Assessment of the Chemical and Ecological Recovery of the Frongoch Stream Following Remediation at Frongoch Lead and Zinc Mine, Mid Wales

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Abstract

Diversion of Frongoch Stream in 2011 reduced inflows to Frongoch Mine, increasing streamflow and diluting contaminants. This, together with subsequent surface water management, capping, hydroseeding and revegetation from 2013-2018, led to overall decreases of 87%, 93% and 87% for dissolved Zn, Pb and Cd respectively. Residual discharges, however, still cause the stream to fail to comply with Water Framework Directive standards and there is only modest evidence of biological recovery to date. Sediment metal concentrations in 2020 indicate that ecological recovery may be impaired by enduring bed-sediment contamination, even where erosion and sediment transport of mine waste is successfully managed.

Keywords: Metal Mine Remediation, Sediment Contamination, MetTol, Macroinvertebrates, Water Quality

Introduction

Treatment of coal mine discharges has protected and improved over 350 km of rivers in the UK in recent decades. Ecological recovery downstream of treated coal mine discharges can be rapid, due to natural washing of ochre from riverbeds (Wiseman *et al*. 2002). In contrast, metal mines remain a major cause of failure to achieve Water Framework Directive (WFD) standards with 1,300 mines adversely affecting over 700 km of rivers in Wales alone. Due to technical challenges and funding limitations, relatively few full-scale remediation schemes have been completed for UK metal mines and, to date, there is limited evidence of ecological recovery following these schemes.

Frongoch Mine, in the upper Ystwyth catchment in Mid Wales, UK (fig.1), was one of the most productive metal mines in Wales, producing over 100,000 tonnes of lead and zinc ore from 1798 until its closure in 1904 (Bick 1996). Large waste dumps were reprocessed to extract metals between 1917 and 1930, but these remained a source of metals pollution to Frongoch Stream thereafter. The mine is in the Silurian, Devil's Bridge Formation, largely composed of interbedded sandstone and mudstone. Annual rainfall is ≈2,000 mm and the surrounding land is mainly rough pasture/moorland. Prior to remediation, pollutant pathways to Frongoch Stream included surface runoff and shallow groundwater discharges, including a discharge from a culvert of unknown origin. Deeper groundwater from the mine drains via Frongoch Adit to the Nant Cwmnewydion in a neighbouring valley (Edwards *et al*. 2016). Frongoch ranked as Wales' second most polluting mine (Mullinger 2004), contributing to failures to achieve WFD standards for Zn, Pb and Cd in Frongoch Stream, Nant Cell, Nant Cwmnewydion, Nant Magwr and Afon Ystwyth (Stokes 2012). Annual metal discharges to Frongoch Stream were 6.5 and 0.5 tonnes of dissolved Zn and Pb respectively, while Frongoch Adit discharged approximately twice those amounts (Edwards *et al.* 2016).

Remediation at Frongoch Mine was completed in four phases from 2011 to 2018. In February 2011, Environment Agency Wales (EAW) stopped Frongoch Stream overflowing from Mill Pond into an open stope at the northeast of the mine by diverting it back into its original watercourse via a culvert from the pond. This reduced the flow to Frongoch Adit and increased dilution of metals in Frongoch Stream downstream of the mine. From January to May 2013, EAW (Natural Resources Wales (NRW) from April 2013) constructed a drainage channel around the northern and western perimeter of the site to intercept clean surface water and divert it away from the mine waste. A flood attenuation pond was built to receive this channeled water and discharge it immediately upstream of the old culvert discharge. From January to June 2015, NRW relocated mine waste to the north of the site where it was re-profiled to enhance runoff. A series of ponds was created to convey the drainage to the flood attenuation pond and provide a habitat for wildlife. A continuous section of imperforated pipe was placed in the perimeter channel to carry relatively clean water through an area of lead-rich tailings lagoon deposits highlighted by a geochemical assessment (Mustard 2013). The ponds and drainage channels were lined with geosynthetic clay liner (GCL) and most of the reprofiled area was covered with a minimum of 300 mm of compacted clay plus 100 mm of restoration soils, seeded with common bent grass. In total, over 23,000 m^2 of the site was capped in 2015, covering approximately 65% of the contaminated mine waste. From May to July 2018, 7,600 m^2 of the previously reprofiled but uncapped area was covered with GCL under a minimum of 350 mm confining layer and restoration soils. In September 2018, a further 5,000 m^2 was sprayed with experimental mixtures including commercial hydroseeding products with biochar, mycorrhizae and plant nutrients.

