THE ADOPTION AND ADAPTATION OF PASSIVE TREATMENT TECHNOLOGIES FOR MINE WATERS IN THE UNITED KINGDOM

Paul L Younger

Department of Civil Engineering, University of Newcastle, Newcastle Upon Tyne NE1 7RU, United Kingdom E-mail: p.l.younger@ncl.ac.uk

Abstract

During the 1990s, passive treatment technology was introduced to the United Kingdom (UK) principally by imitation of US experiences. Early hesitancy on the part of regulators and practitioners was rapidly overcome, at least for the case of net-alkaline mine waters, so that at the start of the 21st Century, passive treatment is now the technology of choice for the long-term remediation of such discharges, wherever land availability is not unduly limiting. As confidence in the technology continues to grow, innovations are beginning to arise in response to problems peculiar to UK conditions, so that there is now a total of six types of passive system in use for mine water treatment:

- aerobic, surface flow wetlands (reed-beds);
- anaerobic, compost wetlands with significant surface flow;
- ◆ mixed compost / limestone systems, with predominantly subsurface flow (so-called <u>Reducing and Alkalinity Producing Systems (RAPS));</u>
- subsurface reactive barriers to treat acidic, metalliferous ground waters;
- closed-system limestone dissolution systems for zinc removal from alkaline waters;
- roughing filters for treating ferruginous mine waters where land availability is limited.

The first four are recognisably North American in origin, while the latter two are, as far as is known, unique to the UK at the time of writing. The limestone systems rely on removal of Zn as the carbonate (smithsonite), whereas the roughing filters remove iron by means of surface-catalysed oxidation of ferrous iron (SCOOFI), whereby ochre is used as a sorptive catalyst. Each of these technologies is appropriate for a different kind of mine water, or for specific hydraulic circumstances. The degree to which each type of system can be considered "proven technology" corresponds to the order in which they are listed above. Many of these passive systems have become foci for detailed scientific research, largely as a part of a \$1.5M European Commission project running from 2000 to 2003. Preliminary results suggest that proton consumption by reduction of ferric iron and precipitation of elemental sulphur may be an important cause of pH increases in compost wetlands, in addition to the more widely appreciated processes of sulphate reduction and limestone dissolution.

Key words: acidity, aerobic, anaerobic, compost, iron, metals, passive, reactive barrier, treatment, water, wetlands

INTRODUCTION

Widespread mine closures in the UK during the early 1990s led to extensive surface water pollution (Younger, 1993, 1998a; National River Authority, 1994). Public outcry prompted the

first steps towards a national strategy for dealing with the legacy of water pollution from recently- and long-abandoned mines. As regulators came to appreciate that mine water pollution often endures for decades (or even centuries) after mine abandonment (Younger, 1997a; Wood *et al.*, 1999), interest quickly grew in alternatives to conventional treatment technologies, favouring in particular solutions which operated with minimal maintenance (Younger, 1997b). The timely publication of the U.S. Bureau of Mines' recommendations on the passive treatment of coal mine drainage (Hedin *et al.*, 1994) proved serendipitous for the UK mining and water industries, and their design guidelines enjoyed rapid and enthusiastic uptake (Younger, 1995; Younger and Harbourne, 1995; Younger, 1997b). By the end of 1997, a total of 8 passive treatment systems were operational at UK mine sites (Table 1; Figure 1; see also Younger, 1997b). The uptake of the technology has continued apace since then, with 23 full-scale systems and a further 5 pilot systems in operation by the Spring of 2000. All but two of the 28 systems (numbers 23 and 28 in Table 1 and Figure 1) address abandoned mine discharges.

The objectives of this paper are:

- (i) To summarise the uptake and performance of these passive mine water treatment systems in the UK, which is the first European country to adopt the technology on such a large scale;
- (ii) To highlight recent innovations in passive treatment in the UK; and
- (iii)To outline recent and forthcoming European research aimed at fostering wider uptake of the technology.

