

# Passive mine water treatment at the former Wheal Jane tin mine, Cornwall: important biogeochemical and microbiological lessons

Kevin B. Hallberg and D. Barrie Johnson

## Abstract

*Acid mine drainage (AMD) discharging from the former Wheal Jane tin mine, Cornwall, is remediated using conventional lime treatment. A pilot-scale passive treatment system was established at the site following the catastrophic release of about 50 000 m<sup>3</sup> of AMD in January 1992. The treatment system is essentially a demonstration and research facility, and all of the water processed in the passive system is returned to the main site for lime-treatment.*

*The pilot plant is a composite treatment system, comprising (in sequence) five constructed aerobic wetlands (aerobic cells), a single compost reactor (anaerobic cell) and a series of ten in-line algal ponds (or 'rock filters'). The aerobic cells are designed to facilitate the removal of iron, via oxidation of ferrous iron and hydrolysis of the ferric iron produced, and arsenic co-precipitation with ferric iron. Chalcophilic metals (e.g. zinc and cadmium) are to be removed as sulphides in the compost bioreactor; while generation of alkalinity in this cell is another key component of the bioremediative strategy. Finally, the rock filters were installed for the dual function of encouraging manganese removal (oxygenic photosynthesis increases the pH of mine water to promote the oxidation and precipitation of manganese) and mineralisation of dissolved organic carbon compounds in water draining the compost reactor. In addition, the pilot plant has the option of pre-dosing the inflowing AMD (with lime) to a desired pH or passive chemical treatment using an anoxic limestone drain, making it a unique facility to study various options for passive biological AMD treatment.*

*Since 1999, a consortium of UK universities and research organisations, under the auspices of the UK government-sponsored LINK scheme, has been carrying out detailed research on the (bio)geochemistry and microbiology of the Wheal Jane passive system, with a view to increasing knowledge of how such systems operate, and how their behaviour might be modelled. It has become apparent that there are many features of the current design and operation of the plant that have little or no impact on AMD remediation or, in some cases, mitigate against the net objectives. The key processes involving the biogeochemical cycling of iron and sulphur, with concomitant production (or consumption) of proton and mineral acidity have been identified and problem areas highlighted. Microbiological studies have revealed an intriguing, and unexpected, level of biodiversity at the site, and have pointed to new opportunities to use microbiological systems in constructed wetlands or in more controllable systems for AMD remediation.*

**Key words:** acid mine drainage, acidophiles, bioremediation, constructed wetland, iron oxidation, manganese removal, sulphate reduction

## INTRODUCTION

The Wheal Jane tin mine, located in Cornwall, England,

closed in 1991 following several hundred years of continual mining operations in the vicinity. In 1992, the failure of an adit plug led to the catastrophic release of acidic, heavy-metal laden water (often referred to as acid mine drainage, 'AMD') that had flooded the closed mine. About 50 000 m<sup>3</sup> of the acidic iron-rich water (200 mg Fe/L), which also contained significant

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concentrations of zinc (100 mg/L), cadmium (0.1 mg/L) and arsenic (6 mg/L), spilled out into the Carnon river and into the Fal estuary, leaving behind an iron-hydroxide coating on the floor of the estuary. Although the impact of this disaster was mainly visual, similar incidents at the Aznacollar mine complex in Spain and Baia Mare gold mine in Romania have had a much more negative environmental impact on the water bodies that they have polluted and also on the surrounding land. As a result of the Wheal Jane incident, a research facility was constructed close to the mine in order to study the effectiveness of a 'passive' treatment system for the remediation of the Wheal Jane AMD. This study site, referred to throughout this paper as the Wheal Jane site, was built in 1993 and operated for a period of almost five years. Data concerning the operation of the treatment system can be found elsewhere (Hamilton *et al.*, 1999). While the term 'passive' is used to describe the treatment system, it is instrumented and controllable.

