Mitigation of pollution from abandoned metal mines

Part 1: Review of passive treatment technologies for metal mine drainage remediation

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Miranda Kavanagh

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Executive summary

A review of passive treatment technologies for metal mines has revealed that a wide range of passive treatment systems are available, which operate with varying degrees of performance. Such systems range from proven technologies operating at full scale, through technologies that appear to offer promise but have yet to be demonstrated at full scale, to emerging technologies that have been investigated at laboratory-scale only.

The main aim of metal mine water treatment is to immobilise metals within a treatment system and, where necessary, raise the pH. Although acidity (low pH) is less commonly a pollutant of metal mine discharges (in contrast to coal mine drainage), pH is an important parameter in treatment system design since it has a crucial control on the geochemical behaviour of metals in aqueous environments. Where pH is low, generation of alkalinity by dissolution of alkali materials or adding alkali chemicals is a critical first step in treatment.

Unlike coal mine drainage remediation, in which iron is removed by the generation of (oxy)hydroxides under oxic conditions, metal mine drainage remediation is more problematic in that other metals have higher hydroxide solubility products than iron and therefore the target pH for their removal is higher. Although, in principle, zinc and other metals in such discharges can be removed via precipitation as their hydroxides within aerobic passive treatment systems (such as settlement lagoons and aerobic wetlands), this review reveals that the area-adjusted removal rate for metals in these systems is very low. Consequently, aerobic passive treatment systems for the remediation of metal mine drainage would need to be very large, leading to problems identifying areas of land of sufficient size and high capital costs of construction.

Alternative processes, such as co-precipitation, adsorption and bioaccumulation, may also result in the immobilisation of metals within treatment systems. Various studies have investigated the sorption of metals onto different types of media, such as ochre pellets, caustic magnesia, zeolites and bauxsol™. However, problems with media exhaustion and armouring (coating) were noted, most experiments were done in laboratory columns, and many studies were only conducted over a short period (months). Therefore issues of scale would need to be investigated before the technology could be applied at full scale, as would issues of system longevity. Nevertheless, further development may offer promise for the future, especially for technologies that regenerate media within the treatment units.

The most promising means of immobilising metals at the pH values typically achievable in passive treatment systems (up to a pH of 8) appears to be anaerobic systems which encourage bacterial sulphate reduction in organic substrates in order to simultaneously generate alkalinity and immobilise metals as sulphides. However, an essential requirement for these systems is the maintenance of bacterial sulphate reduction at sufficient rates to immobilise metals within a reasonable timeframe. With this in mind, many studies have investigated the periodic addition of carbon sources to the compost substrate to maximise rates of bacterial sulphate reduction. Such carbon additives range from alcohols, such as ethanol and methanol, to cheese whey and wine wastes. Other studies have investigated the inoculation of the compost with sulphate reducing bacteria before reactor start-up. The majority of studies, however, have been undertaken at lab scale and do not account for the scale issues or different environmental conditions at actual mine sites. The duration of many of these experiments is also too short to give any confidence that the system would operate effectively over the long time periods necessary for a full-scale system. Further work is therefore necessary before full-scale implementation of compost-based systems can be pursued, such as pilot-scale experimentation, and identification of carbon additives.

Contents

1.   Introduction

This review focuses on the passive remediation of discharges from abandoned metal mines. Passive remediation has been defined as "*improvement of water quality using only naturally available energy sources, in gravity-flow treatment systems (such as wetlands or subsurface-flow reactors) which are designed to require only infrequent (albeit regular) maintenance to operate successfully over their design lives*" (PIRAMID Consortium 2003). Although this defines passive treatment in the strictest sense, the difficulty of successful passive remediation of metal mine drainage in the UK (for reasons outlined in this review) is such that here we prefer to think of there being a sliding-scale between fully passive and fully active treatment. Thus, for example, technologies for metal mine water remediation may embrace many elements of passive treatment, but be supplemented with periodic additions of liquid or solid materials to enhance treatment performance; for which reason we refer in this report to what we term *enhanced passive treatment*. A cursory review of the 50 or so coal mine water treatment systems operated by the UK Coal Authority (CA) shows that mine water treatment typically lies somewhere between fully passive and fully active in practice. The CA operates very few fully passive systems as per the definition above, operates many that rely on pumping (usually to deliver mine water to the treatment system), some that rely on some element of chemical (usually alkali) dosing, and one or two that are fully active with chemical dosing, stirred-tank reactors and so on, such as the High Density Sludge (HDS) plant at Dawdon, County Durham.

Decision-making for mine water treatment is therefore site-specific, and dependent on factors such as contaminant load, topography, land availability and so forth. This report does not provide a site-specific review of treatment options for the many hundreds of abandoned metal mine water discharges in England and Wales. Instead, we offer a detailed review of the types of passive treatment system available and their performance, in the context of the mine water geochemistry and physical environments encountered in the UK. Clearly whilst the context is the UK - and specifically England & Wales - the literature drawn upon in the review is international. It is also worth noting that whilst the focus is England and Wales we often refer to the 'UK' since the breadth of mine water geochemistry and flow rates in England and Wales encompasses that found in other parts of the UK. The review encompasses what may be regarded as proven technologies that have been implemented at full scale, through technologies that appear to offer promise but have yet to be demonstrated at full scale, to emerging technologies that are still at the stage of proof-of-principle testing (usually at laboratory-scale).

Active treatment may be the only feasible solution for discharges with the highest flow rates and highest contaminant metal concentrations (highest metal loads). Such technologies may range from rudimentary Oxidation, Dosing with Alkali and Settlement (ODAS) approaches, through High Density Sludge (HDS) treatment, to highly intensive processes such as biodesalination and membrane processes (such as reverse osmosis, nanofiltration). These technologies are not discussed here, as the review is explicitly focused on passive treatment. However, the interested reader is directed to Chapter 4 of Younger *et al*. (2002) for further details on active treatment.

To set the review in the context of UK mine water geochemistry, it is necessary to identify the key contaminants in UK mine waters. This is dealt with in the section below. In brief, the most common contaminants of non-coal mine discharges in England and Wales are metals. Acidity (low pH) is less commonly a pollutant of non-coal mine drainage and, although it is a critical treatment system design variable, it is rarely a cause of failure of environmental quality standard (EQS) limits in its own right at downstream points, because of simple dilution effects. Sulphate can also be a

pollutant in mine drainage. However, because non-coal mine drainage tends to arise from the oxidative dissolution of monosulphides it is less of an issue than for coal mine waters. Additionally, in the UK it is unusual for sulphate to be present in receiving watercourses at concentrations high enough to be a hazard to aquatic ecology, and dilution effects typically mean it does not exceed EQS limits at downstream locations. The focus of this report is therefore on metal contaminants, albeit with reference to pH because (a) it is a critical variable for treatment system design and (b) mine waters in England and Wales that have low pH are amongst the worst nationally in terms of the metal loading they impart on receiving watercourses.

Much has been written recently about passive mine water treatment by the authors of this report. In particular, recently completed work by Jarvis et al. (2009) contains a targeted review of passive mine water treatment options for abandoned metal mine water discharges. Some of the text from that report is reproduced here since it remains both pertinent and up to date. Of course many other sources of literature are also cited here. However, rather than exhaustively referring to the large volumes of literature available, this review has targeted key publications that serve to illustrate the actual and potential passive technologies available and their performance.

This is one part (Part 1) of a two-part review collated as part of this project. The second part (Part 2: *Review of resource recovery options from the passive remediation of metal-rich mine waters*) considers potential options for resource recovery from metal mine water discharges across England and Wales. Although the two reviews have been compiled separately, the extended Executive Summary (a separate document) draws conclusions from both, and relates these conclusions to each other. This is not least because the type of passive treatment system selected will crucially influence the potential for resource recovery, particularly with respect to metal recovery from spent treatment substrates.

2. Characteristics of metal mine drainage in England & Wales

The most common contaminants of metal mine drainage are metals. An analysis of the data collected as part of the Department for Environment, Food and Rural Affairs (Defra) and Environment Agency project *Identification and prioritisation of abandoned non-coal mines* summarises the characteristics of mine waters in terms of key metal contaminants and their concentrations in metal mine water discharges across England and Wales. Data for around 270 individual discharges were collated as part of that project. The data need to be interpreted with some caution since, for example, not all potentially polluting metals are always determined, and the analyses are sometimes for one-off samples. Nevertheless the data provide a useful indicator of the main metal pollutants. Appendix A summarises data for seven metals/metalloids (As, Cd, Cu, Fe, Pb, Mn, Ni). The data in this appendix shows that cadmium and lead significantly exceed (at least double) their respective EQS values in many metal mine water discharges. Copper and manganese marginally exceed their respective EQS values in a large number of metal mine water discharges, whilst arsenic, iron and nickel are rarely above EQS values (although there are notable exceptions).

Figure 1 illustrates the same data for zinc and flow rate. It is immediately clear from the graph for zinc that the data are heavily skewed to the right (that is, high concentrations are common. Indeed, 124 of the 146 mine waters analysed have a zinc concentration of > 0.03 – 30 mg/l, which is approximately 4 – 4000 times the indicative EQS of 0.0078 mg/l (see Appendix A for explanation of why these are indicative EQS values). In short, zinc is by far the most common contaminant of metal mine water discharges across England and Wales (no other metal or metalloid shows the same right-skewed pattern).

Flow-rate data are also shown in Figure 1 (some of these data are from the Defra/Environment Agency project, but the majority are unpublished data, much of which have been collated by Dr Will Mayes, Hull University). Flow rate is critical for mine water treatment system design, and it is therefore crucial to have an appreciation of the range of flows of metal mine water discharges. The data illustrate that the majority of discharges have flow rates in the range 0.05 – 25 l/s (litres per second).