CURRENT METHODS OF PASSIVE MINE WATER TREATMENT IN THE UK

Six types of passive systems are now in use for mine water treatment in the UK:

- (1) Aerobic, surface flow wetlands, which are often termed "reed beds" in the UK (e.g. Laine, 1997) (Figure 2a);
- (2) Anaerobic, compost wetlands with significant surface flow (e.g. Younger *et al.*, 1997; Edwards *et al.*, 1997; Younger, 1998b; Jarvis and Younger, 1999) (Figure 2b);
- (3) Mixed compost / limestone systems, with predominantly subsurface flow (Figure 2c). These systems were originally labelled "SAPS" (Successive Alkalinity Producing Systems) by their originators, Kepler and McCleary (1994). The label 'SAPS' remains popular in the UK (e.g. Younger, 1998b), though G. Watzlaf and coworkers (personal communication, 2000) have recently re-named such systems "Reducing and Alkalinity Producing Systems" (RAPS), which is arguably a more descriptive name for them, and is the acronym used hereafter and in Table 1;
- (4) Subsurface reactive barriers to treat acidic, metalliferous ground waters (Figure 3a);
- (5) Closed-system limestone dissolution systems for zinc removal from alkaline waters (Nuttall and Younger, 2000) (Figure 3b); and
- (6) Roughing filters for the aerobic treatment of net-alkaline ferruginous mine waters where limited land availability precludes a surface wetland.

The first three types of system have been widely documented in the international literature (see, for instance, Hedin *et al.*, 1994; Walton-Day, 1999), and the principles upon which they are based need no further rehearsal here. The fourth type of system, subsurface reactive barriers (Figure 3a), are no different in concept from the earlier RAPS and subsurface flow compost

Table 1. Passive treatment for mine waters in the UK - a summary of all existing systems. All concentrations are in mg/l (in the case of acidity and alkalinity, as $CaCO_3$ equivalent), save for pH. The mineral worked is coal except where stated in the second column.

(a) Systems in Cornwall and Wales

mine, siderite

orebody with pyritic roof

2000

Site #	tems in Corny Site Name	Date	Mine water	Passive system type	Total	Typical	Typical
Figure 1	(mining district)	Completed	type	Tubbive system eype	area (ha)	influent	effluent
1	Wheal Jane (Cornwall) Tin and zinc	Dec. 1994	Deep mine drainage	3 large pilot systems (aerobic wetlands followed by sealed anaerobic beds, and aerobic rock filters with 3 variations in pretreatmentnone, ALD or lime dosing) (Hamilton <i>et al.</i> , 1997)	2.4	Fe 141 Zn 79 Cu 0.4 Mn 24 As 2.7 pH 4.0	Fe 19 Zn 45 Cu 0.2 Mn 20 As 0.01 pH 3.6
2	Pelenna I (South Wales)	Oct. 1995	Drift mine drainage	Surface flow compost wetlands (Edwards <i>et al.</i> , 1997)	0.09	Fe 20 pH 6.5	Fe 5 pH 7.3
3	Pelenna II	Aug. 1999	Acidic drift mine drainage	Aerobic cell, RAPS, aerobic cell, RAPS, aerobic cell in series	0.75	Fe 35 pH 5.5	Fe 1.5 pH 6.5
4	Pelenna III	Apr. 1998	3 drift mine discharges	2 parallel streams of RAPS followed by aerobic wetland (Edwards <i>et al.</i> , 1997; Younger, 1998b)	1.0	Fe 69 Zn 0.034 Acid. 125 pH 5.0	Fe 4.2 Zn 0.004 Acid. 8.9 pH 7.2
5	Ynysarwed (South Wales)	March 2000	Net-acidic deep mine	Aerobic wetland polishing active treatment effluent until water quality allows use of wetland alone (Ranson and Edwards, 1997; Younger <i>et al.</i> , 1998)	1.0	Not yet available	Not yet available
6	Gwynfi (South Wales)	Aug. 1998	Net-alkaline drift mine	Aerobic wetland (owned by Coal Authority)	0.08	Fe 7	Fe < 1.0
b) Sys	tems in Leices	tershire, La	ncashire and	d Yorkshire (England)	I.	ı	1.
7, 8, 9	Nailstone (Leicestershire)	Sept 1997	Spoil heap drainage	Small surface flow compost wetlands and an aerobic wetland	0.25	Fe 46 Al 47 pH 4.9	Fe 14.6 Al 9 pH 5.6
10	Renishaw Park South Yorkshire	Nov. 1999	Spoil leachate	Subsurface reactive barrier (owned by Coal Authority)	0.004	Not yet available	Not yet available
11	Dodworth Pit Heap South Yorkshire	Sept. 1994	Spoil leachate	Aerobic wetland (Bannister, 1997)	0.25	Fe 30 pH 6.5	Fe < 1.0 pH 7.3
12	Woolley Colliery West Yorkshire	July 1995	Alkaline pumped deep mine water	Aerobic wetland receiving effluent from Coal Authority active treatment plant, which lowers Fe from 40 to 10 (Laine, 1997, 1998).	1.4	Fe 10	Fe < 1
13	Old Meadows Drift (East Lancashire)	Sept. 1999	Net-acidic drift mine discharge	Aerobic wetland polishing receiving effluent from Coal Authority active treatment plant, which lowers Fe from 50 to 5). (Laine, 1998)	0.16	Fe 5	Fe < 1
14	Skinningrove (Cleveland, N Yorkshire) Old ironstone	Pilot: Nov. 1999; full scale: Summer	Net-alkaline, deep mine shaft overflow	Roughing filter to remove Fe from water, located below ground in a steep-sided, urbanised valley, where	0.25	Not yet available	Not yet available

wetlands not feasible

Table 1 (continued)