The site consists of three composite treatment systems that vary only in the handling of the AMD that enters each system (see Figure 1). AMD entering the individual systems: (i) passes through an anoxic cell, containing manure, to strip it of oxygen before passage through an anoxic limestone drain (ALD system); or (ii) the AMD is dosed with lime to pH 5.5 (LD). AMD which flows through the third system (LF) has not undergone any pre-treatment. In all cases, the AMD then enters into a series of five aerobic cells that consist of shallow (20 cm water depth) ponds planted with *Typha latifolia*, *Phragmites australis* and *Scirpus lacustris*. These cells were designed to encourage the oxidation and precipitation of iron, with the concomi-

tant removal of arsenic. Next, the AMD in each system flows through a compost bioreactor, which consists of sawdust and manure buried underground, to encourage sulphate reduction activity for the removal of chalcophilic metals (such as copper, cadmium and zinc) as sulphides. The activity of the sulphate reducing bacteria (SRB) also generates alkalinity. Finally, the AMD flows through a series of shallow pools containing algae-colonised rocks to encourage a further rise in water pH (>8) for the precipitation of manganese, which is not removed in the aerobic or anaerobic cells, and to reduce biological oxygen demand from the compost bioreactor.

Recently, a multi-disciplinary group has been established to study the geochemistry, hydrology and microbiology of the Wheal Jane remediation site to further understand the principles of passive treatment of AMD. As part of this group, we are studying the microbial populations in the treatment systems to discern the role, beneficial or otherwise, of the microbes present in the systems. Here we summarise our studies that have highlighted potential problems with the use of such a system for the remediation of AMD, and we suggest ways to overcome the problems. An intriguing, and unexpected, level of biodiversity at the site has also been revealed, pointing to new opportunities to use microbiological systems in constructed wetlands or in more controllable systems for AMD remediation.

## IRON OXIDATION AND REMOVAL IN THE AEROBIC CELLS

The oxidation of iron in the aerobic cells of the Wheal

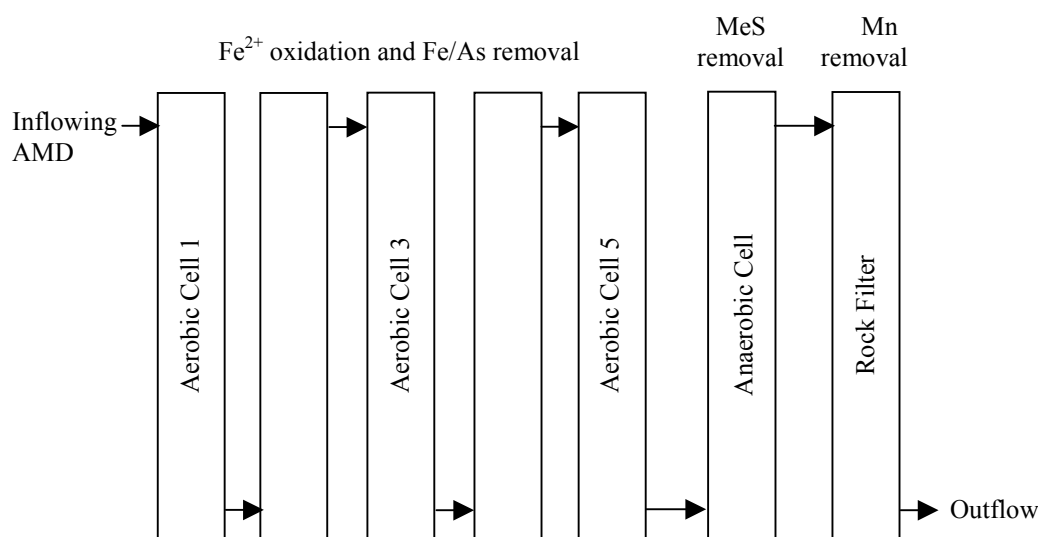


Figure 1. Schematic diagram showing all of the components in one of the treatment systems at the Wheal Jane site

