

A case study of long-term geochemical evolution of coal waste rock drainage and its remediation

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Abstract The paper discusses the geochemical evolution of drainage from an orphan waste rock pile which has been monitored for 15 years. Historically the drainage was acidic, with elevated iron and aluminium concentrations. The drainage from the 35 hectare waste rock heap, in northern England, was intercepted with a small compost wetland system from 1997, and the waste rock heap itself was selectively capped in 1998. Here we show the measurable improvement in water quality resulting from the capping of the heap, and the very modest costs of long-term treatment of such drainage.

Key Words remediation, waste rock, geochemistry, acidic

Introduction

A 35 hectare coal spoil (waste rock) heap has been a long-term source of pollution of a stream which runs through an area of high recreational value in north east England. The site is near the former mining village of Quaking Houses in County Durham, UK. The spoil material at the site was generated during coal mining at two adjacent collieries between 1922 and 1970 (Kemp and Griffiths 1999), and comprises grey and black shale, ash, coal and coal dust (Gandy and Younger 2003). The spoil heap has a depth of 4–10 m (Gandy and Younger 2003), and is founded on an impermeable clay layer. As a consequence a perched water table has developed over a depth of 6–9 m below ground level. It is this water that is the source of contamination of the local stream.

It was not possible to charge any previous mine owner with remediating the pollution arising from the spoil heap in this case. Therefore this is an orphan site, and the remediation effort described here was completed using charitable funding. Such orphan mine sites are a common occurrence across much of the UK and other parts of the world.

The remediation of the spoil heap comprised two phases:

1. Installation of an engineered compost wetland system in 1997, to intercept and treat drainage from the spoil heap
2. Selective capping of the spoil material, in 1998, to limit the level of pollution arising from the spoil heap

The compost wetland system and its performance is discussed in detail by Younger et al. (1997) and Jarvis and Younger (1999). The selective capping of the heap is discussed by Gandy and Younger (2003). Here we focus in more detail on the effect of the capping exercise on the quality of drainage from the heap some 10 years after the capping took place. We also briefly discuss how the combination of selective spoil capping and wetland treatment have served as a truly low cost, long-term, solution to problems of acid mine drainage from this particular abandoned mine site.

Methods

Water samples were collected in pre-washed polyethylene bottles. One aliquot of 125 mL was treated with laboratory grade concentrated hydrochloric acid, for subsequent cation analysis, whilst a second sample, without acid, was collected for anion analysis. All samples were stored at 4°C between collection and analysis. Cation analyses were performed mainly using a Varian Vista-MPX ICP-OES. Anion concentrations were determined principally using a Dionex DX320 ion chromatograph.

Periodic filtering (0.45µm) of samples was undertaken for Quality Assurance / Quality Control purposes, but filtering was not undertaken routinely for logistical reasons. Similarly, replicate samples were collected periodically rather than routinely to ensure acceptable QA/QC in accordance with APHA (1998). Results reported here are total concentrations unless otherwise stated.

Labile determinands were measured in the field at the time of sample collection. Measurements of temperature, pH, Eh, and electrical conductivity were made using a Myron 6P Ultrameter (temperature, pH, Eh and conductivity), calibrated prior to each site visit. A Hach digital titrator was used for alkalinity determination. It was used with 1.6N Sulphuric Acid and Bromcresol-Green Methyl-Red indicator powder, to give results in units of mg/L as CaCO₃.

Acidity concentration was calculated according to the method of Hedin et al. (1994), using the concentrations of Fe, Mn and Al, together with the pH. This approach has been shown to produce acidity concentrations closely representative of those obtained by titration (i.e. within $\pm 5\%$).

Flow-rate was measured using the 'bucket and stopwatch' approach, repeated three times on each occasion, with the mean of the closest two readings taken.

Results and discussion

Table 1 presents annual mean values for key contaminant variables for the period 1998 to 2004. The most notable trend in these data is the gradual decline in the calculated acidity concentration (the mean value for the first 6 months of 2005 was 15.1 mg/L as CaCO₃; $n=8$). Since the acidity is calculated from the concentrations of Fe, Al, Mn and the pH, clearly these decreases relate to reductions in metal ion concentrations, and / or increases in pH. Table 1 illustrates that Al concentration decreased markedly in 2001. The decrease in Al concentration (2.5 mg/L) between 2001 and 2002 is of sufficient magnitude to account for almost the entire drop in acidity concentration in the same period. Decreases in Mn and Fe concentrations between 2003 and 2004 explain the further drop in acidity concentration in this period.