The prevalence of zinc in metal mine water discharges across England and Wales is very significant in terms of the key objectives of passive treatment of metal mine water discharges (discussed at length below). Although it is not a priority substance, geochemically zinc is a conservative metal, the immobilisation of which is therefore challenging. It will be demonstrated below that the use of aerobic wetland systems for passive metal mine water treatment is unlikely to be feasible. Therefore compost-based bioreactors are the most promising option. In such systems divalent metals are immobilised as sulphides. Table 1 shows the solubility products of various metal sulphides, and shows that the solubility product of zinc is higher than lead or cadmium (the second two most prevalent metals exceeding their EQS values). Therefore lead and cadmium will be removed as a sulphide before zinc. Thus, a compost-based reactor designed to remove zinc will also remove lead and cadmium, since they will be immobilised before zinc.
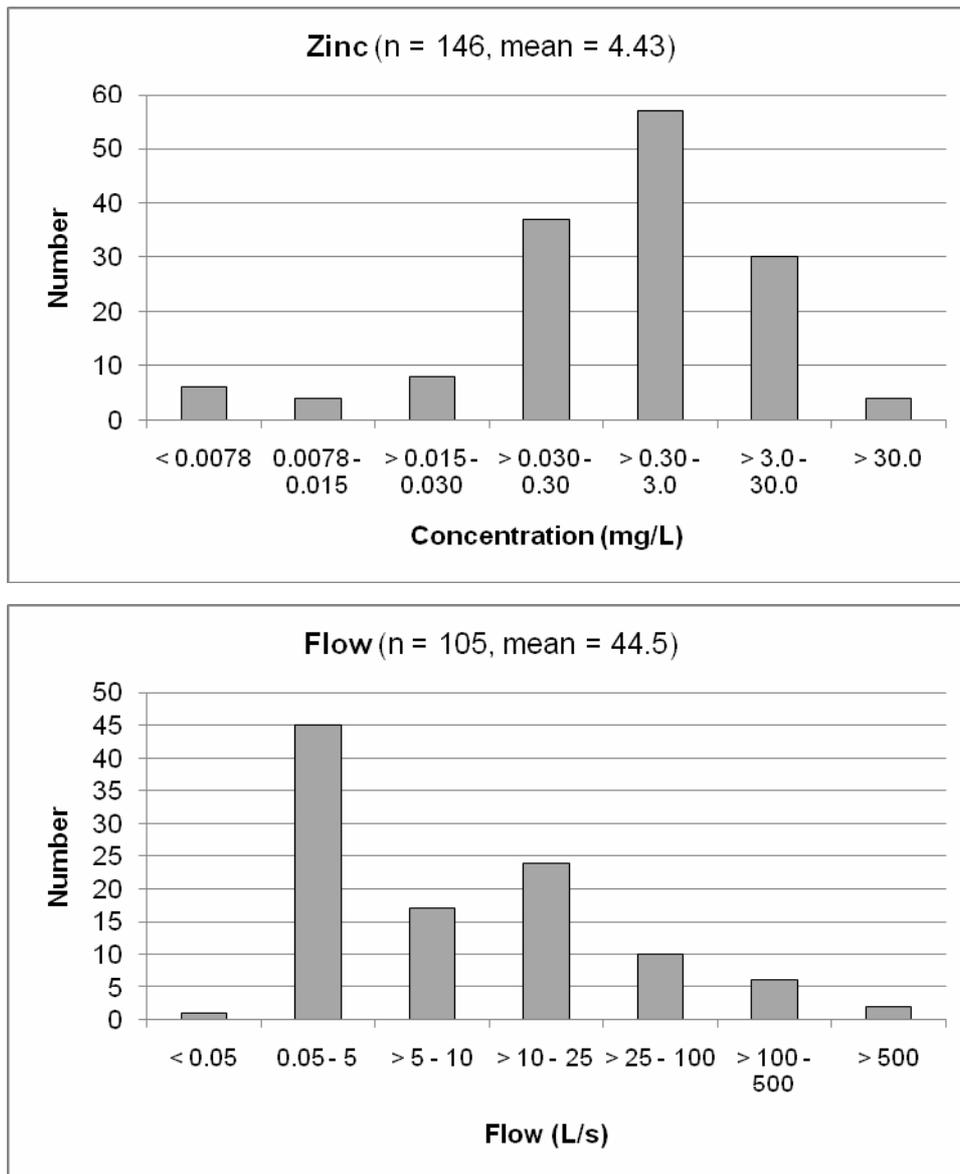
Figure 1: Summary data for metal mine water discharges, showing the concentrations of zinc and flow rates of metal mine waters across England and Wales (see Appendix A for explanatory notes)

Data for pH are not shown graphically here, but previous assessment of data reveals that some 80% of discharges have pH > 6. Neither are sulphate concentration data shown here, for several reasons: (1) it is less commonly determined, (2) it is rarely a regulatory requirement in England and Wales to lower sulphate concentrations and (3) if it was a regulatory requirement to lower sulphate concentrations significantly then passive treatment would unlikely be a feasible option because of the very conservative behaviour of sulphate.

Many of the analyses of metal mine waters to date have been one-off samples. For the design of mine water treatment systems far more comprehensive data are required, in order to appreciate (and factor into design) the variability of mine water discharges. This is particularly the case for flow-rate data, which has historically been omitted from characterisation exercises and/or poorly measured.

3. Biogeochemical principles of mine water treatment

Where the removal of sulphate to statutory limits is not a requirement, the primary aim of non-coal mine water treatment is almost always to raise the pH, where necessary, and immobilise metals within the confines of the treatment system (Hedin *et al.* 1994b; PIRAMID Consortium 2003). Even where pH is not outside regulatory limits close attention needs to be paid to it, since pH is a crucial control on the geochemical behaviour of metals in aqueous environments. Where pH is low, or the water is net-acidic, generation of alkalinity is a critical first step in treatment. The majority of discharges from abandoned metal mines are of near-neutral pH; 80% of all metal mine waters monitored across England and Wales have pH > 6 (unpublished data, Environment Agency). Nevertheless, discharges that exhibit low pH are among the most polluting in the UK, such as Parys Mountain. Therefore, options for elevating pH/generating alkalinity are an important aspect of this review.

Typically, an abiotic increase in pH is accomplished by dissolution of alkaline materials such as calcite, or the addition of alkali chemicals, most commonly sodium hydroxide (caustic soda) or hydrated calcium hydroxide (lime) (Younger *et al.* 2002). The relevant reactions are as follows:

Dissolution of calcite at pH < 5 (Reaction 1) and pH > 5 (Reaction 2):

$$CaCO_3 + 2H^+ \leftrightarrow Ca^{2+} + H_2O + CO_2 \tag{1}$$
$$CaCO_3 + H_2CO_3 \leftrightarrow Ca^{2+} + 2HCO_3^- \tag{2}$$
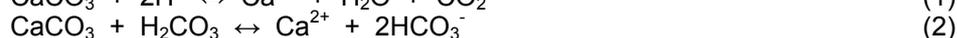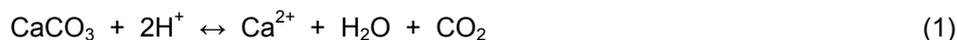
Increasing pH using sodium hydroxide (3) and calcium hydroxide (4):

$$NaOH + H^+ \leftrightarrow Na^+ + H_2O \tag{3}$$
$$Ca(OH)_2 + 2H^+ \leftrightarrow Ca^{2+} + 2H_2O \tag{4}$$

Using alkali chemicals in stirred-tank reactors can elevate pH rapidly (in minutes). In passive treatment systems, in which calcite dissolution is the main source of alkalinity, reaction times are longer. Hedin *et al.* (1994a) suggest optimal contact time for generation of alkalinity in limestone reactors is 14 hours (note the substantial difference between this figure and the actual residence time of waters in many passive systems; Table 3 in particular).

A key process in the remediation of mine water pollution is the generation of solid metal (oxy)hydroxides (and sometimes carbonates) under oxic conditions (this is a key process in every full-scale mine water treatment system currently operating in the UK). The prevalence of precipitation depends on the specific metal and the pH. Specifically, the rate of precipitation of hydroxides increases with pH, which is precisely why the elevation of pH, by one of the means identified above, is required (note that some metals are amphoteric, and therefore become more soluble at very high pH). This is the principal mechanism by which iron is removed from coal mine drainage: mine water is aerated to generate oxidising conditions in which ferrous iron is rapidly converted to ferric iron (5), and the ferric iron then forms a solid hydroxide precipitate which is typically retained in settlement lagoons or wetlands (6).

$$Fe^{2+} + \tfrac{1}{4}O_2 + H^+ \rightarrow Fe^{3+} + \tfrac{1}{2}H_2O \tag{5}$$
$$Fe^{3+} + 3H_2O \leftrightarrow Fe(OH)_3 + 3H^+ \tag{6}$$

Iron is readily removed in this way because its hydroxide solubility product is very small, as indicated in the right hand column of Table 1. Thus, it will form readily in the presence of even low concentrations of hydroxyl ions (OH⁻), and this is reflected in the pH values for formation of hydroxides shown in Table 2, where Fe^{3+} will begin to form at relatively low pH. Furthermore, as pH increases and more hydroxyl ions become available, the rate of removal will increase. Because of the relative ease with which iron can be removed as a hydroxide in passive and active treatment systems, the discussion below does not consider the attenuation of this metal from metal mine drainage in detail, even though it may be present at high concentrations (especially in low pH discharges).

What is also clear from Tables 1 and 2 is that other metals behave differently; the hydroxide solubility products of all the metals shown are greater than that of iron, and the target pH for their removal is higher. This characteristic of differing metal solubility is used to advantage in some (active) mine water treatment technologies which, through careful pH control, facilitate selective precipitation of metals, in order to recover them for possible reuse. However, in passive treatment systems the very high pH required to immobilise some metals as hydroxides (such as manganese) is a significant problem, and this is taken up further in the discussion below.

