow data, since flow gauging in the Gaunless commenced only recently. To the authors' knowledge no concerted effort has ever made to quantify the zinc and lead loads associated with point mine water discharges along the River Allen catchment. This current research endeavours to quantify the in-stream metal loadings and point source contribution more accurately through employing synchronous sampling and flow gauging of both point mine water discharges and instream sample points throughout the two catchments under varying flow conditions. In addition, instrumentation of the point mine waters will facilitate more reliable estimates of flow. In the case of the River Gaunless, the sampling network has also been expanded from previous studies (Younger, 2000) to encompass sampling stations up to the catchment headwaters (previous sampling only went up to the settlement of Butterknowle, some 7km downstream of the first major point mine water discharge) and sampling of major tributaries along the course of the Gaunless. High-resolution reconnaissance surveys of the Rivers Gaunless and Allen (encompassing field walk-by and water sampling) have also aimed to identify any previously unknown point mine waters in the catchments to permit better quantification of point sources.

For water samples collected during the current research, two acidified polypropylene bottles were filled at each sample station, one of which was filtered using 0.2µm cellulose nitrate filters (to quantify dissolved metals in the samples) and one unfiltered (to quantify total metals concentrations in the sample). Samples were analysed for metals using an Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES). Flow at the mine water and in-stream sampling locations was measured via a suite of methods including fixed v-notch and rectangular notch weirs, current meter, Acoustic Doppler Current Profiler (ADCP), bucket-and-stopwatch and hydraulic equations for pipe flow (based on recorded average velocity).

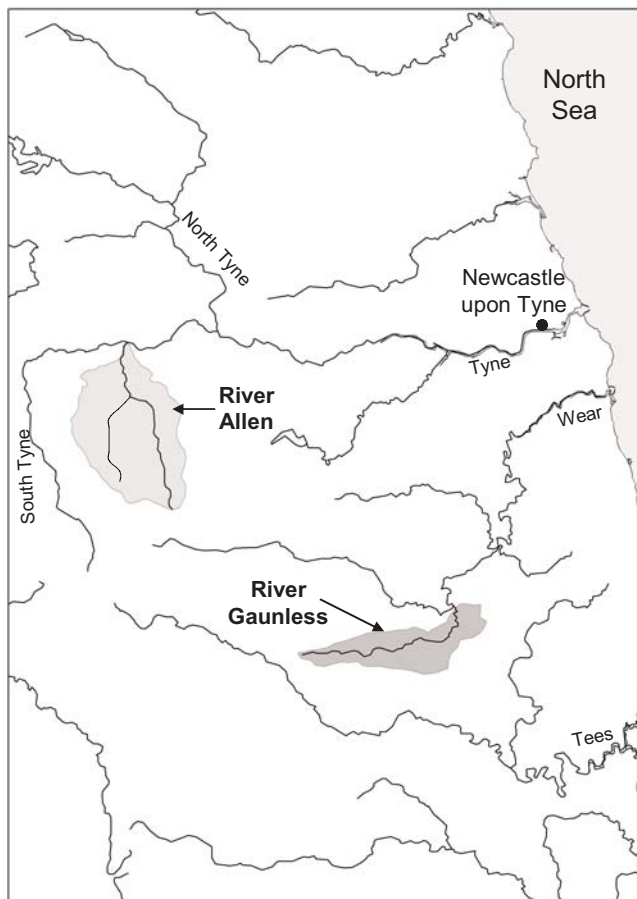


Figure 1: Locations of the River Gaunless and River Allen catchments, County Durham and Northumberland respectively.

RESULTS AND DISCUSSION

The River Gaunless

The River Gaunless catchment contains 6 known point sources of mine water pollution, as illustrated on Figure 2. Total iron concentrations of these discharges range from 3,140 µg/L (Arn Gill) to 26,000 µg/L (Fieldon's). The highest iron load to the river arises from the Lowlands 1 discharge, which has a flow-rate of approximately 25 L/s, and a mean total iron concentration of 7,400 µg/L. Total iron concentrations in the River Gaunless are commonly above 1,000 µg/L, as illustrated by the data in Table 1.

In themselves these discharges are a cause for concern in terms of the degradation of river water quality and ecology that results. For example, Firth *et al.* (1995) showed that the Biological Monitoring Working Party (BMWP) score, which is a measure benthic macro-invertebrate abundance and diversity, decreases from 134 upstream of the Lowlands discharge to 84 downstream of it. However, a key objective of the current work was to evaluate whether addressing these point source discharges alone would enable the River Gaunless to meet WFD objectives.