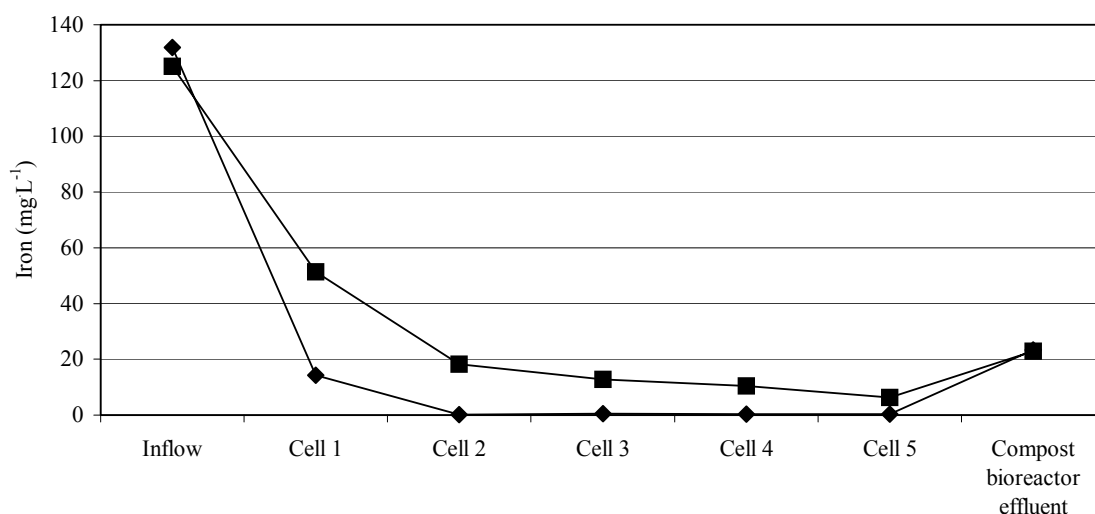


Figure 2. The concentration of soluble (filtered through a 0.2  $\mu\text{m}$  pore-sized filter)  $\text{Fe}^{2+}$  (◆) and total soluble iron (■) in water samples taken, in October 2001, from the inflow, aerobic cells and from the compost bioreactor effluent of the lime-dosed system. Similar trends were found for the other two treatment systems on this date and over a two and a half year period.

Jane passive treatment plant occurs very rapidly. Typically, between 80 and 98% of the ferrous iron is oxidised in the first half of the first aerobic cell for each of the three systems (see Figure 2 for typical data). The soluble (filtered through a 0.2  $\mu\text{m}$  pore-size filter)  $\text{Fe}^{2+}$  in water samples from each of the remaining aerobic cells is usually below 100  $\mu\text{g/L}$ , although it ranges from 0.1 up to 5 mg/L in the latter cells in the ALD and LD system (not shown). The role of microorganisms in the oxidation of iron in these aerobic cells has been well established (Grahame Hall, CEH Windermere, unpublished; Hallberg and Johnson 2002).

There is a simultaneous decrease of the pH in each of the systems from about 5.5 (LD), 5.0 (ALD) and 3.5 (LF) to 3, where the pH remains throughout the rest of the aerobic cells. The similar rates of iron oxidation and removal between the three systems imply that pH adjustment of the AMD prior to entry into the remediation systems is not only unnecessary, but also a costly waste.

The concentration of total soluble iron in each aerobic cell of the three treatment systems remains high (10–20 mg/L, Figure 2). This is quite surprising given the low solubility of ferric iron at pH 3 (calculated to be 0.5 mg/L), but may be explained by the formation of iron colloids, in another wetland site, that have been found to pass through the filters of the same type as we use (Peiffer *et al.* 1999). Of interest, and contrary to the successful remediation of AMD, is the increase of soluble iron in the compost bioreactor effluent. The increase in total soluble iron is puzzling, although on

more than one occasion iron (oxy)hydroxide particles of about 1 mm diameter were seen flowing into the anaerobic cell. We also hypothesise that during periods of high rainfall, the iron (oxy)hydroxides get washed into the compost bioreactors, where the iron is reduced (either directly by iron-reducing microbes or by the sulphide generated by the SRB) and subsequently solubilised. The consequences of this iron ‘remobilisation’ will be discussed below (see the section on manganese removal).