Monitoring methods

Monthly water quality spot sampling was carried out on surface and groundwater discharges and their receiving watercourses from 2011 to 2020. Sampling stations relevant to this study are shown in fig.1. Water quality data were also available for these sites from investigations prior to 2011, but there were gaps in the time series, notably in 2010. Water quality upstream of the discharges to Frongoch Stream was fully compliant with WFD standards, so monitoring stopped here in 2014. Spot samples of water were collected

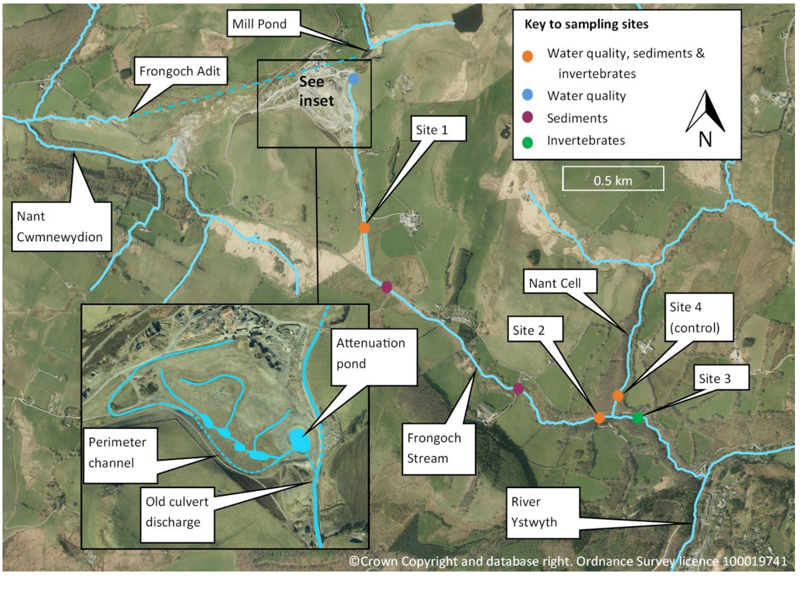

Figure 1 Map of sampling sites and mine features in Nant Cell and Nant Cwmnewydion catchments.

using standard NRW protocols and analysed in NRW's laboratory. Simultaneous flow gauging was carried out on the mine discharges and at Site 1, which was gauged by using a hand-held flow meter to measure velocity and multiplying this by cross-sectional area.

Samples of the macroinvertebrate community were collected from four sites on Frongoch Stream and Nant Cell on three occasions: August 2009, October 2019 and August 2020. These included three sites successively downstream of Frongoch Mine and a control site on Nant Cell (fig.1). All samples were collected using standard NRW protocols and the number of individuals of each taxa captured were calculated for each sample. A newly-developed biotic index to metal pollution (MetTol; Jones *et al.* 2016) was applied to the macroinvertebrate community data to assess any reduction in stress after the remediation. MetTol assigns a score (0-100) to each taxon present in a sample based on its tolerance to metal pollution. The final index value is the average of the scores for the sample, giving a measure of the average metal tolerance of taxa found at the site. MetTol index values typically vary between 25-55, with higher MetTol index values indicating a community under less metal stress. The 90th percentile of assigned MetTol scores was also calculated for each sample to provide a more focussed indication of the prevalence of metal-sensitive taxa. This latter approach is particularly appropriate when seeking evidence of recolonisation of stream sites by metal-sensitive taxa following management interventions.

Bed-sediment metals data for Frongoch Stream and Nant Cell (fig.1) were available from three M.Sc. studies linked to the remediation programme (Morgan 2013, Foggin 2016 and Lort 2017) and from NRW sampling in 2020 using standard NRW protocols for sample collection and analysis. Unfortunately, these surveys used disparate sample collection and analysis methods, which complicates comparisons of sediment metal concentrations over time. In particular, different size fractions of the fine sediment were considered by different surveys and information on the relative weight or volume of different fractions in each sample was not always available.

Results

Diversion of Frongoch Stream in 2011 reduced inflows to underground mine workings, increasing streamflow at Site 1 by ≈300%, providing more dilution of runoff and groundwater discharges from the southeast corner of the mine site. Compared with 2007-2009 data, dissolved Zn, Pb and Cd at Site 1 all reduced by >80% in the period from February 2011 to December 2012 (fig.2).