Figure 3 compares total iron loadings (i.e. concentration multiplied by flow) and concentrations in the River Gaunless itself, with the cumulative iron load of the 6 point discharges of mine water. Under high flow conditions Figure 3A illustrates that there is a general trend of increasing iron concentration downstream. As expected, cumulative iron loading due to point discharges increases downstream also, with additional inputs of mine waters. However, the most striking feature of Figure 3A is the increasing iron loading within the river downstream of Lowlands, despite the negligible increase in cumulative mine water iron load from this point. At Bishop's Park, near the confluence with the River Wear, cumulative mine water iron load is 0.44 g/s, whilst total in-stream iron loading is 24.66 g/s. Therefore only

1.8% of the iron load of the River Gaunless at its outlet can be accounted for by point mine water discharges under high flow conditions. The sources of the additional iron loadings in high flow are likely to be 1) resuspension of ochre from the stream bed (and iron-rich bank sediments), particularly in the perennially ochre-stained reaches downstream of point sources, 2) spoil heap runoff, which may be limited to particular reaches where large exposed spoil heaps are found in close proximity to the river and 3) groundwater input directly to the river. The latter process would be expected to be of lesser significance in high flow than low flow as the flux of contaminated groundwater to the river is likely to be fairly consistent over time given the size of the groundwater bodies underlying the catchment.

There may also be considerable fluxes of iron from sources not connected with mining activity. Peat erosion in the upland parts of the catchment may be a significant contributor; a pattern clearly evidenced by elevated iron concentrations of 7.9mg/L at the headwater sample point upstream of Arn Gill mine water in low flow conditions (Figure 3B). Of greater significance for winter high flow loadings is iron derived from highways runoff (given that highly soluble sodium hexacyanoferrate(II) is used as an anti-caking agent in road deicing salts). Spot samples of road runoff during winter 2005 showed total iron concentrations to typically exceed 15mg/L. Such sources could therefore be responsible for a significant percentage of instream iron loading during winter high flow events in the more built-up lower catchment where numerous CSOs discharge surface drainage from the urban areas directly to the river. Quantification of these additional sources of iron at a catchment scale is however, problematic without the extensive deployment of auto-sampling equipment during high flow events.