Table 1: Solubility products (log K_{sp} at 25ºC) of sulphides and hydroxides of metals, arranged in order of readiness with which the precipitate will form (from top to bottom) (data from Diaz *et al*. 1997)

Metal	Sulphide	Metal	Hydroxide
Cu^{2+}	-35.9	Fe^{3+}	-38.6
Cd^{2+}	-28.9	Pb^{2+}	-19.9
Pb^{2+}	-28.1	Cu^{2+}	-19.8
Zn^{2+}	-24.5	Fe^{2+}	-16.3
Ni^{2+}	-21.0	Zn^{2+}	-16.1
Fe^{2+}	-18.8	Ni^{2+}	-15.3
Fe^{3+}	-	Cd^{2+}	-14.3
Mn^{2+}	-13.3	Mn^{2+}	-12.7

Table 2: Target pH values for removal of various metals as their hydroxides (from Younger *et al.* 2002). Note that optimal pH values for rapid removal may be higher than the values indicated

Metal	pH
Fe^{3+}	3.5
Cu^{2+}	6.8
Pb^{2+}	6.8
Zn^{2+}	8.2
Fe^{2+}	8.5
Cd^{2+}	9.8
Mn^{2+}	10.2

pH is not the only variable that requires consideration with respect to the immobilisation of metals. Oxidising conditions are required for precipitation of metals as their hydroxides. Redox potential (Eh) is therefore also an important control on the state of metals in aqueous environments, and the pH-Eh relationship can be used to predict the behaviour of metals. A pH-Eh diagram for zinc is shown in Figure 2. What can be seen here is that the solid phases $ZnCO_3$ (smithsonite) and $Zn(OH)_2$ are generally favoured in oxidising conditions in which the pH is ~ 8 for $ZnCO_3$ and > 8 for $Zn(OH)_2$. Note the sharp contrast to Fe^{3+}, the main contaminant in coal mine drainage, which will begin to form a solid hydroxide, $Fe(OH)_3$, at pH > 3.5 under oxic conditions (Table 2).

The discussion thus far has focused on abiotic mechanisms for increasing pH and removing metals as hydroxides under oxic conditions. However, pH may also be increased under anoxic conditions by an important biotic process, which may simultaneously immobilise divalent metals as sulphides. Dissimilatory bacterial sulphate reduction (BSR) is a much cited process in mine water treatment (see Blowes *et al*. 2004; Hedin *et al*. 1994b; Walton-Day 1999; Younger *et al*. 2002), particularly in relation to passive remediation. The principle is that sulphate, which is invariably present at elevated concentrations in mine waters, can be reduced under anoxic conditions by sulphate reducing bacteria. The reduction of sulphate by BSR can

consume protons (Reaction 7), generate alkalinity (Reaction 8), and simultaneously release sulphide to form a precipitate with divalent metal ions (Reaction 9) (Walton-Day 1999).

$$2CH_2O \ + \ SO_4^{2-} \ + \ 2H^+ \ \rightarrow \ 2CO_2 \ + \ H_2S \ + \ 2H_2O \qquad (7)$$
$$SO_4^{2-} \ + \ 2CH_2O \ \leftrightarrow \ H_2S \ + \ 2HCO_3^- \qquad\qquad (8)$$
$$H_2S \ + \ M^{2+} \ + \ 2HCO_3^- \ \rightarrow \ MS \ + \ 2H_2O \ + \ 2CO_2 \qquad (9)$$
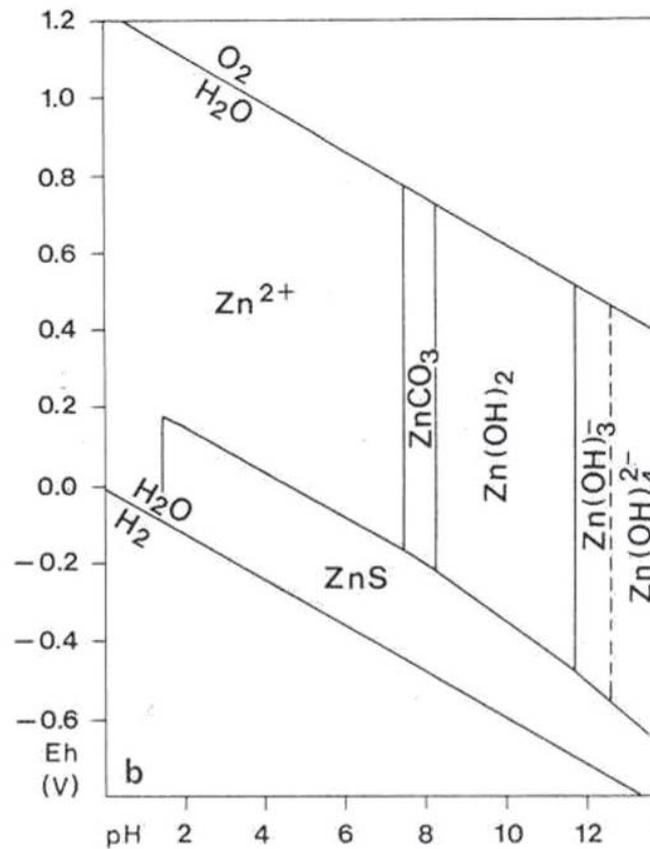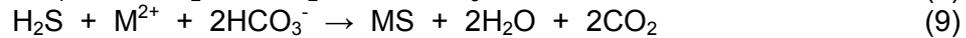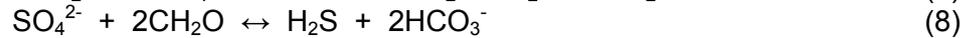
Figure 2: Eh/pH diagram for the Zn-O-H-S-C system at 25ºC and 1 atm (Salomans and Förstner 1984)

In order to promote anoxic conditions, and provide a source of carbon to enable sulfate-reducing bacteria to metabolise, the substrate for passive treatment systems harnessing BSR is some form of organic medium such as compost. To further enhance the generation of alkalinity, limestone is often mixed with the organic substrate, resulting in calcite dissolution. Sulphate reduction is harnessed in both active and passive systems. In the UK the focus has been use of BSR in passive systems, with the principal aim of raising the pH and immobilising at least a portion of the metal contaminants within the organic substrate. In other countries sulphate reducing bioreactors have been developed as active systems, with the aim of lowering sulphate concentrations to regulatory limits. This has been particularly the case in semi-arid and arid countries, in which mine water may need to be treated to standards appropriate for subsequent potable use.
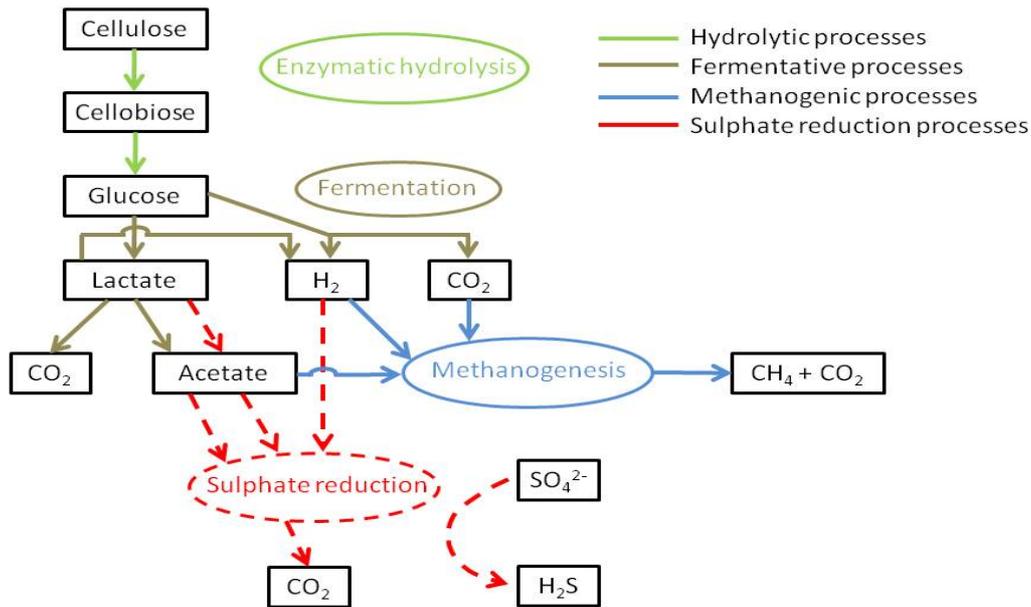
Figure 3: Illustrative process diagram for the breakdown of cellulose to facilitate sulphate reduction. The rate-limiting step is the breakdown of cellulose to cellobiose (from Wildeman and Gusek 2009).

Given the solubility products of the sulphides and hydroxides of metals (shown in Table 1), it is apparent that BSR may well offer a more feasible means of immobilising metals such as zinc at pH values typically achievable in passive treatment systems (it is difficult to achieve pH 8 in passive systems). However, a key requirement for accomplishing this is the maintenance of BSR at rates sufficient to immobilise divalent metals within a reasonable timeframe (if the timeframe is too long, system size is likely to be unrealistically large). A review of literature published to date strongly suggests that currently rates of BSR achieved in passive systems are not high enough to immobilise metals within treatment systems with a residence time of approximately 24 – 48 hours (the approximate target residence time for systems in the UK to make passive treatment a realistic option). A critical reason for this appears to be the availability of carbon within the compost substrate which, crucially, is required for sulphate reducing bacteria to metabolise. The breakdown of complex carbon compounds into available carbon compounds is a limiting step to the rate of BSR. The process of cellulose breakdown, ultimately to facilitate sulphate reduction, is illustrated in Figure 3. The rate-limiting step is the initial breakdown of cellulose to cellobiose. Whilst compounds such as acetate are therefore desirable to encourage BSR, there in fact needs to be a combination of available carbon and complex carbon sources to ensure ongoing BSR but also maintain system longevity, such that the complex carbon compounds are broken down more slowly over time, facilitating long-term BSR. Much research effort has been devoted to identifying suitable carbon sources for maximising rates of BSR, and this is discussed below.