## IRON REDUCTION IN THE AEROBIC CELLS

On occasional sampling, it was noticed that the concentration of ferrous iron increased as the water passed through the cells of the LD and ALD systems. The major difference between these systems is the luxuriant plant growth in the former relative to the latter. The rise in ferrous iron concentration in the water indicates that iron reduction is occurring in these cells, consistent with the isolation, from sediment samples of the aerobic cells, of acidophilic microbes that can couple the reduction of iron with the oxidation of organic compounds (Hallberg and Johnson 2002).

To further study the relationship between the plants and the reduction of iron, the chemistry of the sediment pore water was characterised. The pore water samples from aerobic cells 1, 3, and 5 of the ALD and LD systems had much higher (up to 100 times) ferrous iron than the overlying surface water. They also had from

two to ten times the concentration of dissolved organic carbon compared to the corresponding surface water. The pore water from the ALD cells consistently exhibited higher concentrations of both iron and organic carbon. The strong correlation between organic carbon and metal solubility found here has been observed in other vegetated wetlands (Beining and Otte 1996; Goulet and Pick 2001).

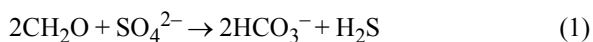
Sediment samples were taken from the first aerobic cell of the ALD and LD systems and placed in a synthetic medium, sparged with nitrogen to prevent iron oxidation, containing soluble ferric iron and either no substrate or glucose as substrate. The sediment samples that were incubated with no substrate produced very little ferrous iron from four to ten days (Figure 3). In contrast, those sediment samples that had been incubated in a glucose-containing medium generated significant ferrous iron over the same time period. These results indicate that the iron-reducers found in the samples are indeed active and could be the cause of the increase in ferrous iron found in the aerobic cells. The overlying water of the aerobic cells has always been found to be saturated with regard to oxygen, but no such data for the sediment pore waters have been obtained. It should be noted here that the iron-reducing microbes isolated from the Wheal Jane site belong to the genus *Acidiphilium*, which are capable of reducing ferric iron even in the presence of oxygen (Johnson and Bridge 2002).

The reduction of ferric iron seen in the aerobic cell sediment samples may compromise the efficiency of the remediation process in this treatment system. In addition to the remobilisation of the iron, any other

metals (or metalloids such as arsenic) that have co-precipitated with the iron will also be resolubilised. The latter metals may well be retained in the compost bioreactors that follow the aerobic cells, but, as will be discussed below, the ferrous iron may not.

## COMPOST BIOREACTOR OPERATION

The compost bioreactors are underground chambers filled with a mixture of straw, sawdust and manure. The straw and sawdust were used to provide solid substrate support and also to serve as a long-term source of organic carbon, while the manure was added as an immediate source of organic carbon and as a source of inoculum for sulphate-reducing bacteria (SRB). The purpose of these bioreactors is to remove chalcophilic metals not removed in the aerobic cells by sulphidogenesis, and also to increase the pH of the water through the generation of bicarbonate (Eq. 1). Due to operational problems, the LF system bioreactor has been closed for a period of over six months and will be dealt with separately.



Routine monitoring of the effluents from the ALD and LD compost bioreactors for over two years has revealed that these reactors are not functioning as they should. As mentioned above (see the section on iron oxidation and removal), the effluents of these bioreactors contain more soluble iron (exclusively as  $\text{Fe}^{2+}$ )