In low flow conditions, the contribution of point sources is

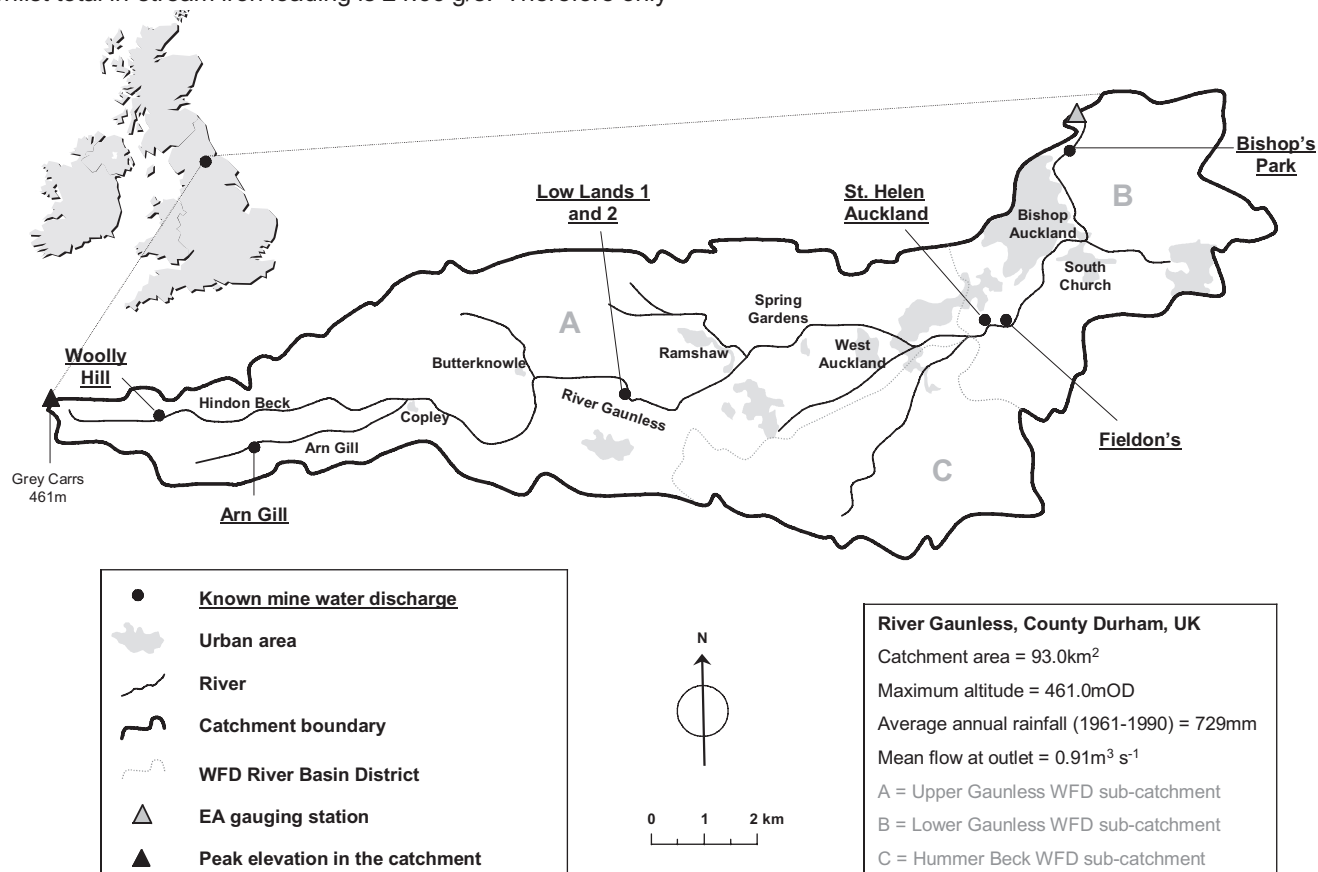


Figure 2: Schematic map of the Gaunless catchment highlighting known point mine water discharges, urban areas and Water Framework Directive management boundaries (after Mayes et al., 2005).

Table 1.: Summary statistics for dissolved and total iron ($\mu\text{g l}^{-1}$) in the River Gaunless, January 1990 - June 2005 (U/S = upstream; D/S = downstream; MW = mine water; DC = District Council). (Updated from Younger, 2000)

Site name	Grid ref. (Prefix NZ)	n =	Dissolved Fe			Total Fe		
			Mea	S.D.	Max	Mea	S.D.	Max
			n			n		
U/S Butterknowle	115250	40	138	107	409	425	891	5450
U/S Lowlands	133251	112	397	316	2520	886	1363	9700
D/S Lowlands	135250	120	818	395	1700	1356	423	2130
U/S Ramshaw	154260	100	258	178	1490	927	940	6680
Spring Gardens	173266	36	213	111	563	919	1829	10700
West Auckland	184267	150	187	134	630	-	-	-
Fieldon's Bridge	204266	46	158	91	423	907	1531	8520
U/S Fieldon's MW	205266	17	153	67	946	354	70	9520
D/S Fieldon's MW	206267	17	149	53	237	419	73	481
Wear Valley DC Depot	218285	33	199	139	676	1210	1497	7350
South Church	219284	101	201	139	722	1274	1407	8250
Cemetery (Bishop Auckland)	216292	47	174	97	417	1262	1960	12800
A689 (Bishop Auckland)	213300	37	177	119	700	981	1025	6330
U/S Bishop's Park	217302	7	907	2070	5600	1399	2352	7190
U/S Wear confluence	214306	99	170	121	593	978	927	7530