(c) Systems in Durham, Cumbria and Northumberland (England)

Site #	Site Name	Date	Mine water	Passive system type	Total	Typical	Typical
Figure 1	(mining district)	Completed	type		area (ha)	influent	effluent
15	St Helen Auckland (South Durham)	Aug. 1999	Mine shaft overflow	Aerobic wetland (English Partnerships).	0.25	Fe 3 Alk. 500 pH 6.3	Fe 0.3 Alk. 480 pH 6.7
16	Bowden Close (West Durham)	Sept. 1999	Spoil leachate / drift mine drainage?	Pilot-scale RAPS followed by small aerobic wetland (Durham County Council).	0.1	Fe 20 Al 8 pH 5.0 Alk. 4	Fe 3 Al 0.1 pH 7.2 Alk. 225
17	Nenthead (Cumbria) Abandoned Pb- Zn mine	July 1998	Zinc-rich, alkaline mine water	Pilot closed-system limestone dissolution reactor (Nuttall and Younger, 2000)	0.0007	Zn 8	Zn 3
18	Tailrace Level (Durham) Recently abandoned Pb- Zn-F mine	Feb 2000	Zinc-rich, alkaline mine water	Closed-system limestone dissolution reactor (Environment Agency)	0.25	Zn 40	Not yet available
19	Edmondsley Yard Drift (Central Durham)	Sept. 1999	Net-alkaline drift mine water	Aerobic wetlands (Coal Authority).	1.25	Fe 27	Fe 0.1
20	Quaking Houses (Northwest Durham)	Pilot: Feb 95 Full-scale: Sept 97	Acidic spoil heap leachate	Surface flow compost wetland. (Younger <i>et al.</i> , 1997; Jarvis and Younger, 1999; Kemp and Griffiths, 1999)	0.04	Fe 10 Al 53 pH 4.5	Fe 1 Al <0.5 pH 6.5
21	Oatlands (West Cumbria)	Sept. 1998	Acidic spoil heap leachate	Aerobic wetland (see Warner, 1997)	0.16	Fe 85 pH 4.0	Fe 25 pH 3.1
22	Shilbottle Northumberland	1995	Acidic spoil heap leachate	Aerobic wetland (reactive barrier under construction)	0.3	Fe 100 pH 3.5	Fe 25 pH 3.0
(d) Sys	tems in Scotla	nd				_	
23	Craigenbay (Galloway, Scotland) Aggregates with low-grade mineralisation	August 1998	Open pit and stockpile drainage	4 aerobic cells, 2 with RAPS pre-treatment (Norton <i>et al.</i> , 1998)	0.14	pH 3.5	pH 5.5
24	Dalquharran (Ayrshire, Scotland)	September 1994	Deep mine drift drainage	Aerobic wetland (Marsden <i>et al.</i> , 1997)	0.095	Fe 200	Fe 50
25	Monktonhall (East Lothian, Scotland)	July 1998	Deep mine pumped effluent	Aerobic wetland polishing effluent from active treatment plant	0.5	Fe 10	Fe < 1
26	Minto Colliery (Ore Valley, Fife)	April 1998	Deep mine shaft overflow	Aerobic wetland (Coal Authority)	1	Fe 18	Fe < 1
27	Mains of Blairingone (Clakmannan coalfield)	November 1995	Old adit draining back-filled opencast	Aerobic wetland (Marsden et al., 1997; Younger, 2000a)	6.2	Fe 38	Fe 2.3
28	Foss Mine (Aberfeldy) Barite in pyritic and sphaleritic orebody	August 1998	Active mine workings pumped drainage	Pilot scale system comprising RAPS followed by aerobic wetlands, then closed-system limestone bed	0.005	Fe 2 Mn 14 Al 8 Zn 32 pH 5.4	Fe 0.4 Mn 12 Al 1.5 Zn 25 pH 5.8