Other mechanisms may also serve to immobilise metals within treatment systems. Such processes include adsorption, co-precipitation and bioaccumulation. Whilst treatment systems are not in general designed explicitly on the basis of such processes, they may be significant in certain situations, as discussed below. Like precipitation many of these processes are pH-dependent, with implications for the rate of removal. As an example, Figure 4 illustrates the pH-dependency of sorption of various metals onto hydrous ferric oxide (HFO). It is noteworthy that (a) there is a finite

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capacity for sorption of metals onto a fixed mass of sorbent and (b) desorption of previously sorbed metals may occur if pH/Eh conditions change, potentially releasing metals back into solution.
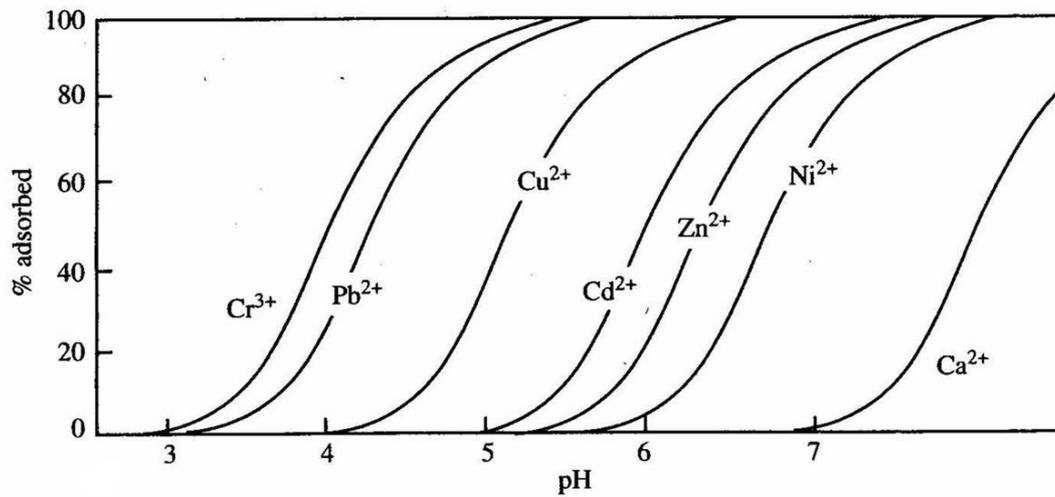
Figure 4: pH specific adsorption edge profile (for adsorption onto hydrous ferric oxides) for a range of cations (from Gaillardet *et al*. 2003)

4. Passive treatment technologies and their application to metal mine water discharges

The principles and design of passive treatment systems for mine water treatment have been discussed in detail by the PIRAMID Consortium (2003) and Younger *et al.* (2002) among others. Both volumes are widely available and exhaustive details are not repeated here other than where it is pertinent to the discussion. This section focuses on the potential application of these treatment systems to the problem of metal mine drainage in England and Wales.

The UK has a track record of using passive treatment systems for coal mine drainage remediation that is unrivalled in Europe. More than 50 full-scale systems are currently in operation, the majority installed by the UK Coal Authority. However, with the exception of the HDS plant at Wheal Jane, all of them have the primary aim of raising pH (where necessary) to facilitate the removal of iron within the treatment system. For non-coal mine drainage, iron is not the only cause for concern (and even where it is present, the Coal Authority's track record is evidence enough that it can be dealt with effectively). Removal of other metals, such as zinc and cadmium, requires different geochemical conditions to that needed for removal of iron, particularly with respect to the pH.

In principle at least, zinc and other metals present in non-coal mine drainage can be removed via precipitation as their hydroxides within aerobic passive treatment systems, such as settlement lagoons and aerobic wetlands (*cf* Figure 2). In addition, Nuttall (1999) illustrates the possibility of removing zinc as its carbonate (smithsonite) in anoxic limestone drains (ALD) (*cf* Figure 2). Adsorption and co-precipitation may also be an effective means of attenuating metals such as zinc, using high surface area media under appropriate geochemical conditions (Figure 4). The performance of a range of passive treatment systems in which the authors of the publications report removal of zinc are provided in Table 3 (whilst the majority were designed for metal removal, not all were explicitly designed to do so). Whilst Table 3 does not provide an exhaustive list of performance data for passive systems which receive zinc as a contaminant, it nevertheless offers some illuminating data with respect to the potential application of current passive treatment technologies to the remediation of non-coal mine drainage.

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Table 3: Performance of different types of passive treatment technology, using various assessment metrics (figures derived where not explicitly given in text of references) (from Mayes et al. 2009; citations therein referenced again here for convenience)

1	2	3	4	5	6	7	8	9	10	11
Wastewater type	System type	Monitoring period (months)	Area (m^2) Volume (m^3)	Flow (l/s)	Residence time (days)	Zn influent/ *effluent* (mg/l)	Load removed (g/day)	Treatment efficiency (%)	Area-adjusted removal rate (g/m^2/day)	Volume-adjusted removal rate (g/m^3/day)
Mine water[1]	Pilot-scale ochre drain	10	0.35 m^2 0.21 m^3	0.03	0.02	1.7/*1.2*	1.30	32	3.71	6.19
Mine water[2]	Pilot-scale aerobic wetland/algal mat	48	420 m^2 240 m^3	0.04 - 0.17	16 - 79	16/*1.2*	191.8	91	0.47	0.80
Mine water[2]	Full-scale aerobic wetland/algal mat	24	13,200 m^2 6,000 m^3	8.5	8.2	14.4/*9.9*	3,300	31	0.25	0.55
Mine water[3]	Pilot-scale anoxic limestone drain	1.5	4.5 m^2 2.25 m^3	0.02	0.65	6.91/*5.74*	2.02	17	0.45	0.90
Not specified[4]	Subsurface flow wetland	-	- -	-	-	2.50/*0.73*	-	71	0.11	-
Urban runoff[5]	Full-scale surface flow wetland	12	8,250 m^2 -	18	-	0.065/*0.057*	12.4	12	0.002	-
Domestic wastewater[6]	Full-scale gravel subsurface flow wetland	9	650 m^2 325 m^3	6.25	6.7	0.113/*0.014*	53.5	88	0.08	0.16
Zn-containing wastewater[7]	Lab-scale hydrosoil surface wetland	2	0.896 m^2 0.224 m^3	0.003	1.00	1.76/*0.34* 1.76/*0.52*	0.319 0.278	81 70	0.37 0.31	1.42 1.24
Smelter water[8]	Lab-scale subsurface wetland	10	0.231 m^2 0.069 m^3	8.3 x 10^{-5}	7.00	0.140/*0.043*	7 x 10^{-4}	69	0.003	0.01

Sources of data: [1]Mayes *et al.* (2009); [2]Kalin (1998); [3]Nuttall (1999); [4]Kadlec and Knight (1996); [5]Scholes *et al.* (1998); [6]Lesage *et al.* (2007); [7]Gillespie *et al.* (1999); [8]Song *et al.* (2001)

Of all the data shown in Table 3, Column 10 is arguably the most significant. This reports the area-adjusted removal rate for zinc within each of the systems. The area-adjusted removal rate shown in this column is the mass of metal removed per unit area of treatment system per unit of time. It is calculated by dividing the metal load (in g/day) removed within the treatment system by the area of the treatment system (in m^2). Measurements of the area-adjusted removal rate in wetland systems treating coal mine drainage have revealed that on average iron is removed at an area-adjusted removal rate of 10 $g/m^2/day$ in aerobic wetland systems receiving water with circum-neutral pH (Hedin *et al.* 1994b). This figure of 10 $g/m^2/day$ remains the basis for design of coal mine drainage treatment wetlands for removal of iron under aerobic conditions, using the following formula:

$$A = \frac{Q_d(C_i - C_t)}{R_A}$$

where:

A = required wetland area (m^2)

Q_d = average daily flow rate to wetland (m^3/day)

C_i = average daily influent iron concentration (mg/l)

C_t = target iron concentration (mg/l)

R_A = area adjusted contaminant removal rate ($g/m^2/day$)

The formula above shows that the contaminant load (the numerator) must be known to calculate wetland area and, most importantly in the current context, the value of the area-adjusted removal rate has a major bearing on the overall size of the wetland.

Returning to column 10 of Table 3 it can be seen that the area-adjusted removal rate of zinc in the passive treatment systems is with only one exception less than 0.5 $g/m^2/day$, and in a number of cases at least an order of magnitude lower. Thus, from the data shown in Table 3, an aerobic wetland for attenuation of zinc would need to be at least 20 times the size of such a system for equivalent remediation of iron pollution (a similar principle applies to settlement lagoons, for which hydrolysis and precipitation of metals are also the main processes). Given the upland locations, and consequently steep topography, of many non-coal mine discharges around England and Wales, the likelihood of identifying areas of land sufficient to accommodate such large wetland systems is slim at best. Quite apart from this consideration, the capital costs of constructing such large systems are very high, principally due to the earth-moving/import costs. Other metals are similarly difficult to remove from solution as their hydroxides, as shown by the solubility products of metal hydroxides in Table 1. Specifically, nickel, cadmium and manganese are in fact all more difficult to remove than zinc (quantitative manganese removal has been noted in some aerobic wetland systems operated by the Coal Authority. However, it is not clear whether (a) the rate is high enough in terms of area-adjusted removal rate or (b) what the precise mechanism of removal is e.g. sorption, removal as oxide (personal communication, Dr Ian Watson, Coal Authority, UK)).