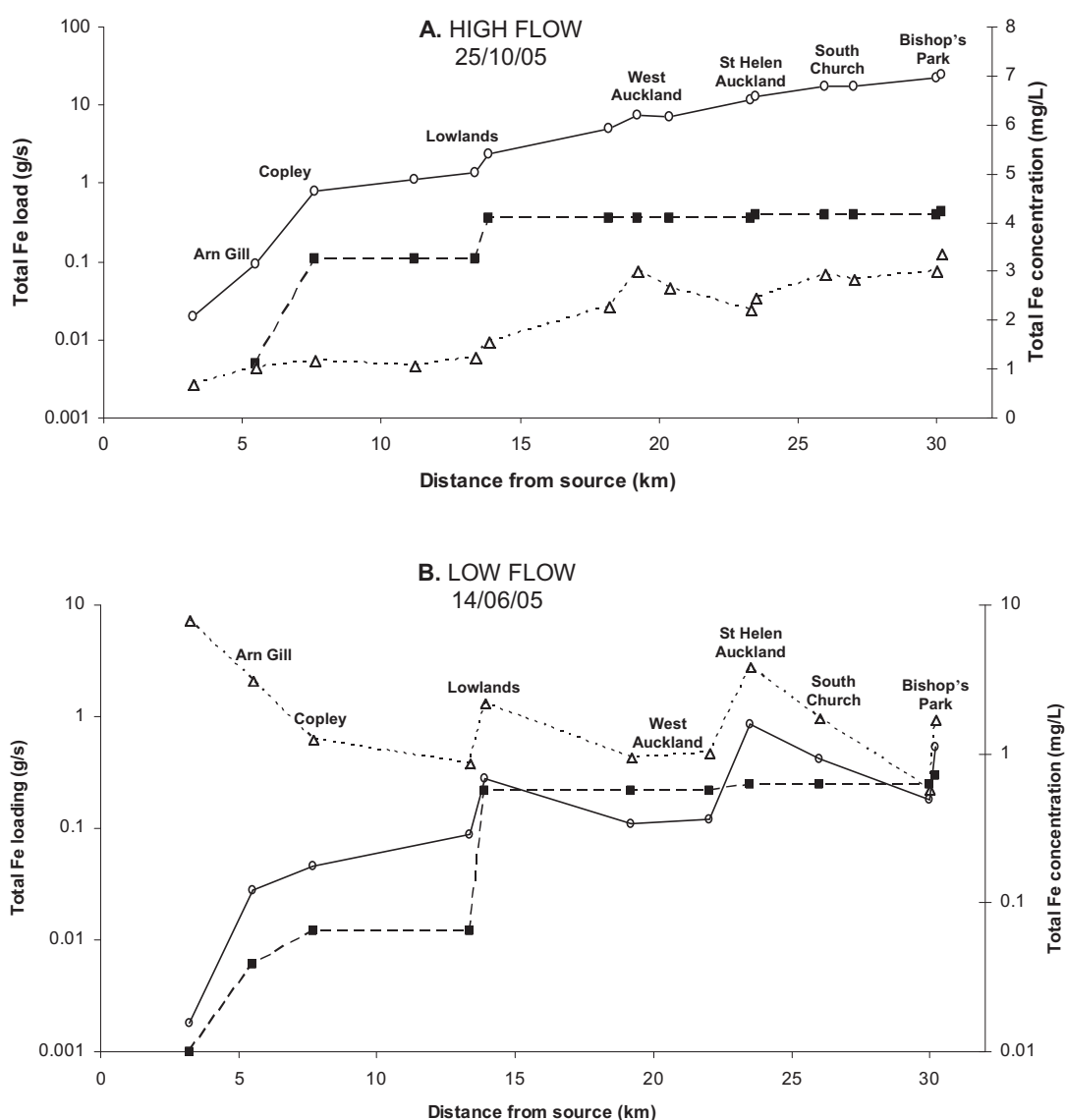


Figure 3: Total iron load and concentration profiles in the River Gaunless under high flow (Fig 3A) and low flow (Fig 3B) conditions (Closed squares = cumulative mine water total iron load; Open circles = total iron load in the river; Open triangles = total iron concentration in the river) (updated from Mayes et al., 2005).

much more apparent in the 'saw-toothed' iron profile (Figure 3B). Abrupt instream peaks in iron loading are apparent downstream of point mine water inputs (e.g. at Lowlands, Fieldon's and Bishop's Park). At Lowlands the point mine water accounts for over 100% of the instream iron load increase (suggesting immediate loss of ochre from the water column to the streambed), while at Fieldon's and Bishop's Park the point sources account for less than 60% of the instream increase in iron load. The latter suggests significant diffuse components in the immediate vicinity of the point sources. The peaks in instream load downstream of the mine waters are followed by subsequent decreases in the reach downstream of the measured rise. This again suggests that loss of iron as ochre on the stream bed is significant in attenuating iron loading in the water column in the reach 2-3km downstream of point sources, with iron load in the river returning to similar levels as those measured upstream of the mine water. At the most downstream sample point (Bishop's Park), the known point mine waters account for no more than 55% of the recorded instream loading toward the catchment outlet. This again clearly suggests major diffuse inputs of iron in the vicinity of Bishop's Park (equating to 21.1 kg day⁻¹ of Fe on 14/06/05), which presumably enter the river via diffuse seepage around the point source, or as direct groundwater discharge to the river via the hyporheic zone. Ongoing work in the catchment is investigating in more detail the groundwater-river interactions, particularly in the reach around St Helen Auckland where groundwater levels are very close to river levels.