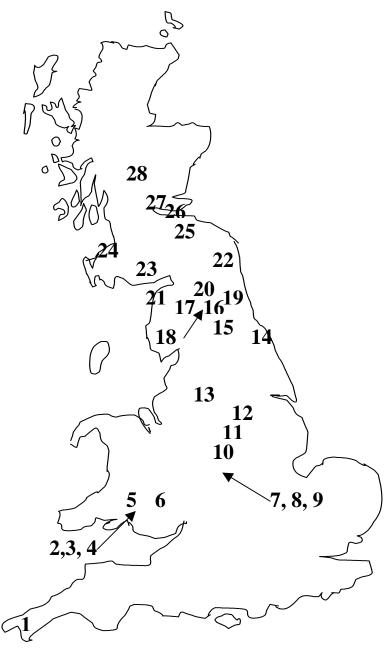


Figure 1. Locations of passive mine water treatment systems in the UK as at 30th April 2000. The identities of the numbered locations are given in Table 1. The systems marked here include six "pilot" systems (sites 1, 14, 16, 17, 22 and 28), and four sites at which passive treatment forms only part of an overall treatment system which includes "active treatment" unit processes (sites 5, 12, 13 and 25).

wetlands, save that they are employed in situations where it is possible to treat groundwater *in situ*, rather than waiting for it to discharge at the land surface as a mine water outflow. The first application of a subsurface reactive barrier to treat groundwaters polluted by mining activities was at the Nickel Rim site in Canada (Benner *et al.*, 1997). Similar systems are now being installed in Europe. The first was installed at Renishaw Park, UK, in 1999 (site 10, Table 1 and

Figure 1), and a second is scheduled for construction during 2000 at Shilbottle, Northumberland, UK (site 22 in Table 1 and figure 1). Probably the largest barrier of this type in the world is currently under construction at Aznalcóllar, Spain (C. Ayora, CSIC, Barcelona, *personal communication*, 2000).

The fifth and sixth types of passive system listed above are, as far as is known, unique to the UK at the time of writing. They are therefore discussed in further detail later in this article.

(a) Surface flow aerobic reed bed, for net-alkaline, ferruginous mine waters

(b) Surface flow compost wetland, for acidic mine waters where head is limiting

(c) <u>Reducing</u> and <u>Alkalinity Producing</u> <u>Systems (RAPS) for acidic mine waters where there is sufficient head</u>

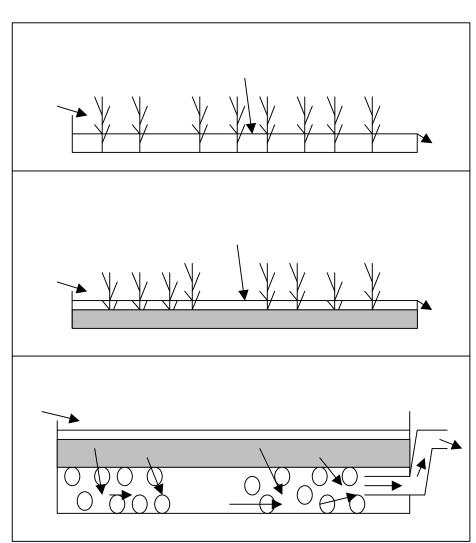
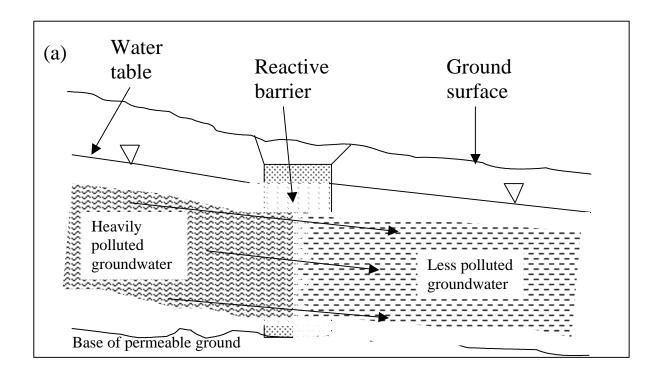


Figure 2. Schematic illustrations of the three most common types of passive treatment system used for polluted mine waters in the UK.