Possible passive treatment units for metal mine drainage, and their performance, are reviewed in the remainder of this document. The review is conducted in light of these very low area-adjusted removal rates for the primary metal contaminants in metal mine drainage, and thus conclusions are drawn on the prospects for effective passive remediation of metal mine drainage across England and Wales.

Mitigation of pollution from abandoned metal mines

4.1   Aerobic passive treatment systems

The primary mechanism of metal removal in aerobic systems is oxidation and hydrolysis to promote precipitation of a hydroxide precipitate. Aerobic systems are best suited to the removal of iron, arsenic and to some extent manganese. In contrast, anaerobic (compost) systems may attenuate zinc, cadmium and copper (Lamb *et al*. 1998). Since the latter are among the main metals of concern in metal mine drainage, the discussion of compost systems is a particular focus here. Nevertheless, aerobic systems are reviewed for completeness.

For coal mine drainage, in which the principal metal pollutant is iron, the main aerobic units used for this metal removal are settlement lagoons and aerobic wetlands (whilst the term aerobic wetland is used, such systems also contain anoxic/anaerobic zones below the water-substrate interface, which may well play a role in metal attenuation). Here, the focus is on the performance of such systems for the removal of metals (other than iron) typically found in metal mine discharges. Literature on this subject mainly relates to constructed wetland systems, some of which also incorporate (as part of the wetland or as a stand-alone unit) algal mat treatment. Not all studies relate specifically to remediation of mine drainage (e.g. highways runoff, urban drainage), but they are nevertheless insightful in terms of the rates and mechanisms of metal removal.

Various investigations have explored the potential utility of aerobic wetland systems for metal removal. Scholes *et al.* (1998), for example, evaluated the performance of a full-scale (> 8,000 m^2) system for removal of zinc from urban runoff, but removal efficiency was just 12%. Furthermore, calculation (by these authors) of the area-adjusted removal rate reveals a figure of just 0.002 g/m^2/day (see Table 3), far too low to be realistically applied to metal mine drainage treatment. Gillespie *et al.* (1999) also investigated transformations of Zn, but in this case in two laboratory-scale wetlands comprising hydrosoil. Removal rates of 81% and 70% were recorded for the two wetlands over a 24-hour retention time. Thus, at first glance these results are more promising, with the authors noting that 98% of the attenuated Zn was partitioned to the sediment or hydrosoil, with 2% adsorbed to plants. However, the experiments were only conducted for a period of two months (see Table 3). The experience of these authors, in conducting research for the Environment Agency (Mayes and Jarvis 2008) and ongoing research for the Waste Resources Action Programme (WRAP), is that over this timeframe adsorption processes dominate attenuation (consistent with 98% partitioning to sediment and soil), but after 3 – 6 months adsorption capacity becomes exhausted, and treatment performance rapidly deteriorates in the absence of other biogeochemical mechanisms of attenuation. Thus, it is concluded that the insight of the work of Gillespie *et al.* (1999) is rather limited.

Song *et al.* (2001) studied the performance of laboratory-scale constructed wetlands to treat neutral synthetic lead mine drainage. The work overcomes the limitations of that by Gillespie *et al.* (1999) insofar as the system was monitored over a period of 300 days. During this time the average removal of metals was 90% for Pb and 72% for Zn, and 70% for sulphate, which is likely by conversion to sulphide by BSR according to the authors. Three primary metal removal mechanisms were concluded to be responsible for the attenuation: (1) adsorption or exchange onto sediments in the wetland, (2) sulphide precipitation in reducing zones catalysed by bacterial metabolism, and (3) co-precipitation with (or adsorption onto) Fe and Mn oxides. On this evidence the remediation is therefore effective. However, additional data presented in Table 3 shows the limitation of the systems: residence time of mine water within the system is 7 days (and consequently area-adjusted removal rate is very low at just 0.003 g/m^2/day). This is far too long to be a realistic option for a passive system in the UK, given restrictions on land availability.

Lesage *et al.* (2007) identified similar metal removal mechanisms to those of Song *et al.* (2001), but investigating full-scale systems, using domestic wastewater, and over three years of operation. All metals except Fe and Mn were efficiently removed. Removal efficiencies varied between 49% for Ni and 93% for Al with >84% for Al, Zn and Cu. However, careful reading of the manuscript reveals the same difficulty: hydraulic residence time of the system is 6.7 days, far too high to be a realistic option in the UK.

Kalin (1998) studied both pilot-scale and full-scale biological polishing ponds which comprised an aerobic wetland with an algal mat. Four Zn removal mechanisms were identified by Kalin (1998): (1) co-precipitation or adsorption of Zn onto surfaces of ferric iron hydroxides precipitated as groundwater enters an oxygenated flooded pit, (2) adherence of suspended solids containing Zn to algal biomass surfaces, (3) adsorption of dissolved Zn onto algal cell walls, and (4) precipitation of Zn carbonate onto algal mats. Although the pilot-scale system had a Zn removal rate of 88%, residence time within the systems was between 16 and 79 days. The full-scale system subsequently operated had a residence time of 8.2 days (still above a realistic residence time for UK systems), and an annual average of 31% Zn removal. Although better performance was observed during the summer – thought to be due to higher temperature, pH and oxygen concentration in the water, together with growth of algae – such a system is unlikely to be practical for the UK.

Boshoff *et al.* (2004) and Rose *et al.* (1998) have also investigated the use of algae in passive mine water treatment systems. However, in this case the algae were utilised as a carbon source for BSR, and therefore these systems are discussed further in the section on anaerobic and anoxic passive systems, below.

4.2 Removal of metals by adsorption, co-precipitation and bioaccumulation

Sorption processes refer to the removal of metals from solution via binding of the metals to the charged surfaces of oxides and organic matter (Appelo and Postma 2006). Co-precipitation attenuates metals by binding them within the lattice of another precipitating metal. Co-precipitation of heavy metals with iron hydroxide is one common example. Finally, bioaccumulation is the quantitative uptake of metals by flora in particular, such as reeds and algae. These processes may be used to treat metal mine water discharges individually or in combination with other processes. Efforts to harness these processes in treatment systems are reviewed here.

Alternative approaches to passive remediation of zinc and other metals include harnessing high rates of sorption onto various types of media (*cf* Figure 3), and/or co-precipitation at high pH. The pilot-scale system investigated by Mayes *et al.* (2009) using ochre pellets (hydrous ferrous oxide sourced from coal mine water treatment systems) is one such example, the performance data for which are shown in the first row of Table 3. This particular system exhibited a comparatively good area-adjusted removal rate of 3.7 g/m^2/day over the 10-month period of operation, with an average treatment efficiency of 32%. The principal mechanisms of removal were sorption to the ochre pellets used in the system, and co-precipitation with calcite-dominated secondary minerals at high pH. Elevated pH early in the trial (up to pH 11.8) was attributed to dissolution of the cement used as a binder to create the ochre pellets. Although such high pH values clearly encourage the removal of metals, rapid precipitation of calcium carbonate minerals can cause problems of armouring of the ochre, and the costs of full-scale implementation may be prohibitive given the cost of the cement used as a binder. The problem of armouring could be overcome by pre-weathering/rinsing of the ochre pellets prior to use, using alternative binders or using a smaller volume of cement.

Mitigation of pollution from abandoned metal mines

PIRAMID (2003) describe the use of caustic magnesia (main constituents MgO and CaO) for the attenuation of metals such as Zn and Cu at laboratory scale. Significant metal removal, associated with high pH, may have been linked to co-precipitation of metals with amorphous Fe and Al oxyhydroxides. However, pH dropped before the end of the experiment due to armouring of the caustic magnesia by Zn and Cu precipitates. Low pH led to re-dissolution of precipitates and an increase in effluent Zn and Cu. In an attempt to increase porosity and reduce problems of clogging, the experiment was repeated using a mix of caustic magnesia and quartz. Using at least 50% caustic magnesia, the effluent pH remained consistently around 9 and removal of Zn was highly efficient. A residence time of 1.3 to 1.6 hours was required.

PIRAMID (2003) also investigated, in laboratory experiments, the use of zeolites (hydrated aluminium silicates of Ca, Na, K and Ba) for sorption of metals as they are known for their strong ion exchange capacities. Preliminary experiments showed that zeolites were capable of retaining a significant proportion of the Zn and Cu in influent waters despite relatively high concentrations of Ca which competes for exchange sites on the zeolite. The volume of waste produced is approximately four times greater than that produced by conventional lime treatment, so its greatest use may be as a polishing treatment particularly for removal of Cu, Zn and Cd.

Perkins *et al*. (2007) report on an alternative approach which utilises dealginated seaweed as a sorbent for metals such as Pb, Zn and Cd. In contrast to others, the authors show that pH does not exert a strong control over the adsorption process. The material is selective in its adsorption of metals, showing a greater tendency to adsorb Pb than Zn and Cd. Initially, removal rates are high for short residence times but sorption capacity of material appears to become exhausted quickly, raising issues of how such systems might be maintained over the medium to long term.

Ríos *et al*. (2008) evaluated, at laboratory scale, the use of low-cost sorbents like coal fly ash, natural clinker and synthetic zeolites to remove heavy metals from the AMD generated at the Parys Mountain Cu-Pb-Zn deposit, North Wales. The pH played a very important role in the sorption/removal of the contaminants and a higher adsorbent ratio promoted the increase of pH, particularly using natural clinker-based faujasite (pH increased to 7.7-9.43), and a decrease in metal concentration. Selectivity of faujasite for metal removal was, in decreasing order, Fe > As > Pb > Zn > Cu > Ni > Cr. The immobilisation of heavy metal ions by ion exchange and adsorption is likely to be accompanied by precipitation of metal hydroxide complexes (mainly of Fe) on active sites of the particle surface. However, different sorbents contain considerable amounts of accessory phases that can partly dissolve during the batch reaction and result in a sudden increase or decrease in metal concentration. The release of these elements is controlled by the dissolution of the sorbent.