The River Allen

Reconnaissance studies of point mine water sources have identified 40 point mine water sources, 13 of which were deemed significant for flow and hydrochemical monitoring (Figure 4) in this sampling programme (Gozzard *et al.*, 2006). Total zinc concentrations in these point sources range from 40 to 5420 µg/L, with lead concentrations ranging from 16 to 276 µg/L. Instream contaminant profiles under varying flow conditions have been established for the catchment and show some similar patterns to those in the Gaunless. Zinc loadings under high and low flow conditions are presented for the West Allen in Figure 5. The concentration curves highlight that throughout much of its course, particularly in low flow, much of the West Allen is in breach of the zinc Environmental Quality Standard (EQS) of 8?g/L. In high flow, a cumulative rise in instream zinc load is apparent for the upper 8km of the West Allen, which levels off for the lower 10km reach. The major point mine water at site 20 accounts for 60% of the instream rise in zinc loading in that reach, suggesting a significant diffuse component is associated with the point discharge. The total contribution of point sources to the instream zinc load at the catchment outlet (just upstream of the confluence with the East Allen) is just 10%. This indicates that the remaining 90% of instream zinc must arise from diffuse sources. These high flow sources are likely to be dominated by resuspended metal-rich sediments, although spoil heap runoff (particularly in the upper reaches) and direct discharge of contaminated groundwaters to the river via the hyporheic zone may also be significant.

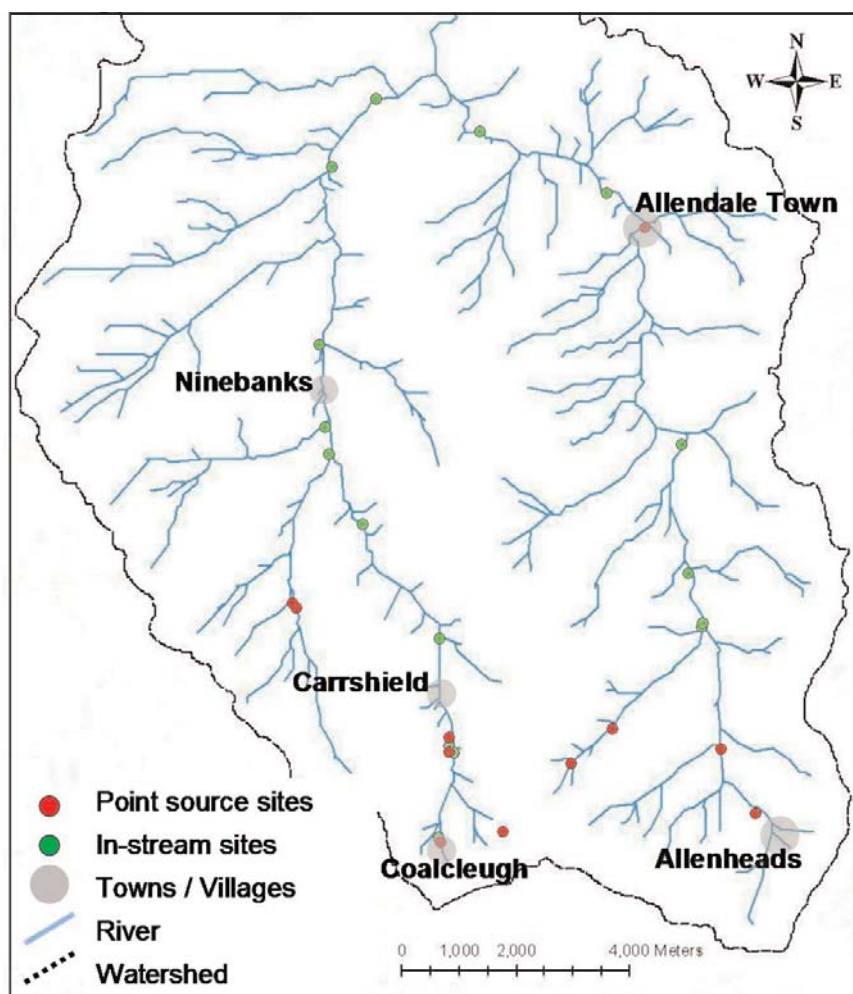


Figure 4.: Schematic map of the River Allen catchment highlighting known point mine water discharges, sampling locations and major settlements.