Each of the above technologies is appropriate for a different kind of mine water, or for specific hydraulic circumstances. The degree to which each type of system can be considered "proven technology" corresponds to the order in which they are listed above. This ranking of confidence is reflected in their relative abundance in the UK. In April, 2000, there are twelve full-scale



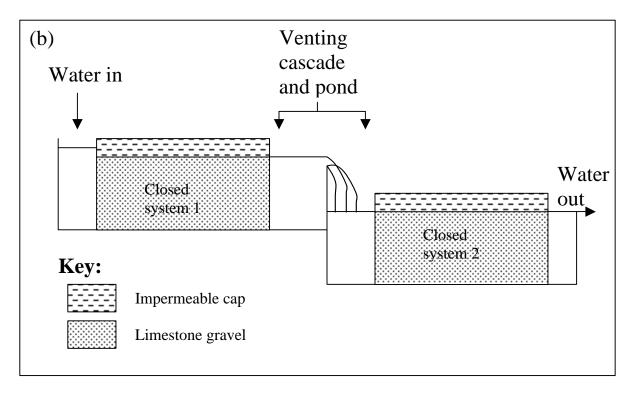


Figure 3. Schematic drawings of two less common types of passive treatment system: (a) subsurface reactive barrier for acidic, metalliferous ground waters found at mine sites, and (b) successive closed-system carbonate dissolution units for subsurface treatment of alkaline, zincrich mine waters.

reed-bed systems (of which seven are operated by the Coal Authority, a national government agency), three RAPS, three anaerobic reed-beds, one reactive subsurface barrier, one full-scale closed-system limestone dissolution unit for zinc removal (with another at pilot scale) and one roughing filter.

RECENT INNOVATIONS IN PASSIVE MINE WATER TREATMENT IN THE UK

Optimising Ochre Accretion with Roughing Filters

The population density of the UK is about ten times greater than that of the USA. This simple demographic fact results in passive treatment frequently being far more difficult (and/or expensive) to implement in the UK. Indeed the UK's leading developer of passive treatment systems, the Coal Authority, regards restricted availability of land as the single greatest restraint on the wider application of passive mine water treatment (Parker, 1997). In response to this problem, significant research effort has been dedicated to finding methods for removing iron from net-alkaline mine waters at cramped and/or steep, sites.

The principle avenue explored in this research has been the possible use of surface-catalysed oxidation of ferrous iron (SCOOFI) using as the catalyst the ochre (i.e. ferric hydroxide) which naturally precipitates from such waters. The SCOOFI process occurs naturally at virtually all mine water discharges: it is operative wherever ochre can be seen to be accreting from mine water flowing at velocities in excess of that which would foster natural settlement of colloids from suspension. SCOOFI is a simple two-step process. In the first step, ferrous iron is removed from the water by sorption onto the highly polar surface of fresh ochre. Once the ferrous ions are attached, they are oxidised *in situ* by dissolved oxygen, with the ferric hydroxide acting as a catalyst, accreting a further layer on the surface of the ochre. Oxidation of ferrous iron in this manner is a far more rapid process than oxidation by dissolved oxygen in free solution. Hence on cramped sites, systems efficiently employing SCOOFI offer the potential for achieving "passive treatment without wetlands". Recent UK research has focused on the use of reactors packed with media of high specific surface area on which ochre can be accreted and SCOOFI initiated and sustained. The experiments have used commercial PVC trickling filter media and blast furnace slag, an abundant waste material characterised by a naturally high specific surface area.

Most experiments to date have been undertaken using unsaturated flow conditions, with water trickling through the media. In two reactors fed with an average influent iron concentration of 1.43 mg.l⁻¹ over a six-month period, mean effluent iron concentrations were 0.41 mg/l and 0.38 mg.l⁻¹. Variations in influent iron loading rates (achieved by varying the flow rate) demonstrated that iron removal rates increase linearly with loading rate, up to around 14 g.d⁻¹ (Figure 4). Although the relationship apparently becomes non-linear at higher loading rates, the rate of removal remains as high as 50% for a loading rate of nearly 32 g.d⁻¹ Fe. Residence time in these reactors is as little as 70 seconds. The combination of short residence time and efficient removal of iron at low influent concentrations strongly suggests that these reactors may be an effective passive alternative to aerobic wetlands, particularly at sites where topography and/or land area are restrictive. The preliminary results suggest that such reactors may be as much as an order of magnitude more efficient than aerobic wetlands (Jarvis and Younger, 2000).