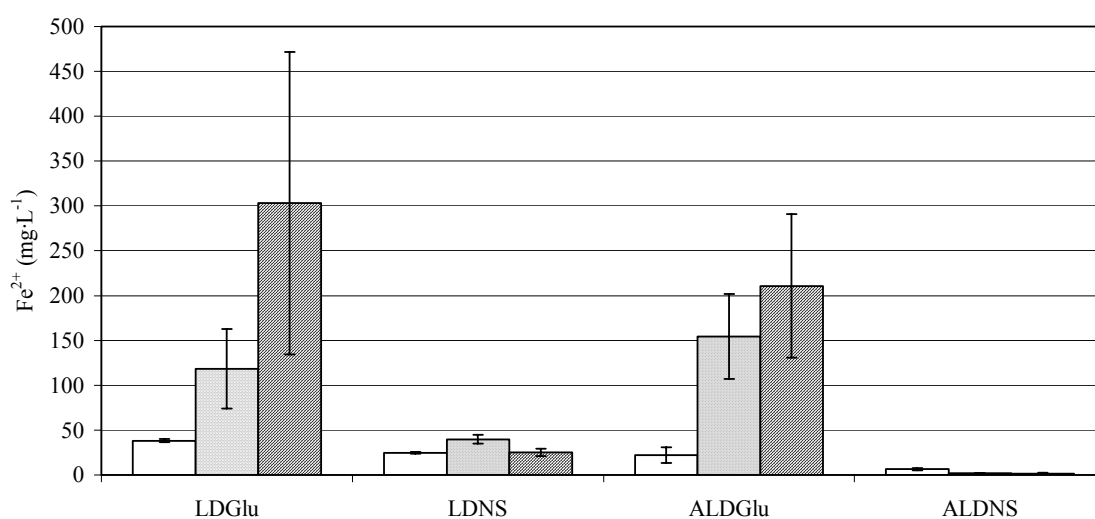


Figure 3. Changes in ferrous iron concentration catalysed by sediment samples from the LD or ALD aerobic cell 1 after 48 hours (white bars), 168 hours (dotted bars) or 240 hours incubation (striped bars). The data are the means from duplicate incubation experiments incubated with glucose (Glu) or with no exogenous substrate (NS) with the standard error for each duplicate.

**Table 1. Sulphur and iron mass balance data in mine water before and after passage through Wheal Jane compost bioreactors. These data were taken on one sampling visit; similar trends have been noted for more than two years**

| System: water source | Sulphate-S (mg/L) | Sulphide-S (mg/L) | Ferrous iron (mg/L) | Ferric iron (mg/L) |
|----------------------|-------------------|-------------------|---------------------|--------------------|
| 'LD' : influent      | 130               | <0.2              | 1.6                 | 16                 |
| : effluent           | 113               | 10                | 56                  | <0.1               |
| : Rock Filter 1      | 118               | <0.2              | 49                  | <0.1               |
| 'ALD' : influent     | 143               | <0.2              | 2.4                 | 9.8                |
| : effluent           | 73                | 12.5              | 13                  | <0.1               |
| : Rock Filter 1      | 98                | <0.2              | 18                  | 2.0                |

than the influent (Table 1). Also, the pH of the water draining these bioreactors is only moderately increased, from ~3.0 to, on average, ~5.5. This low pH precludes the formation of iron sulphides and hence the removal of iron from solution.

Due to the configuration of the bioreactors, the microbiology inside the systems has been difficult to study. Enumeration and characterisation of the microbes in the effluents, however, has been carried out. Very few (<100 m/L) SRB have been detected in these effluents. In contrast, high numbers ( $10^3$ – $10^4$  m/L) of iron- and sulphur-oxidisers have been detected in these effluents. These include microbes such as *Acidithiobacillus ferrooxidans* (Fe- and S-oxidiser), *At. thiooxidans* (S-oxidiser) and Fe- and S-oxidising *Thiomonas* species (Hallberg and Johnson 2002). As these microbes were present at much lower numbers in the compost bioreactor influents, it is apparent that they are growing within the bioreactors. Bacteria of the genus *Acidithiobacillus* are often considered to be obligately aerobic, but it is well known now that *At. ferrooxidans* is capable of growth on reduced inorganic sulphur

compounds ('RISCs', including sulphur) with ferric iron as electron acceptor in the place of oxygen. *At. thiooxidans* is incapable of such growth, but it appears to be able to grow anaerobically by disproportionation (inorganic fermentation) of RISCs (Hallberg *et al.* 2001), and it is presumed that the *Thiomonas* species can either couple the oxidation of RISCs to ferric iron reduction or may also grow by disproportionation of reduced sulphur. Thus it appears that microbes with the wrong phenotype are growing in these bioreactors, leading to poor remedial function.