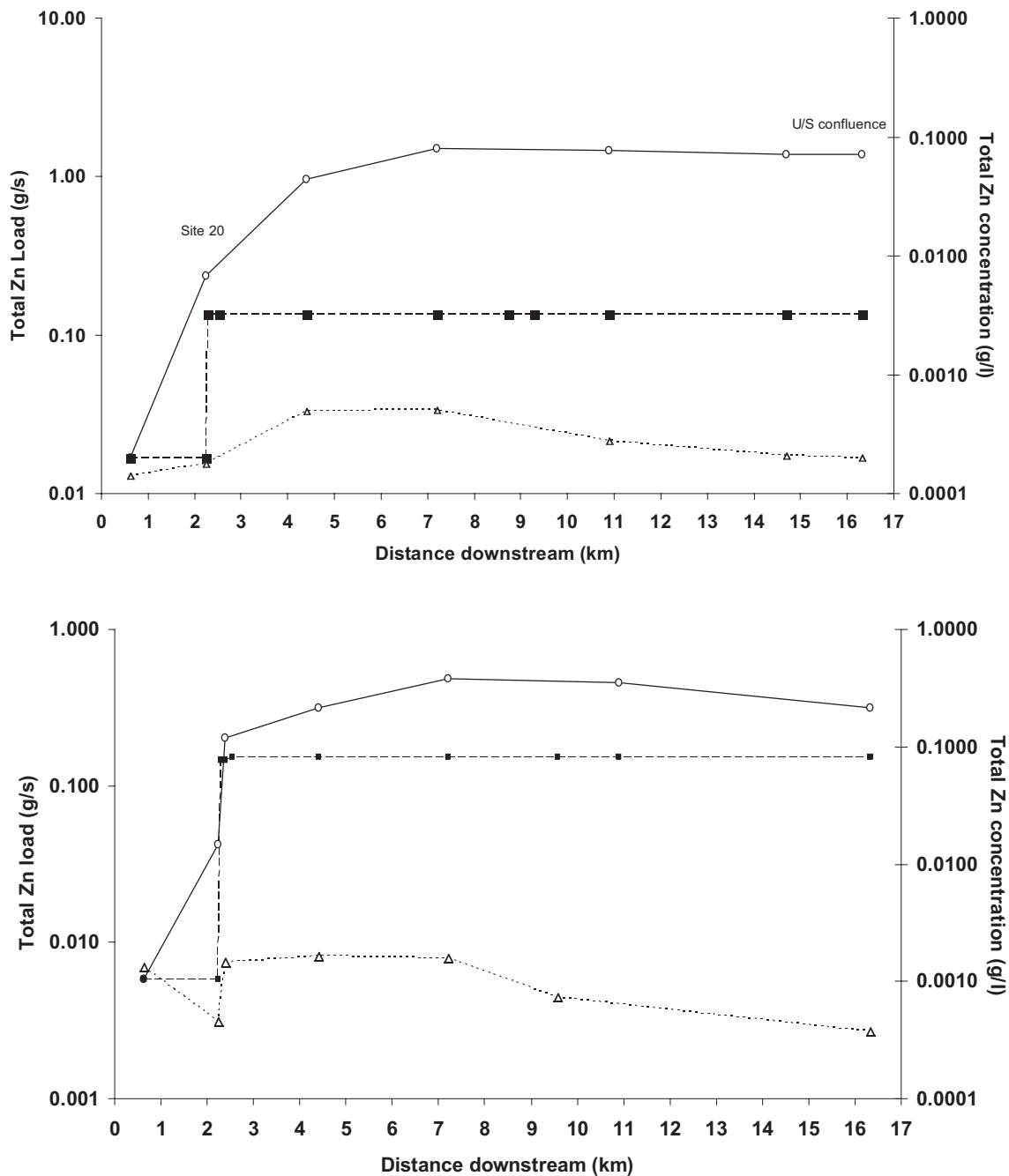


Figure 5. Total zinc load and concentration profiles in the River West Allen under high flow (Fig 5A) and low flow (Fig 5B) conditions (Closed squares = cumulative mine water total zinc load; Open circles = total zinc load in the river; Open triangles = total zinc concentration in the river).

In low flow conditions, the point source mine water contribution becomes a lot more significant to the instream zinc load. Site 20 accounts for 88% of the instream rise in zinc loading in the reach around the discharge, again suggesting a diffuse component to the mine water discharge to prevail under all flow conditions. At the monitoring point furthest downstream, the point mine water discharges account for 48% of the instream zinc load. This still suggests a significant diffuse component to low flow instream zinc loadings, which is likely to be dominated by the discharge of metal-rich groundwater directly to the stream via the hyporheic zone. Ongoing research in the catchment is assessing river-groundwater interactions and investigating the mobility and partitioning of metals on bed sediments