In practice, the application of SCOOFI at full-scale requires consideration of a number of issues. For discharges of high flow rate, SCOOFI-based systems may need to be run under saturated flow conditions. This can be expected to have negative implications for oxygen transfer rates, which might be addressed by incorporating venturi devices into the influent pipework of the systems. Other practical issues relate to system maintenance requirements, such as the logistics and frequency of removal of some of the media for de-sludging, and the disposal or re-use of the ochre sludge. Current research at Skinningrove, Cleveland (site 14, Table 1 and Figure 1) is examining both the logistics of saturated flow SCOOFI applications and the practicalities and economics of long-term operation and maintenance. The two substantial Skinningrove discharges arise from adjoining, flooded ironstone workings that were abandoned in the 1960s (Younger, 2000b). The larger of the two discharges has a mean flow of about 2073 m³.d⁻¹, while the other discharges at about 1900 m³.d⁻¹. The discharges arise from shafts a few tens of metres apart, and have virtually identical chemistry, being brackish and net-alkaline with mean dissolved iron of 16.3 ± 0.82 mg.l⁻¹. The discharges emerge into the local river, the Kilton Beck,

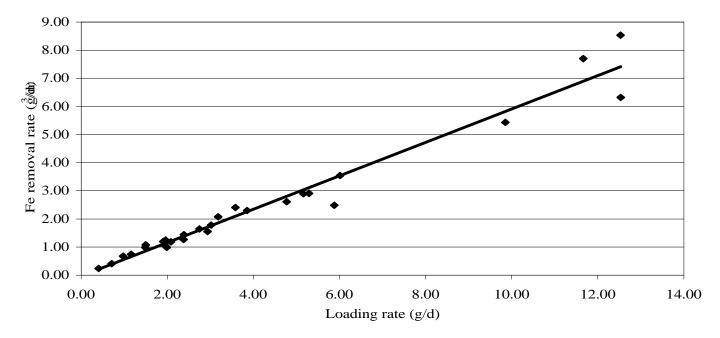


Figure 4. Iron removal rate as a function of loading rate for unsaturated flow of mine water through two pilot reactors filled with media of high specific surface area.

immediately upstream of the village of Skinningrove, the dwellings of which occupy all available flat land in the valley floor along the full length of the Beck to the North Sea. Apart from the visual impact of the discharge, it also acts as a significant barrier to migratory salmonid fish, which have to pass through the impacted reach of the river to access spawning grounds upstream. With no "problem owner" upon whom a duty to undertake active treatment might have been enforced, and with no flat land available for a wetland system, the possibility of using a SCOOFI-based system has been examined for this case. The intent is to construct the system beneath a car parking area, which can retain its primary use whilst affording subsurface space for mine water treatment. A pilot SCOOFI reactor was constructed to receive half of the flow of the

larger of the two discharges, and it was operated from October 1999 for six months. Influent and effluent samples were collected weekly by local residents. The reactor was only 9.5m in length, and comprised 8 gabions packed with blast furnace slag, arranged in series. Residence times in the reactor cannot have exceeded 10 minutes at any time during the period of operation, and were probably closer to four minutes on average. Substantial iron removal was observed (Figure 5). These data are being used to constrain designs for a full-scale SCOOFI-based reactor at this site (scheduled for construction in June 2000). The type of system envisaged will physically resemble an established low-cost water treatment technology, the "roughing filter", which is occasionally used to filter turbid surface waters in river intakes. Roughing filters are long channels packed with stones or other filter media, through which sediment-laden waters are passed to achieve rough filtration prior to disinfection. Although the principle of operation of the Skinningrove SCOOFI reactor differs somewhat from that of a conventional roughing filter, the same name is being applied to it on account of the similarity in form.

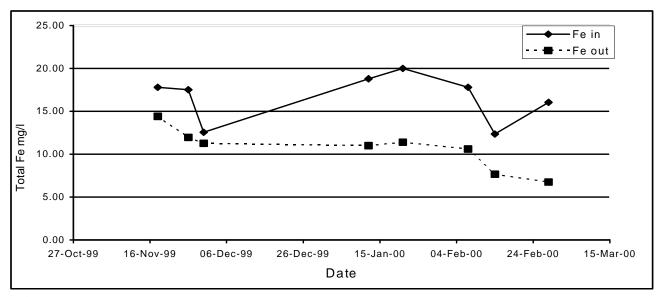


Figure 5. Influent ('Fe in') and effluent ('Fe out') iron concentrations for the pilot roughing filter at Skinningrove, Cleveland (site 14 on Figure 1 and in Table 1). Note that the filter was treating the full mine water flow (2073 m³.d⁻¹) up to 14th January 2000, after which half of the flow was diverted leaving 1036 m³.d⁻¹ to pass through the filter.