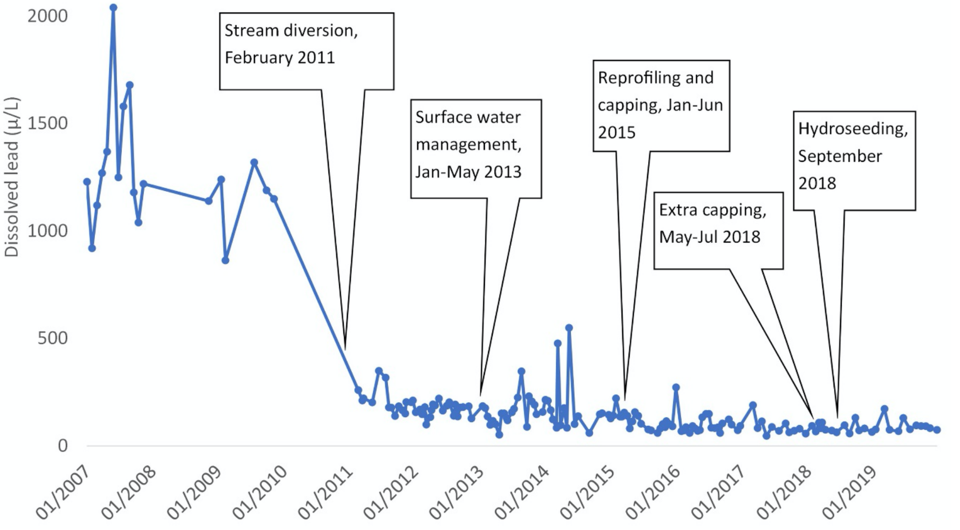

***Figure 2** Dissolved lead concentrations at Site 1 on Frongoch Stream.*

Frongoch Adit flow reduced by ≈80% following the diversion and although metal concentrations in the residual flow increased, the Zn, Pb and Cd loads reduced by 44%, 63% and 58% respectively. Reprofiling works in 2013 may have caused spikes in concentrations of metals in Frongoch Stream in 2013-14, but there was an overall reduction in metal concentrations over the period from March 2011, after the stream diversion, to January 2020, when monitoring stopped due to Covid-19 restrictions. A non-parametric Mann-Kendall test combined with a Theil-Sen estimator detected significant ($P<0.05$) downward trends over this period for Zn, Cd and especially Pb. There were, however, no obvious step reductions following the remediation phases after March 2011.

Based on a comparison of mean data from 2007-2009 and 2017-19, the combined remediation measures reduced dissolved Zn, Pb and Cd concentrations in Frongoch Stream by 87%, 93% and 87% respectively. Residual sources of these metals, however, still cause the stream to fail to comply with WFD standards. Bioavailable fractions of Zn and Pb at Site 1 in 2017-19 were 160 and 74 times higher than WFD standards respectively, while dissolved Cd was 63 times higher than the WFD standard based on hardness band. Zn, Pb and Cd concentrations were only ≈25% lower at Site 2 than at Site 1, so they also greatly exceeded WFD standards at this site. The Nant Cell control site (Site 4) is compliant with WFD standards for Zn and Cd, but marginally fails the standard for Pb, based on 2017-19 data.

The number of macroinvertebrate individuals and taxa captured in samples from sites 1-3 increased substantially between summer 2009 and autumn 2019, either side of the remediation, but a similar pattern was seen in the Nant Cell control site (tab. 1). Further sampling in summer 2020 found a consistent decrease in the number of individuals and taxa captured relative to the previous autumn, suggesting that seasonal and inter-annual variation may be important confounding factors to be considered when interpreting the data. Applying the MetTol index to the community data enables better identification of the unconfounded metal stress signal over the three sampling occasions. At sites 2 and 3 there have been distinct improvements with the macroinvertebrate community beginning to include more metal-sensitive taxa, such as the caddis flies *Silo pallipes*, *Sericostoma personatum*, the mayfly *Ecdyonurus*, and the stonefly *Perla bipunctata*. Considering 90%ile MetTol, there appears to be a sustained increasing trend at both these sites. At Site 1, just below the mine, there is less conclusive evidence for sustained improvements; MetTol values have decreased slightly but 90%ile MetTol has increased over the sampling period suggesting the first tentative steps

***Table 1** Community indices calculated for sites in Frongoch Stream/Nant Cell catchment.*

Site name	Date	Number of individuals	Taxon richness	MetTol	90%ile MetTol
Site 1	Aug 2009	20	8	43	52
	Oct 2019	753	25	38	50
	Aug 2020	308	23	36	57
Site 2	Aug 2009	61	11	29	40
	Oct 2019	645	28	36	50
	Aug 2020	240	14	35	54
Site 3	Aug 2009	244	20	31	41
	Oct 2019	579	34	40	52
	Aug 2020	107	24	38	58
Site 4	Aug 2009	53	15	38	53
(control)	Oct 2019	247	28	41	53
	Aug 2020	81	23	41	50

towards recovery may be occurring. The metal-sensitive taxa *S. pallipes*, *S. personatum*, and the non-biting midge *Chironomini* have been recorded intermittently at the site. At the Nant Cell control site, MetTol (38-41) and 90%ile MetTol (50-53) values varied less between surveys, but were not consistently greater than at sites 1 and 2 in the impacted Frongoch Stream.