Management considerations

Approaches to mine water management at the catchment scale often focus towards identifying principal point sources

of pollutants detrimental to catchment water quality and ecology, and undertaking targeted remediation at the sites where limited funds will reap the maximum improvements to water quality (e.g. Kimball et al., 1999). Hypothetical estimates of residual loadings can be made for the Gaunless catchment if such a management approach was adopted. The three point sources of mine water at Lowlands 1, St Helen Auckland and Bishop's Park would be obvious candidates for remedial action as they produce the highest iron loads (note: the St Helen Auckland site already flows via a treatment wetland which is currently decommissioned). If target effluent total iron concentrations of 0.5 mg/L are assumed (a suitable estimate given the influent concentrations), the difference between point source contribution at present and under the hypothetical remediation scenario can be subtracted from the recorded instream loadings presented in Figure 3B. Figure 6 displays these residual total iron concentrations (Fig 6A) and loadings

(Fig 6B) based on the data collected on 14/06/05 under low flow conditions. Given that remedial action at point sources will be of greatest influence at low flow, such an exercise highlights the period at which potential remedial action would be at its most effective.

Figure 6A shows that instream total iron concentrations remain unchanged (and above 1.0 mg/L) for all the sample locations upstream of the first potential treatment site at Lowlands. Downstream of Lowlands the instream concentrations remain below 1.0 mg/L to Fieldon's Bridge. It is in this reach downstream of Lowlands where the most pronounced benefits of any remedial work would be seen and over time, with scouring of the perennial ochre deposits from the streambed, instream loadings may fall to negligible levels. In the reach around St Helen Auckland and Fieldon's Bridge however, remedial work would be less effective due to the large diffuse iron contribution from groundwater sources. Here, the predicted instream concentrations are 2.8 mg/L and remain close to 1.0 mg/L up to the catchment outlet. It is along this reach where complaints of cloudy water (caused by an iron-organic complex) have been repeatedly made by local residents (Mayes *et al.*, 2005). This brief exercise highlights that although there are significant localised improvements in iron loadings and concentrations under the remediation scenario, the diffuse sources in the catchment are likely to cause a continuation of high iron concentrations (to levels which may threaten WFD compliance), particularly

in the lower catchment during low flow conditions.

Wider management options for diffuse mine water pollution remain limited at present, with most 'proven' mine water treatment technologies being designed for point sources. Monitored natural attenuation (MNA) is a remedial paradigm that has been developed principally for applications with polluted aquifers. However, it may also have potential applicability to diffuse mine water pollution in the surface water environment. MNA relies on natural processes (e.g. attenuation of contaminants on the stream bed or in the hyporheic zone) to achieve the best overall environmental result. It also recognises that many active interventions have environmental costs of their own (e.g. quarrying limestone in an area of high amenity value to treat mine water elsewhere). For MNA to be fully justified, it needs to be based on well-designed monitoring and modelling coupled to rigorous economic analyses (e.g. Younger *et al.* 2005a). Of course, in the process of attempting to 'justify' MNA in this manner, it may well emerge that treatment of one or more point sources in a catchment is worthwhile after all (cf Younger *et al.* 2005a), as may well be the case in the Gaunless and Allen catchments.

In some cases, it may be possible to use established technologies (e.g. permeable reactive barriers at the toe of heavily polluting spoil heaps) to directly intercept and remediate diffuse mine water pollution sources (see, for