Removing Zinc as a Carbonate

Particular difficulties have been encountered in the UK with attempts to passively treat hard, netalkaline mine waters with elevated dissolved zinc (up to 40 mg/l). These waters are relatively common in the former Pb-Zn mining field of the North Pennines, where they cause significant ecological damage in receiving streams (Nuttall and Younger, 1999). Such waters are not readily amenable to sulphidisation in compost-based systems (Younger, 1997c), as ZnS does not readily precipitate at neutral pH. Experimental work has demonstrated that dissolution of limestone under closed-system conditions can create conditions favourable for the precipitation of zinc as its carbonate, smithsonite (ZnCO₃). Hence closed-system limestone gravel beds are a possible means of passive treatment for such waters (Nuttall and Younger, 2000). In basic form, these closed-system limestone dissolution units differ little from conventional anoxic limestone drains (ALDs; e.g. Hedin *et al.*, 1994). However, closed-system cells for zinc removal require venting to the atmosphere after a few hours of retention time in order to restore pCO₂ to relatively aggressive values. In this respect, their construction differs markedly from that of standard ALDs (Figure 3b), in which aeration steps would never be incorporated. Experimental data obtained to date suggest that a retention time of around four hours may be sufficient to remove 50% of the zinc from a water containing 5 - 10 mg.l⁻¹ Zn (Nuttall and Younger, 2000). Successive closed-system tanks in series can result in considerable net removal of Zn. Practical problems with the use of these systems may arise after some months of operation due to blinding of the reactive limestone surfaces with smithsonite. It is possible that this problem can be overcome by the use of fluidised bed configurations, though this will require further investigation.

PROCESS-LEVEL RESEARCH: FINDINGS AND PROSPECTS

Many of the passive systems listed in Table 1 have become foci for detailed scientific research, and are also being used as demonstration systems to foster the wider adoption of passive mine water treatment elsewhere in Europe. The emerging research agenda in the UK is for detailed, process-level investigations of the biogeochemistry of passive treatment systems. It is hoped that the outcomes of such research will assist in the gradual replacement of existing empirical design criteria with more scientifically-based design theory. One preliminary example of this type of process-level research, which relates to the identification of processes that raise pH in compost wetlands, is given below for illustration.

Compost-based systems can cope with a wider range of influent pH and a broader cocktail of contaminant metals than can aerobic reed-beds. Contaminant removal processes in most compost wetlands are predominantly microbial (e.g. Hedin *et al.*, 1994; Walton-Day, 1999). Bacterial sulphate reduction generally governs both the raising of pH <u>and</u> the removal of those metals that form sulphides at ambient temperatures and pressures (Fe²⁺, Zn²⁺, Cu²⁺, etc). The coupling of sulphate reduction to bicarbonate generation is often written:

$$2CH_2O + SO_4^{2-} \rightarrow H_2S + 2HCO_3^{-}$$
 (1)

Reaction of this bicarbonate with protons present in the water yields a rise in pH (at least as long as the CO_2 produced is able to vent to the atmosphere):

$$HCO_3^- + H^+ \rightarrow H_2O + CO_2$$
 (2)

This rise in pH fosters the precipitation of hydroxide minerals, which can help to remove metals that do not form sulphides at ambient temperatures and pressures (e.g. Al³⁺ and Fe³⁺).

Equations 1 and 2 are the most common formulation of the treatment pathway presumed to be occurring in compost wetlands (e.g. Hedin *et al.*, 1994; Walton-Day, 1999). However, preliminary process studies in the Quaking Houses pilot wetland (site 20, Table 1) suggest that these processes may not contribute so greatly to pH buffering (and hence to overall treatment

efficiency) as has hitherto been envisaged. Although sulphate reducing bacteria (SRB) were found to be abundant and active in the wetland substrate, and despite the known presence of limestone within the compost, the observed rise in pH was greater than could be explained by observed levels of SRB activity (as indicated by changes in dissolved SO₄²⁻ concentrations) and/or limestone dissolution (as revealed by changes in Ca and Mg concentrations). Freeze-coring and analysis of the wetland substrates showed that total sulphur was present in three forms, distributed in the following proportions:

On the basis of these observations, it is postulated that the pH rise in excess of that explicable by calcite dissolution and reactions (1) and (2) is explicable by the consumption of protons through a reaction chain involving reduction of ferric hydroxide and precipitation of native sulphur. While proton consumption by ferric hydroxide dissolution has previously been recognised as a potentially important process in lake sediments and constructed wetlands (Anderson and Schiff, 1987; Walton-Day, 1999), the inclusion of native sulphur precipitation in the reaction chain has not previously been considered. The simultaneous removal of iron from solution (which is clearly demonstrated by the data (Table 1) (Jarvis and Younger, 1999) and formation of significant quantities of S° is consistent with the following coupled reactions:

$$5H^{+} + 2FeOOH + HS^{-} \rightarrow 2Fe^{2+} + S^{o} + 4H_{2}O$$
 (3)

$$Fe^{2+} + H_2S + 2HCO_3 \rightarrow FeS + 2H_2O + 2CO_2$$
 (4)

Equations (3) and (4) explain how a net consumption of H⁺ can be driven by bacterial sulphate reduction (and partial re-oxidation of sulphide, but only as far as S^o) whilst having very little effect on alkalinity concentrations. For instance, HCO₃⁻ released by calcite dissolution, or by the sulphate reduction process represented by reaction (1) is consumed in reaction (4). If the sequence of reactions represented by (3) and (4) are realistic, then compost wetland treatment systems may be even more complex (and perhaps also more finely-balanced) than has previously been envisaged.