The iron and sulphur in the compost bioreactor effluents affect the overall performance of the treatment system. As would be expected, the sulphide in the effluents is very unstable upon re-oxygenation of the effluents, with a half-life in the order of minutes. The ferrous iron, on the other hand, is relatively more stable, but over time is oxidised to ferric iron upon re-oxygenation of the effluents in the presence of bacteria (Figure 4). The combination of substrates and the presence of iron- and sulphur-oxidisers in these effluents leads to re-acidification of the effluents (pH 3 to 3.5

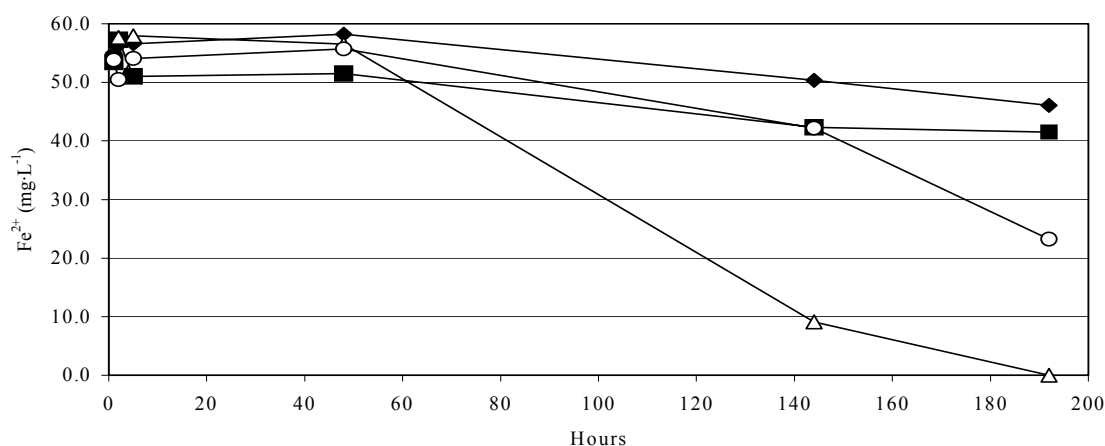


Figure 4. Oxidation of ferrous iron in duplicate samples of the compost bioreactor effluent from the LD system in the absence (solid symbols) and in the presence of indigenous microbes (open symbols)

from 5.4), while the slow abiotic oxidation only leads to slight acidity (4.7 to 4.6 from 5.4). The iron and the sulphide in the ALD effluents are much more labile (even in the absence of bacteria), indicating that the two systems are not behaving identically.

Operational problems at the treatment site have led to the closure of the LF system for a period of almost 1 year. To protect the compost bioreactor, it was decided to close the system so that sulphate-poor water (i.e. rain water) would not enter the system and thus starve it. Upon resumption of AMD flow to the LF system, a change in the effluent chemistry from the compost bioreactor system was noted. The pH of the effluent increased from 3 to between 6 and 7 and the sulphide and iron (again exclusively ferrous) decreased as well. After four months of continuous operation, the iron in the effluent decreased to levels below that of the influent (<0.5 mg/L and 1.5 mg/L, respectively). During the same time period, SRB were detected at much higher numbers (100 times) in the LF effluent than from the ALD or LD bioreactor effluents. It thus appears that this compost bioreactor is now behaving as expected.

#### MANGANESE REMOVAL IN THE ROCK FILTERS

Soluble manganese ( $Mn^{2+}$ ) is often found in considerably greater amounts in AMD than in unpolluted streams and groundwater (Banks *et al.* 1997). As with iron and aluminium, manganese also contributes to the total mineral acidity of mine waters. It is readily precipitated as  $Mn^{4+}$ , but in contrast to ferrous iron, little oxidation of  $Mn^{2+}$  occurs in solutions below pH 8. The lack of affinity of manganese for sulphide prohibits any significant removal as a sulphide in compost wetlands and bioreactors. Therefore, a biological approach for removing manganese from solution, by causing solution pH to rise above 8 via oxygenic photosynthesis, has been developed (Bender *et al.* 1994; Phillips *et al.* 1995).

This approach has been used at the Wheal Jane passive treatment site with varying results. Periodical monitoring of the rock filter effluent chemistry has shown that, in general, the rock filters have failed. The pH of the effluents was typically between 5 and 6 over a two-year period, and the manganese concentration has shown that very little, if any, removal has occurred. One potential reason for this is that Cornwall receives relatively little sunlight and thus the algae are unable to raise the pH sufficiently. This appears not to be the case, however, as no seasonal fluctuations were recorded.