Bauxsol^{TM} has received attention as a potential media for combined precipitation and sorption of metals from mine drainage. Lapointe *et al*. (2006), for example, describe laboratory-scale experiments investigating the suitability of such media in a permeable reactive barrier application. This media has the advantage that it is produced from the red mud waste from alumina production. Its high alkalinity promotes elevation of pH, and therefore precipitation of metals such as iron in particular. Lapointe *et al*. (2006) used a highly acidic (pH ~ 2), metal-rich water in their experiments (8,560 mg/l Fe, 31.2 mg/l Cu, 5.1 mg/l Ni, 5.3 mg/l Zn, 1.1 mg/l Pb), which was leached through bauxsol^{TM} columns twice weekly over a period of approximately three months. Performance was generally good, although it appeared to deteriorate over the course of the experiments. Primary mechanisms of removal cited are precipitation on outer surfaces of bauxsol^{TM} for iron, and adsorption with the pellets for other metals (LaPointe *et al*. 2006). Although apparently successful, longer-running experiments would be required prior to any larger scale application in the UK, for a number of reasons: (1) three months is a rather short experimental period (particularly under non-continuous flow conditions), (2)

issues of substrate exhaustion/clogging would need to be investigated and (3) although not specified, assuming a substrate porosity of 50%, residence time of water in the columns was in the order of eight days in LaPointe *et al.* (2006) experiments, far too long to be realistic for a UK setting.

Despite limitations such as those noted above, further development of such passive technologies may offer promise for the future, especially where there is a potential to utilise waste ochre from coal mine drainage treatment systems for the remediation of non-coal mine water discharges. Particular development needs, given that the capacity of any media for sorption is finite, are to develop technologies that either regenerate media within such treatment units, or to engineer systems such that removal (and disposal) and replacement of exhausted media can be undertaken straightforwardly during operation.

4.3   Carbonate-based reactors

Nuttall (1999) and Nuttall and Younger (2000) investigated the use of anoxic limestone drains (ALD) to immobilise zinc. The principle is that at a pH of approximately 8.2 zinc will form its carbonate mineral, smithsonite, as shown on the phase diagram in Figure 2. Table 3 (Row 4) shows that whilst residence time in the pilot-scale system was reasonable (14 hours), area-adjusted removal rate was still low (0.45 g/m^2/day), and treatment efficiency was comparatively poor at 17% (probably a function of the residence time). The laboratory-scale system performed somewhat better, with an average reduction in Zn of 50%. This difference between laboratory and pilot scale is probably due to the difference in temperature and the difference in limestone particle size. This approach is currently being investigated further at a pilot-scale system on the Red River, Cornwall. Although data for the latter system have yet to be thoroughly interrogated, first indications are that the system is remediating zinc to good effect. The key issue will be whether the rate of the process is sufficiently fast to make this a feasible option for full-scale deployment.

Wang and Reardon (2001) propose the use of a packed-bed reactor filled with siderite and calcite to remove As and Cd from water which is already low in dissolved Fe. Siderite dissolution is achieved in a saturated flow reactor, and then water is aerated over calcite clasts where Cd is precipitated as its carbonate ($CdCO_3$, otavite). Simultaneously, ferric hydroxide is precipitated, which acts as a strong sorbent for AsO_4^{3-}. Removal of As and Cd to concentrations below detection limits was achieved for a residence time of two hours. Therefore, such units may hold promise for metal mine water discharges where cadmium is the primary concern.

Anoxic limestone drains (ALD) are widely cited as a means of elevating pH for acid mine drainage. However, a common problem with traditional ALD (typically used for acidic coal mine drainage, though not to date applied in the UK) is that they suffer problems due to clogging and coating of the reactive grains with precipitates, especially when used to treat very high metal concentrations, within a few months of operation. In order to overcome this problem, Rötting *et al.* (2008) developed a novel reactive material called dispersed alkaline substrate (DAS), which contains an inert pine wood shavings matrix, to supply a high porosity and to reduce clogging, along with a fine-grained reagent (such as limestone) to increase reactivity. This material was tested in a pilot plant (Caraballo *et al.* 2009) which included two tanks filled with limestone and wood shavings and one tank filled with caustic magnesia powder (MgO) and wood shavings to remove divalent metals, such as Zn, Ni, Cd and Co. The water reached a pH of 8-9 in the MgO tank and after nine months of operation the system achieved an average removal of 100% Al, Cu, As, Pb, >95% Cr, > 70% Fe, 25% Zn, almost 20% Ni, 10% Co and 5% Cd. Preferential flow along the walls of the tank was responsible for its low treatment performance and almost all reagent in the MgO tank was exhausted by

the end of the experiment. A secondary, but important, problem with this type of reactor is that availability of MgO in the UK is very limited (personal communication, Dr Tobias Rötting, Newcastle University, 2009).

Aziz *et al.* (2008) undertook a series of batch studies using a variety of media of differing particle size, and found that limestone removed >90% of most metals at a final pH of 8.5, whereas crushed bricks removed 80%, gravel 65% and aeration and settlement without solid media < 30%. The removal of heavy metals was influenced by the media and not directly by the pH. Adsorption and precipitation as metal oxides and probably carbonates contributed to the removal of metals from solution. At higher pH levels, heavy metals are removed from solution by co-precipitation with calcite or by sorption on Fe or Mn oxides that are precipitated on carbonate particles. The results indicate that a rough surface of solid media and the presence of carbonate are beneficial in the removal of heavy metals from water.

From the literature reviewed here, it appears that either (a) metal removal rates in carbonate-based systems are poor or (b) where metal removal rates are high, they tend to suffer from clogging problems.

4.4　Anaerobic and anoxic passive treatment systems

Current passive treatment options include a number of systems which work under anoxic conditions with the aim of encouraging bacterial sulphate reduction (BSR) in organic substrates, to simultaneously generate alkalinity and immobilise metals as sulphides, as illustrated in Reactions 7 to 9. To thrive, sulphate reducing bacteria (SRB) require a strict anaerobic environment with a pH in the range of 5 to 8 (Cohen, 1996). When pH and/or redox conditions are not optimum, the rate of microbial sulphate reduction declines. This in turn reduces metal removal capacity.

Compost wetlands and Reducing and Alkalinity Producing Systems (RAPS) are the most common configurations for treating mine water where it emerges at surface. It is evident from the solubility products for metal sulphides (shown in Table 1), that some of the key metals present in non-coal mine drainage are removed preferentially over the iron which is ubiquitous in coal mine drainage. Thus, in theory at least, the order of removal of metals as sulphides, under appropriate geochemical conditions, is Cu > Cd > Pb > Zn > Ni > Fe^{2+} > Fe^{3+} > Mn. In principle, therefore, the use of compost-based passive systems, operating under anoxic conditions, appears more promising as a means of removal of metals such as zinc than alternative systems operating under aerobic conditions, and/or systems reliant on sorption processes or immobilisation of metals as carbonates.

Many studies have explored the potential of such compost-based systems for the remediation of mine drainage. In South Africa, developments have focussed on harnessing very high rates of BSR in what are effectively active treatment systems (since the aim is to remove sulphate to levels which make the water usable for irrigation or even potable supply). Nevertheless, much can be learned from these experiences in terms of possibilities for enhanced passive treatment. Coetser *et al.* (2006), for example, noted that the chemical composition of the substrate critically influences its ability to drive BSR, noting in particular that high protein content encourages BSR, whilst high fibre content restricts it. Gibert *et al.* (2004), investigating BSR in passive units, found similar results from an investigation of compost, sheep manure, poultry manure and oak leaf, identifying substrates with the least lignin content and highest 'easily available substances' (EAS) as the most effective at encouraging sulphate removal.

Recognising the importance of the characteristics of the carbon source for driving BSR, many investigators have explored the possibility of adding carbon sources to the compost substrate to maximise rates of BSR. Many have used ethanol, acetate and methanol as a carbon source in laboratory-scale reactors, since these compounds provide an immediately available source of carbon (for example Bilek 2006; Mayes and Jarvis 2008; Nevatolo *et al.* 2010). Alternatively, or additionally, the compost may be inoculated with sulphate reducing bacteria before or during reactor start-up (for example Pereyra *et al.* 2008). Details of some of these investigations are provided below.

Mayes and Jarvis (2008) undertook continuous-flow laboratory column trials to test the ability of substrates containing carbonate-rich material mixed with woodchip, manure and anaerobic digested sludge to remove metals from a synthetic, acidic, metalliferous mine water. The column bioreactors were designed to maximise removal of metal contaminants such as zinc, lead and copper through microbial sulphate reduction. To enhance the sulphate reduction process, the effects of addition of a refined low-molecular weight organic compound, in this case methanol, were tested. Residence time was approximately 12-14 hours. Methanol was added to the feedstock once breakthrough in dissolved Zn occurred, to serve as an electron donor to microbial communities for sulphate reduction. Removal of >99% of dissolved Zn was sustained for nearly four months before any breakthrough in dissolved Zn was apparent (removal rate of 1g Zn/m^3/day). The breakthrough of zinc corresponded to increasingly aerobic conditions (evident by rising Eh). However, after addition of methanol more reducing conditions returned, resulting in further sustained removal of all contaminant metals and sulphate, and enhanced alkalinity generation. Carbonate-rich material, mixed with the organic substrate, comprised either shells or limestone. Shell treatment provided slightly better alkalinity generation and pH buffering, likely due to the higher specific surface area of the shells, permitting greater dissolution of carbonate minerals than with the more spherical limestone clasts. An effluent pH in the region of 6.6-7.6 was consistently maintained in the columns. After addition of methanol ceased, there was a rapid fall in alkalinity over 10 days to pre-methanol addition levels, along with an increase in dissolved Zn. Within one week of methanol addition being ceased, there was an immediate and rapid upturn in Eh values, similar to those when breakthrough of Zn was originally observed. This is of concern to sustaining suitable conditions for microbial sulphate reduction in the absence of methanol addition. Column trials demonstrated that effective metal removal through bacterially mediated sulphate reduction can be maintained in the column bioreactors through deployment of appropriate substrates and addition of methanol to the feedstock.