To consolidate and expand upon process-level studies such as this, the European Commission has recently funded a \$1.5M, three-year project entitled 'Passive In-situ Remediation of Acidic Mine / Industrial Drainage' (PIRAMID). Full details of the PIRAMID project are available at: http://www.piramid.org.

PIRAMID involves ten research and consultancy organisations in five EU Member States (UK, Spain, Sweden, France and Germany) and a candidate Member State (Slovenia), and is funded from March 2000 to February 2003. The objectives of PIRAMID are:

- 1. To assemble a database of European experiences with passive in situ remediation (PIR) of acidic mine / industrial drainage, covering both surface and subsurface PIR systems;
- 2. To develop process-based models of PIR system performance to support future design improvements;
- 3. To critically evaluate the potential application of PIR in areas of Europe which still do not have the technology;

- 4. To test novel approaches to PIR in both the laboratory and the field, targeted at other contaminants and using novel substrates; and
- 5. To develop engineering guidelines for PIR application at new sites throughout the EU.

The work programme designed to achieve these specific objectives is heavily weighted in favour of process-level research into passive treatment, whilst prioritising particular experiments on the basis of utility for end-users. Particular issues to be addressed include:

- <u>The hydraulics of passive treatment systems</u>: Although influent and effluent flow volumes are generally well-constrained by engineered structures, the internal hydrodynamics remain poorly understood, especially in systems where flow occurs in both surface and subsurface zones. Carefully planned tracer tests and low-rate pumping tests will be implemented to improve our understanding of hydraulic processes.
- Design criteria for passive systems: Tarutis *et al.* (1999) have made a convincing case for the replacement of unrealistic sizing models for aerobic wetlands, which implicitly assume "zero-order" kinetic models, with more appropriate first-order kinetic models. Although Tarutis *et al.* (1999) did not consider compost wetlands, data from the Quaking Houses and Pelenna III systems in the UK strongly suggest that a similar case can be made to revise sizing criteria for compost wetlands. The basis for such criteria will be examined by detailed sampling of pore waters and sediments supported by rigorous statistical analysis and kinetic geochemical modelling.
- Long-term performance of passive systems: Of prime concern to the owners and funders of passive treatment systems is an as-yet-unanswerable question about the longevity of such systems. While simple calculations of the longevity of aerobic wetlands can be made by assuming consumption of freeboard by steadily accumulating hydroxides, complications such as diagenesis of the hydroxides and remobilisation of iron have yet to be studied in detail, let alone incorporated into management guidelines. The uncertainties are considerably greater for anaerobic, compost-based systems. For instance, we need to know the rates at which labile organic carbon will be consumed by SRB, and the rate at which it will be replenished by seasonal die-back of macrophytes. Further issues arise in relation to rates of pore clogging by sulphide precipitates etc. Again, careful sampling and analysis of pore waters and sediments, including determinations of stable isotope ratios, are expected to yield substantial advances in understanding.

CONCLUSIONS

Passive treatment technology for mine waters was introduced to the UK during the 1990s, principally by adoption of U.S. Bureau of Mines experiences (Hedin *et al.*, 1994). Regulatory acceptance was soon gained, and passive treatment is now the technology of choice wherever possible for the long-term remediation of mine water discharges. Innovations in passive treatment technology are now beginning to arise in the UK in response to issues of limited land availability and the occurrence of peculiar net-alkaline mine waters very rich in zinc. Roughing filters, which remove iron from mine waters by the SCOOFI process, are now being developed to full-scale as an alternative to wetland treatment where space is very limited. Zinc removal from waters of neutral pH appears to be feasible using a variant of ALD technology. Scientific research into passive system performance is being marshalled via a \$1.5M European Commission project (PIRAMID), running from 2000 to 2003. While PIRAMID will incorporate

activities relating to collation of existing data and dissemination of engineering guidelines, it also encompasses basic process-level research into the hydraulics, reaction kinetics and longevity of passive systems.

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