Table 1 illustrates that there was a step change in Al concentration around the middle of 2001. The data in Table 1 suggests that step changes in acidity concentration have also occurred. However, Figure 1, which presents all values of acidity concentration for the period 1997 - 2005, illustrates that the overall attenuation of acidity has in fact been a gradual trend.

The decreasing trend in acidity concentration throughout the period of monitoring is clearly illustrated by Figure 1. What might be the reason for this gradual improvement in water quality? Evans et al. (2003) and Gandy (2005) have modelled, using a 1-D advective-reactive transport model and an object-oriented particle tracking model respectively, the evolution of pyrite oxidation in the spoil heap. Since there is a finite mass of pyrite within the heap, and oxidative dissolution of pyrite is ongoing, ultimately that portion of the pyrite that is susceptible to weathering must become exhausted, and pollution will therefore cease. However, both Evans et al. (2003) and

Table 1 Annual mean concentrations (in mg/L) of metal contaminants, acidity (mg/L as CaCO₃) and pH in the spoil heap leachate (S.D. = standard deviation; n = number of measurements)

		1998	1999	2000	2001	2002	2003	2004
pH	Mean	6.09	5.98	7.00	7.25	6.53	6.44	6.78
	S.D.	0.59	0.65	0.76	0.94	0.52	0.32	0.43
	n	44	32	21	13	22	22	15
Acidity _{calc.}	Mean	45.1	47.4	40.1	46.7	30.7	28.3	21.8
	S.D.	19.2	20.3	21.0	21.4	11.0	10.7	8.0
	n	42	31	14	6	22	22	15
Fe	Mean	4.1	6.6	4.4	4.2	5.1	4.6	1.7
	S.D.	3.13	4.47	2.67	2.82	3.10	3.16	1.19
	n	54	43	19	13	22	22	15
Mn	Mean	3.8	3.6	2.7	2.9	2.5	2.5	1.7
	S.D.	1.83	1.77	1.28	1.30	1.25	1.18	0.98
	n	54	43	17	13	22	22	15
Al	Mean	5.4	5.1	5.2	5.6	3.1	2.8	2.8
	S.D.	2.35	2.57	3.38	2.48	0.88	0.89	1.01
	n	50	43	19	6	22	22	15

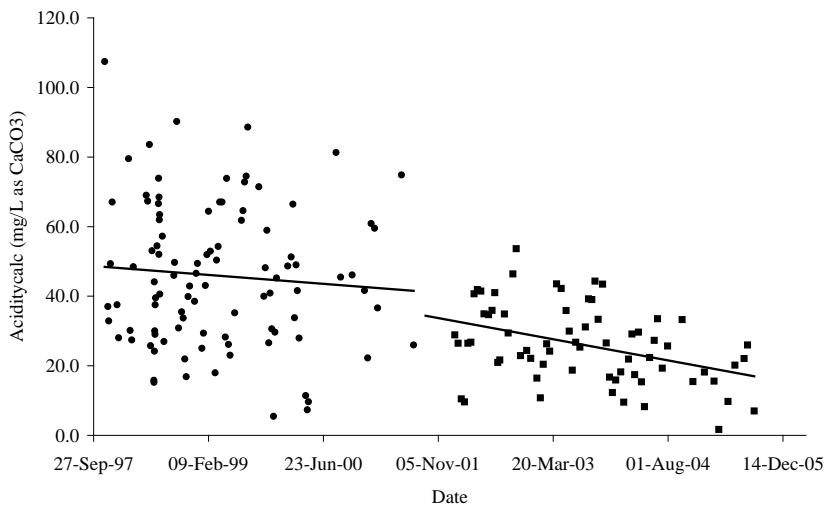


Figure 1 Changes in acidity concentration of the spoil heap leachate between November 1997 and July 2005; trendlines are for the periods November 1997 to July 2001 and August 2001 to August 2005 – see text for explanation

Gandy (2005) predict that the time-scales for such natural attenuation to occur are in the order of centuries. Since Table 1 and Figure 1 illustrate that pollutant concentrations have depleted by upwards of 50% within a decade, exhaustion of reactive pyrite seems unlikely to be the reason for the improvement in quality of the spoil leachate.