A more likely explanation for the failure of the rock filters to remove manganese is the failure of the com-

post bioreactors that lie before the rock filters. The ingress of sulphide and iron into the rock filters leads to acidification of the water in the pools. Ferrous iron has been found at concentrations of greater than 10 mg/L in the second and even the third rock pool (of ten consecutive pools). It is also likely that the acidity, and possibly other toxic metals that are present in the bioreactor effluents, inhibits algal activity. Careful monitoring of the pH and manganese concentrations in each individual pool shows the correlation between low pH and high manganese concentrations in the LD rock pools (Figure 5A). This is in contrast to the rock pools of the LF system (sampled after the system was restarted), where the high pH found in the latter rock pools correlates well with low manganese concentration (Figure 5B).

#### CONCLUSIONS

The microbiological studies presented here, coupled with chemical monitoring of the Wheal Jane composite wetland remediation scheme, have allowed us to draw some useful conclusions concerning the use of wetlands for the remediation of acid mine drainage.

Careful study of the aerobic cells has shown that iron oxidation and precipitation occur very rapidly in these cells. It is thus apparent that excess capacity exists, nearly four and a half cells, in which further oxidation can occur. This is important to note, as careful modelling of the system will require knowledge of the area in which iron removal is occurring, rather than simply calculating removal rates on the area of the wetland itself.

The knowledge that microbes are responsible for the oxidation of iron is also an important finding. Previous studies (e.g. Kirby *et al.* 1999 and references therein) that have drawn the conclusions that iron-oxidation in wetlands of pH 3–6 is primarily abiotic are probably incorrect. Such conclusions are often based on the lack of iron-oxidising microbes, though in these studies only extremely acidophilic (growth <pH 3) microbes were sought. Iron oxidation by the microbes isolated from Wheal Jane is inhibited below pH 3, as is *in situ* oxidation by microbes attached to sediment material from the aerobic wetlands (Grahame Hall, CEH, unpublished). This information can be used to redesign the aerobic cells, such that periodic pH adjustment is included, for example, through successive alkalinity production systems (Barton and Karathanasis 1999).

Similarly, microbiological studies have shown that iron-reducing microbes also reside in the aerobic cells. Chemical analysis of sediment pore water indicated high ferrous iron content where there were large concentrations of dissolved organic carbon. These sedi-