Given the longevity of metal mine water discharges it would be preferable to use waste sources of carbon rather than laboratory chemicals such as methanol. It is for this reason that additional, as yet unpublished, work is ongoing to evaluate possible waste carbon sources that can be used as an amendment to systems such as that investigated by Mayes and Jarvis (2008). Similarly, wine wastes as a carbon source have been studied by Costa *et al.* (2009). The results of that work showed that efficient neutralisation and high sulphate removal (> 90%) were attained with 97% removal of Zn and Cu. Costa *et al.* (2009) note that the choice of carbon source for SRB activity can be the key to ensuring high performance, long-term efficiency and economical viability of the treatment. Selection of a suitable carbon source and electron donor for biological sulphate reduction was based on three factors in their experiments:

(1)   degradability of the carbon source and hence its capacity to allow complete sulphate reduction by SRB;

(2)   its cost per unit of sulphate converted to sulphide;

(3)   its availability.

The waste from the wine industry had a similar composition to that of diluted red wine, with pH 3.8-3.9. The results of Costa *et al.* (2009) showed that neutralisation occurred but a slight decrease in effluent pH was observed with non-optimal values for SRB (<5.5) after 150 days of operation. Similarly, sulphate reduction was high until pH < 5.5, and then sulphate concentration increased. Hence, the results emphasise that biological sulphate reduction is limited by pH. Arguably more significant was that the hydraulic retention time of waters within the reactors operated by Costa *et al.* (2009) was eight days, and it was necessary to adopt a daily carbon addition regime to maintain reactor performance. A key objective of future investigations, in the context of passive mine water treatment in the UK, needs to be establishing whether it is feasible to effect good treatment performance at lower residence times, and with less frequent requirements for carbon additions.

Irrespective of the carbon source used as an amendment over the medium to long term, the start-up conditions of these reactors appears to be important also. Both Johnson and Hallberg (2005) and Mayes and Jarvis (2008) noted that a period of maturation of the organic substrate (four to ten months) prior to use appears to improve subsequent performance, presumably due to the development of microbial populations. In an effort to improve initial microbial populations, and maintain active colonies, mine waters and/or substrates may be inoculated with microbial populations at the outset to encourage their establishment (Pereyra 2008).

Johnson and Hallberg (2005) studied the compost bioreactor components of three composite passive remediation systems constructed to treat acid mine drainage at the former Wheal Jane tin mine in Cornwall. Although an increase in pH and decrease in metal concentrations was observed in each of the bioreactors, only one showed effective removal of the two dominant heavy metals, Fe and Zn (>99% removal). This bioreactor had been shut down for 10 months prior to the monitoring period due to operational problems and this appears to have allowed more successful development of sulphate-reducing bacteria. The authors recommend that compost bioreactors be allowed to "mature" in terms of developing adequate populations of compost-degrading microorganisms, SRB, and other important microbes, before throughput of mine water is initiated.

Pereyra *et al.* (2008) concluded similarly when comparing different microbial inocula in batch tests investigating sulphate and metal removal and pH neutralisation. The study showed that pre-acclimated material from a sulphate-reducing permeable reactive zone was ideal. The authors also note that the handling and storage of the inoculum are important aspects that must be taken into account. Jong and Parry (2003) used BSR inocula when investigating the treatment of mildly acidic, metal- and sulphate-contaminated waters using BSR in a bench-scale upflow anaerobic packed bed reactor. The pH in these experiments increased from 4.5 to 7.0 while the reactor removed >82% sulphate and more than 97.5% of Cu, Zn and Ni. The reactor was initially inoculated with 400 ml of a mixed culture of BSR with continuous flow started 14 days after inoculation. Each experiment was then conducted for 14 days (far too short a period to give confidence in long-term performance). In these experiments, Cu was the first metal to be removed, followed by Zn, Ni and finally Fe, which exactly reflects the trend in solubility products of the respective metal sulphides (see Table 1).

Notwithstanding these issues of SRB inocula and ongoing carbon requirements, in principle selective precipitation of metals could be achieved using BSR reactors, due to the differing solubility products of metals. This has implications for potential resource (metal) recovery and re-use. Selective precipitation has been the subject of research for some investigators. For example, Sahinkaya *et al.* (2009) were able to separate Cu and Zn precipitation using sulphide produced in an anaerobic baffled reactor. The reactor was fed with ethanol and sulphate. Cu was precipitated at low pH (<2) using sulphide transported from the reactor effluent via N_2 gas followed by Zn recovery using

reactor effluent containing sulphide and alkalinity. Removal efficiency varied between 84 and 98% and pH was increased from 3 to 6-7.5. Separate precipitation was achieved by controlling the reactor pH since Cu can be precipitated as CuS at very low pH (< 1) without precipitating other heavy metals, whereas Zn does not precipitate as ZnS until pH 1.3. To accomplish this it is necessary to separate the biological sulphide reduction process from metal precipitation, in order to optimise the conditions for both SRB (pH 7-8) and selective Cu precipitation over Zn (pH < 2) when sulphide is in excess.

Whilst selective precipitation has been shown to be feasible in principle, there is little doubt that such units would in all likelihood be active systems due to the close process control required, particularly with respect to pH. One such 'active biological system' is described by Johnson *et al.* (2006) who were able to demonstrate selective removal of zinc as ZnS using a consortium of acidophilic bacteria that is active at low pH (pH 3 - 4). The system used glycerol, acetic acid and hydrogen as energy sources. Such systems, however, are only cost-effective at high concentrations and due to the close process control required, they represent a very expensive treatment option at a remote site.

As an alternative to a compost-based system, the US Environmental Protection Agency installed a compost-free sulphate reducing bioreactor system to treat acid rock drainage at Leviathan Mine, California (USEPA 2006). The system consists of a pre-treatment pond, two bioreactors, two settling ponds and an aeration channel. Sodium hydroxide solution is added to the metal-rich influent within the pre-treatment pond, together with ethanol as a carbon source, before the influent is passed through two bioreactors containing river rock. After passing through the bioreactors, sodium hydroxide solution is again added to the effluent to raise the pH to neutral. The total system hydraulic residence time is 107 hours (~ 4.5 days) and the system has been able to achieve a target metal removal efficiency of 95%. The pH of the influent is also increased from 3.0 to 7.0 within the system. The use of a liquid carbon source and a rock matrix has the advantage over a compost and wood chip matrix of improved control over biological activity along with improved hydraulic conductivity and precipitate flushing. However, the use of continuous carbon and alkali additions is an active intervention, with implications for operation and maintenance costs. The intention of the current research is to investigate the possibility of periodic, rather than continuous, carbon additions.

One final variant of the use of BSR for metal mine water treatment is the use of algae. Rose *et al.* (1998), for example, investigated the use of waste stabilisation ponds (WSP) as a treatment unit for co-treatment of organic waste streams and acid mine drainage. The algae developed as a consequence of the organic waste stream serve as a carbon source for BSR, which therefore immobilise metals within the WSP. Hydraulic residence time within the systems investigated by Rose *et al.* (1998) were in the region of three days, and therefore are more in line with requirements for the UK situation. Boshoff *et al.* (2004) subsequently demonstrated the potential use of dried algae as a direct carbon source for feeding BSR reactors.

The potential to co-treat organic and inorganic wastewaters is attractive, but it is not clear how much opportunity there would be to adopt such approaches in the UK, particularly given the remote locations of many UK metal mine water discharges. The use of algal systems may offer some potential in the UK, but there would likely be issues with maintaining performance during the winter months (noted as an issue by Kalin (1998)), unless artificial light sources could be installed (with obvious implications for operation and maintenance). It is worth noting that the work of both Boshoff *et al.* (2004) and Rose *et al.* (1998) was undertaken in South Africa, where climate may be more amenable to this type of approach.

In conclusion, there seems little doubt that in general terms the deployment of compost-based passive systems offers more promise for the remediation of non-coal mine drainage than aerobic wetland systems. Nevertheless, further development work is required to provide the confidence in performance needed before full-scale installation proceeds. Particular issues that require resolution include:

- Pilot-scale experimentation to establish performance under field conditions, determine modes of metal removal, and begin to quantify scale-dependence issues. Most studies are undertaken at laboratory scale (in preparing this review it was found that of 130 recent publications on compost-based systems, only two were conducted at pilot or full scale under field conditions), and therefore do not account for issues of different environmental conditions at actual mine sites, or of the very substantial differences in performance that may result by increasing the scale of operation by several orders of magnitude (due, for example, to non-ideal hydraulic performance).

- Identification of potential carbon additives and sources of microbial inocula, and the precise behaviour of SRB under various start-up conditions. A number of carbon additives have been explored, including methanol (for example Bilek 2006; Mayes and Jarvis 2008), ethanol (for example Nevatalo *et al.* 2010; Sahinkaya *et al.* 2009; USEPA 2006), cheese whey (Martins *et al.* 2009) and wine wastes (for example Costa *et al.* 2009). However, although an increase in metal attenuation is observed immediately following carbon addition, the carbon source tends to be rapidly exhausted with frequent additions necessary to maintain system performance. In the course of the project, various solid and liquid carbon additives will be investigated at both laboratory and pilot scale, including waste from the brewery industry and CellMatt, a solid waste product produced by steam autoclaving. For solid carbon sources simple placement of the media within the substrate will be undertaken once effluent Zn concentrations begin to increase. Also, very slow addition, or periodic addition, of liquid sources may be investigated, since mixing and retention of such liquids may be more effective at pilot scale than at full scale.