In part because of the longevity of spoil heap leachate predicted by workers such as Evans et al. (2003) and Gandy (2005), it is clearly desirable to endeavour to minimize contamination potential by limiting pollutant release at source. In 1997, in parallel to construction of the compost wetland, 3 boreholes were installed in the north-eastern part of the spoil heap. Analyses of water samples from these boreholes indicated that the severity of pollution within the heap was far from homogenous (Pritchard, 1997). 'Hotspots' of contamination, in areas of especially active pyrite weathering, were identified in certain areas of the heap. Within these areas Pritchard (1997) identified highly elevated concentrations of Fe (£ 276 mg/L), Al (£ 121 mg/L) and Mn (£ 94 mg/L) (cf. concentrations in the leachate shown in Table 1). In an effort to limit the potential for continued pollution from these areas selective capping of the spoil heap was undertaken in February 1998. Gandy and Younger (2003) reported the results of continued monitoring of water quality of the perched water table, and concluded that contaminant concentrations (Fe, Al and SO_4) decreased following completion of the cap. Furthermore, early results indicated that concentrations of key contaminants in the discharge to the local stream also decreased, some 3–4 months after the capping exercise was complete (Gandy and Younger, 2003). The 3–4 month lag time was comparable with the calculated travel time of water through the spoil heap to the discharge point, based on measurements of the hydraulic conductivity and porosity of the spoil material, and the hydraulic gradient and distance to the discharge point (Gandy and Younger, 2003).

Additional monitoring data for the spoil heap leachate since Gandy and Younger (2003), enables further important conclusions to be drawn regarding the influence of the clay cap on the spoil heap. Figure 1 illustrates that calculated acidity concentration steadily decreased following installation of the clay cap. However, the trendlines on Figure 1, which illustrate the trends in acidity concentration for the periods November 1997 to July 2001 and August 2001 to August 2005, show that the rate of decrease of acidity concentration has accelerated during the latter period. The timing of this more rapid attenuation of acidity corresponds to the decrease in Al concentration illustrated in Table 1. It appears to be principally the reduction in Al concentration that is responsible for the overall improvement in water quality.

Given that absolute exhaustion of pyrite available for weathering is unlikely in light of the conclusions drawn by Evans et al. (2003) and Gandy (2005), the selective clay capping of the spoil

heap appears to be responsible for the improved quality of the spoil heap leachate. Specifically, it seems that reduced infiltration is limiting the supply of oxygen (present in dissolved phase in the infiltrating water) for pyrite oxidation, and / or the selective cap is limiting oxygen diffusion into the pyritic heap. This in turn appears to be reducing the potential for leaching of Al from clay minerals and silicates, and therefore limiting the concentration of Al in the discharge to surface. The reduction in metal concentrations in the spoil heap leachate has in fact resulted in the discharge increasingly exhibiting net-alkaline (i.e. alkalinity > acidity), rather than net-acidic, characteristics.

Conclusions

The selective capping of a coal spoil heap in the northern England has resulted in significant improvements in leachate water quality. Modelling of pyrite weathering within the heap suggests that these improvements are a result of the capping, and not a consequence of a natural reduction in the extent and rate of pyrite weathering. The clearest improvements in water quality of the leachate relate to aluminium and acidity concentrations.

This paper has focused on the long-term evolution of the spoil leachate following selective capping with clay. However, as noted earlier, a compost wetland system has also played an important role in the remediation of the drainage. The wetland system was installed for a modest cost of around £60,000 (Jarvis and Younger 1999). In 2005 the wetland was completely renovated for a cost of approximately £50,000 (unpublished data), and the wetland continues to effectively remediate residual pollution from the spoil heap to this day. Thus, a combination of relatively low-cost selective capping of the heap, and passive treatment of the spoil drainage, has resulted in sustained water quality improvements in the stream below the spoil heap. It is hoped that this case study of low cost remediation of leachate from an orphan spoil heap may serve as a useful example to others involved in addressing pollution from such sites.

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