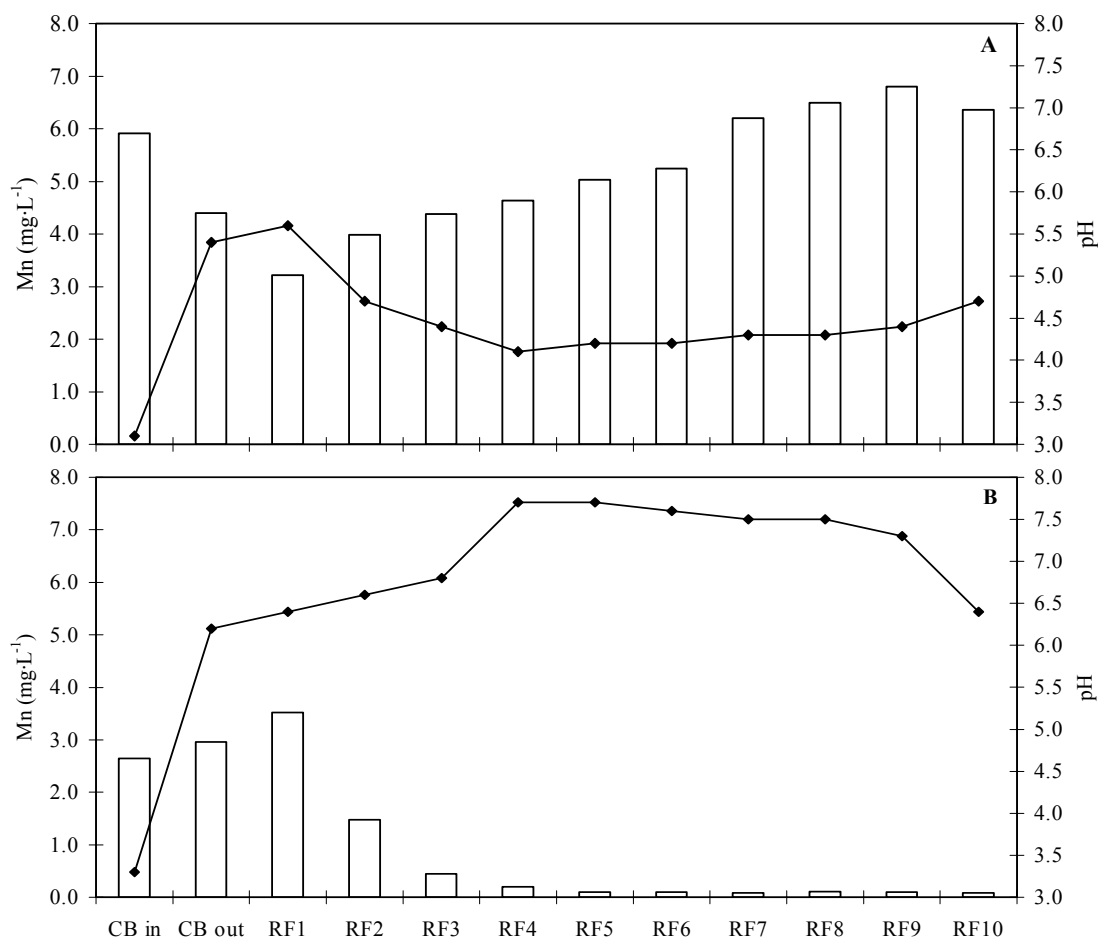


Figure 5. Manganese and pH trends in the compost bioreactor influent (CB in) and effluent (CB out) and in each of the ten rock filter pools (RF1–RF10). The samples were taken in April 2002, from the LD (A) and LF (B) systems. Bars show soluble manganese and the lines show pH.

ments came from the aerobic cells with the most luxuriant plant growth. The same sediment samples were able to catalyse the reduction of soluble ferric iron to ferrous iron only when given an organic substrate. It thus appears that the presence of plants in the aerobic wetlands may be counter-productive in terms of iron retention. Furthermore, the flow of AMD in the aerobic wetlands with more plant growth is channelled down the sides, and thus the exposure to the oxidising sediment material is limited. The use of plants in aerobic wetlands designed for the treatment of high iron-containing AMD should be questioned, considering that the aerobic cells of the LF system (with virtually no plant growth) are as efficient in promoting iron oxidation and removal as those of the other two systems with plants.

For complete remediation by this composite system, each of the components must work efficiently. As

shown here, the compost bioreactors in the LD and ALD systems are not functioning as designed. Currently they are simply reversing the work done in the aerobic cells. The importance of the proper function of each of the components of such a composite treatment system is illustrated by the poor manganese removal by the rock filters of the ALD and LD systems, in comparison to the LF system where the compost bioreactor is functioning properly.

Two important observations should be considered when using such a system for AMD remediation. The first is the importance of keeping the solid ferric iron out of the compost bioreactors. A deep pool prior to the inlet of the bioreactors may allow proper settling of the iron (oxy)hydroxides from solution before entry. The second important observation to this end is the shutdown of the LF system, which is now functioning as designed. This shutdown-time must have allowed the

SRB to become established and active, such that they can now cope with the acidity of the influent. As an alternative, one must look to better sources of inoculum for such systems, such as the acidophilic/acidotolerant SRB that we found in acid mine drainage impacted streams (Sen and Johnson 1999).

By providing examples where microbiological studies on the Wheal Jane AMD passive treatment plant have highlighted shortcomings of such a system, we hope that we have highlighted the fact that the uncertainties about using constructed wetlands and compost bioreactors to remediate AMD described in this article will only be answered by programmes of basic, multi-disciplinary research into these complex ecosystems.

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