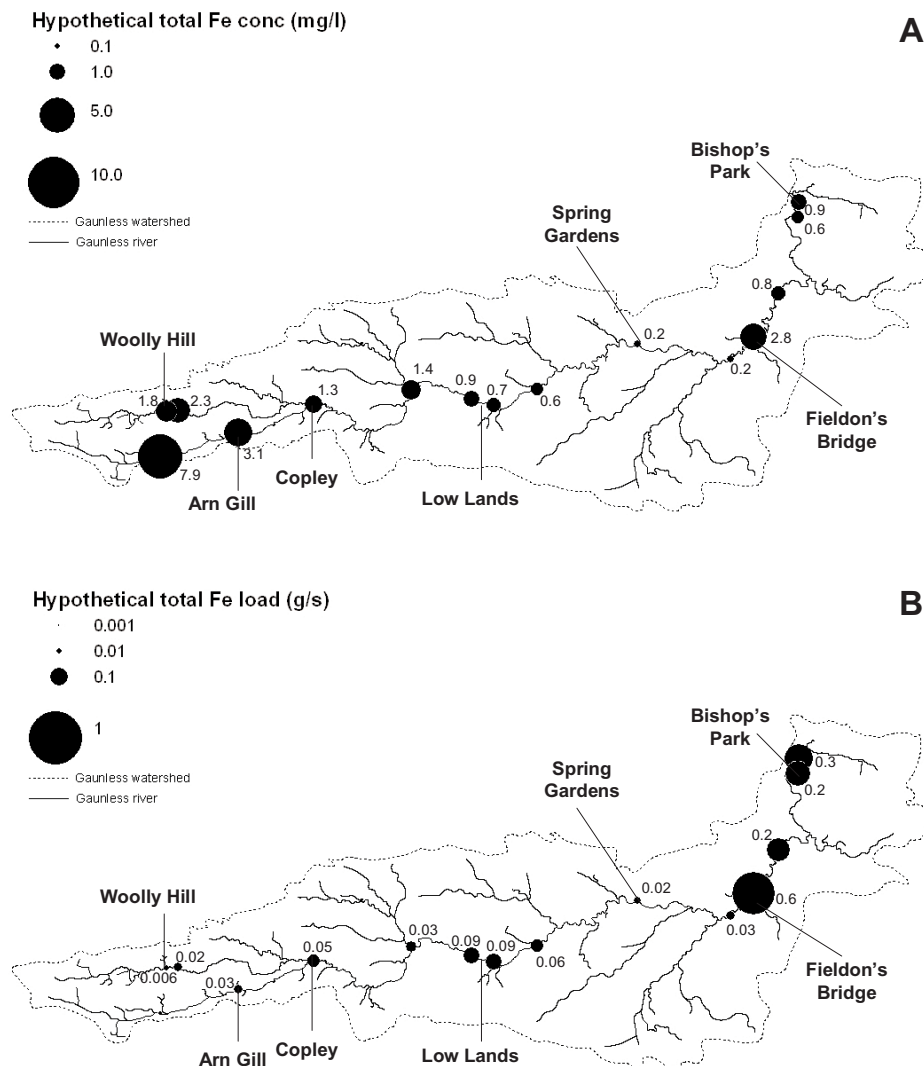


Figure 6: Hypothetical iron load and concentration under a point source remediation scenario and low flow conditions in the Gaunless catchment (after Mayes *et al.*, 2005).

example, Younger et al., 2005b). Other technological interventions can also be imagined, such as near channel, sub-surface dosing systems and gradient control pumping to minimise groundwater impacts during periods of low flow. Before substantial progress is likely to be made in the development of new technologies along these lines, a more profound understanding of diffuse pollutant release processes will need to be developed. Intensive catchment-scale sampling programmes such as those described here provide valuable estimates on the nature and extent of the diffuse mine water pollution problem. However, further monitoring would still be desirable to partition and quantify the flux of specific diffuse sources over time. This could include more detailed reach-scale monitoring, for example at channel reaches draining important spoil heaps or reaches of bedrock-controlled channel where groundwater-surface water interaction is clearly evident.

The outcomes of the ongoing investigations in the Gaunless and Allen catchments aim to improve this understanding of diffuse mine water processes through providing more robust estimates of the contribution and partitioning of diffuse sources under varying flow conditions. In addition, sedimentological and geochemical studies in the Allen catchment aim to yield important information about the mobility and long-term fate of metal-laden sediments in the catchment. Elsewhere in the UK, studies of hyporheic zone processes in mine-water impacted rivers in England and Wales are currently underway which will also go some way to improving the basis for management of diffuse sources.

CONCLUSIONS

The data collected to date in the Gaunless and Allen catchments have highlighted the significant contribution that diffuse sources make to instream metal loadings in both low (55% and 48% in the Gaunless and Allen respectively) and high flow (98% and 90% respectively). In low flow conditions, this diffuse input appears to be dominated by direct groundwater discharge into the streambed and seepage around point sources. At higher flows, the remobilisation of ochre or metals from bed sediments appears to be the major contributor to instream metal loadings in addition to spoil heap runoff.

Projected low flow loadings under a hypothetical remediation scenario for the three main point sources in the Gaunless catchment suggests clear localised improvements in iron loadings and concentrations, but the diffuse sources will continue to keep much of the catchment uncertain of compliance with WFD water quality objectives.

Wider planning for remediation of diffuse sources requires a greater appreciation of the modes of pollutant release in mined catchments. In particular, the partitioning between groundwater outflows and surface runoff, especially where complex hyporheic zone cycling of contaminants is feasible, will require substantial further study in many catchments. In addition, it will often be important to establish whether diffuse pollutants are truly 'new' to the river channel (e.g. by ferrous iron entry through groundwater upflow through the streambed) or remobilised pollutants that were previously present in the bed sediments. Ongoing research in the Gaunless and Allen catchments will explore some of these hydrogeochemical issues under varying flow conditions. Even after such issues have been resolved, it will still be necessary to undertake rigorous economic analysis (using the approaches outlined by ERMITE Consortium, 2003) if rational, defensible remediation of diffuse sources is to be

pursued.

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