Concentrations of Zn, Cd and Pb in bed-sediment at all sites on Frongoch Stream sampled in 2020 continue to far exceed the thresholds recommended by Jones *et al.* (2016) for fine sediments (<2000 µm) based on species sensitivity distributions derived from ecological field data (tab. 2). In particular, Pb concentrations are still over 50-100 times higher than the threshold values. Concentrations at the control site on Nant Cell in 2020 were below these thresholds. A time-series of consistently analysed sediment data from sites 1-4 is not available, but the data collected do not suggest a consistent reduction in sediment metal concentrations in Frongoch Stream over the 2013-2020 period.

Conclusions

Remediation at Frongoch Mine has controlled the release of contaminated mine waste and reduced metal concentrations in Frongoch Stream, but only modest biological recovery has been observed to date. Continuing impacts on the macroinvertebrate fauna can be attributed to enduring high concentrations of Zn, Cd and Pb in both the water column and streambed sediments.

Analysis of water quality and flow data for sampling sites at Frongoch Mine by Morgan (2020) concluded that the old culvert discharge is the largest remaining source of metals to Frongoch Stream. Other remaining sources include a separate groundwater discharge, which enters Frongoch Stream via the flood attenuation pond, and surface runoff, most of which also discharges via the pond outfall. With the exception of the 2011 stream diversion, reductions in metal concentrations in the Frongoch Stream have occurred progressively rather than in step changes following remediation phases. Morgan (2020) found that capping in 2015 and 2018 improved the quality of surface runoff from the mine site, but effects on groundwater sources were less evident.

Metal concentrations have continued to far exceed WFD standards even during periods where there was no visible discharge to Frongoch Stream via the old culvert or the flood attenuation pond, e.g. during the drought of summer 2018. This suggests that metal concentrations are augmented by diffuse groundwater inputs during periods of low flow, as observed by Byrne et al. (2020) in the neighbouring Nant Cwmnewydion

Table 2 *Sediment metal concentrations at sites in Frongoch Stream/Nant Cell catchment.*

Site name	Date	Zn mg/kg		Cd mg/kg		Pb mg/kg	
		<2000 µm	<63 µm	<2000 µm	<63 µm	<2000 µm	<63 µm
Site 1	2013 - 2017	Range 3034 - 36171*		Range 0.31 - 33*		Range 1318 - 12100*	
	Aug-20	9480	12700	48.1	34.1	1620	15300
	Sep-20	1660	5360	3.74	13.3	2970	14400
Site 2	2013 - 2017	Range 45 - 9156*		Range 0.77 - 11*		Range 2150 - 5221*	
	Aug-20	3900	9330	8.29	22.8	4530	8150
	Sep-20	2870	6930	6.79	20.3	2230	8450
Site 4 (Control)	2013 - 2017	Range 119 - 316*		Range 0.02 - 0.7*		Range 50 - 1100*	
	Aug-20	144	428	0.183	0.883	30	212
	Sep-20	136	298	0.185	0.664	32	222
Threshold concentration**		447	N/A	4.7	N/A	133	N/A

*Range includes variable size fractions <2000 µm, **Jones *et al.* (2016)

catchment. These diffuse sources may reduce the benefits of future treatment of the point source discharges.

The biological signal from sites on the Frongoch Stream suggests that the first tentative steps towards recovery are taking place, but the sediment data tempers hope that recovery will be rapid, even if groundwater sources of dissolved metals are treated. This is supported by Clements *et al*. (2010), who found a significant delay in macroinvertebrate recovery following stream restoration, partly due to residual sediment metal contamination. The macroinvertebrate community in Frongoch Stream will remain under stress for as long as the fine streambed sediment that many of the animals consume is contaminated to this extent. If the remediation has sufficiently reduced inputs of metal-laden water and sediment to the stream, much of the entrained and adsorbed metal should eventually be flushed downstream, allowing more metal-sensitive taxa in Nant Cell and the catchment upstream of Frongoch Mine to colonise and persist.

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