- Assessment of the robustness of SRB communities under field conditions.

- Determination of area-adjusted removal rates in pilot-scale systems, to carry forward to full-scale design.

Arguably the single biggest issue to be addressed in a UK context is resolution of the problem of hydraulic residence time in compost systems. In the vast majority of cases, high performance systems have relied upon residence times that are simply too long to be realistically feasible in UK settings. From published literature to date it would appear that the key to solving this problem is the identification and application of suitable inocula and carbon additions for such compost-based systems.

5. Conclusions and recommendations

This review focuses on the passive remediation of discharges from abandoned metal mines, in particular the different types of passive treatment system available and their performance. The principal conclusions are:

1. The most common contaminants of metal mine discharges are metals, while acidity is less commonly a pollutant of metal mine discharges. However, pH has a crucial control on the geochemical behaviour of metals in aqueous environments so is an important parameter to consider during treatment system design. Redox potential is also a key control on the state of metals in aqueous environments and the pH-Eh relationship can be used to predict the behaviour of metals.

2. The primary aim of metal mine water treatment is to immobilise metals within the confines of the treatment system and raise the pH, where necessary.

3. The area-adjusted removal rate (mass of metal removed per unit area of treatment system per unit of time) is a key indicator of treatment system performance. Compared to aerobic wetland systems treating coal mine drainage (which primarily remove iron), the area-adjusted removal rate of zinc in passive treatment systems is significantly lower. An aerobic wetland for attenuation of zinc would therefore need to be considerably larger than an equivalent system for remediation of iron pollution.

4. The principal mechanism by which iron is removed from coal mine drainage is by the precipitation of (oxy)hydroxides. However, zinc and other metals common in metal mine discharges have higher hydroxide solubility products than iron and therefore require a higher target pH for their precipitation which represents a significant problem in their remediation.

5. Other mechanisms, including co-precipitation, adsorption and bioaccumulation, may also serve to immobilise metals within treatment systems although many are pH-dependent, with implications for the rate of metal removal. In addition, sorption media may become exhausted and it is possible, if pH/Eh conditions change, for desorption of metals to occur, potentially releasing metals into solution.

6. The most promising means of immobilising metals appears to be by the precipitation of sulphides in anaerobic systems in which bacterial sulphate reduction in organic substrates simultaneously generates alkalinity and immobilises metals as sulphides. The solubility products of the sulphides of some of the key metals present in metal mine discharges (such as zinc) are lower than that of iron sulphide and thus these metals should be removed preferentially over iron. However, a key requirement is the maintenance of bacterial sulphate reduction and much research has been devoted to identifying suitable carbon sources for maximising rates of bacterial sulphate reduction.

The principal recommendations for future research are:

1. Pilot-scale experimentation to establish the performance of the technologies described in this review under field conditions, determine modes of metal removal, and begin to quantify scale-dependence issues. The majority of studies, particularly those utilising BSR in anaerobic systems, have been

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undertaken over short time periods at laboratory scale and therefore do not account for issues of different environmental conditions at actual mine sites.

2. Identification of potential carbon additives and sources of microbial inocula in order to reduce the hydraulic residence time of anaerobic compost-based systems along with an assessment of the robustness of SRB communities under field conditions.

3. Determination of area-adjusted removal rates in pilot-scale systems so that designs can be carried forward to full scale.

4. Further development work on the process of sorption of metals onto different types of media, in particular for technologies that either regenerate media within the treatment units or the engineering of systems such that replacement of exhausted media can be undertaken straightforwardly during operation.

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APPENDIX A: Occurrence and concentration of metals and metalloids in metal mine drainage across England and Wales

1. Each histogram shows the distribution of concentrations of each metal or metalloid (in units of mg/l) across all mine waters analysed.

2. The first category of concentration represents a typical environmental quality standard (EQS) for the particular metal or metalloid; for example, for arsenic the EQS is 0.05 mg/l, and therefore the first category is < 0.05 mg/l. Note that these are indicative EQS values as (a) some EQS values vary depending on other chemical variables and (b) EQS values are subject to change.

3. In the title of each graph n = number of samples analysed and mean is the mean concentration of all analyses.

4. Note that a low value of n may in principle simply be indicative of how often that particular metal or metalloid is determined, rather than a reflection of how often it occurs at elevated concentrations.

5. Values of zero recorded in the original data have been ignored for the purposes of this assessment.

6. Data for zinc and flow rate are shown in the main text.

Arsenic (n = 16, mean = 0.085)

Cadmium (n = 105, mean = 0.035)

Manganese (n = 86, mean = 4.86)

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Nickel (n = 51, mean = 0.06)

Copper (n = 84, mean = 0.57)

Iron (n = 95, mean = 32.8)

Lead (n = 121, mean = 0.58)

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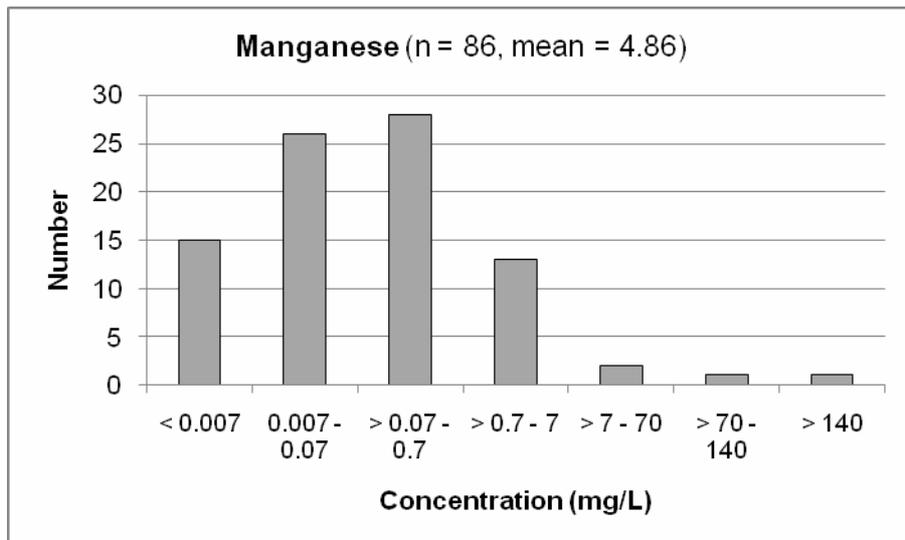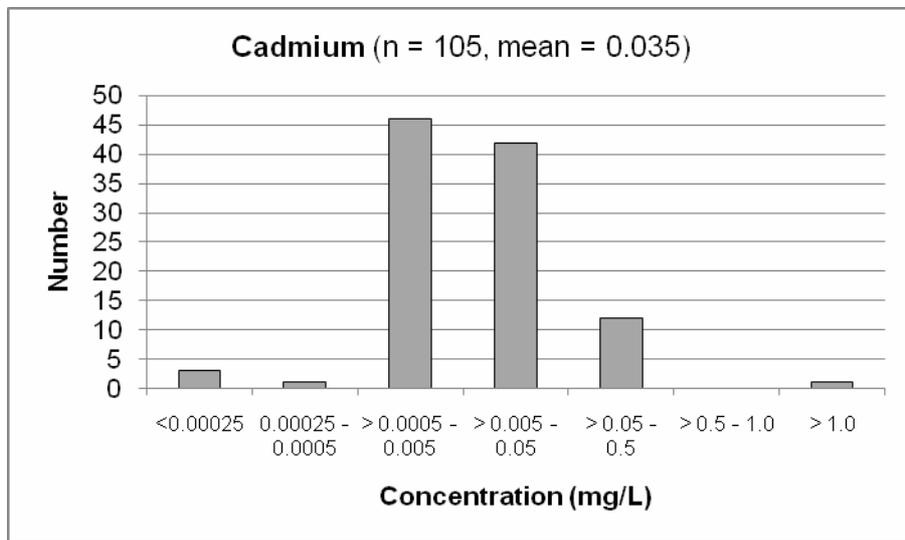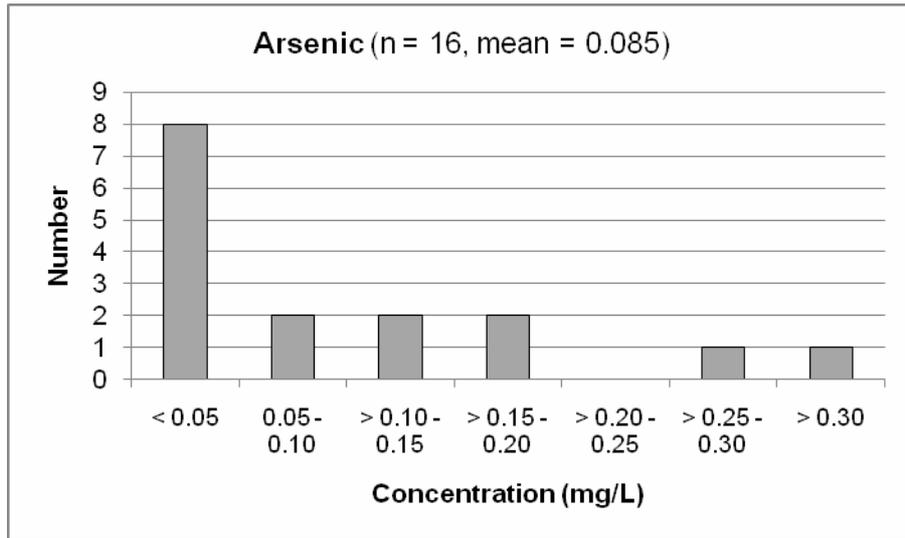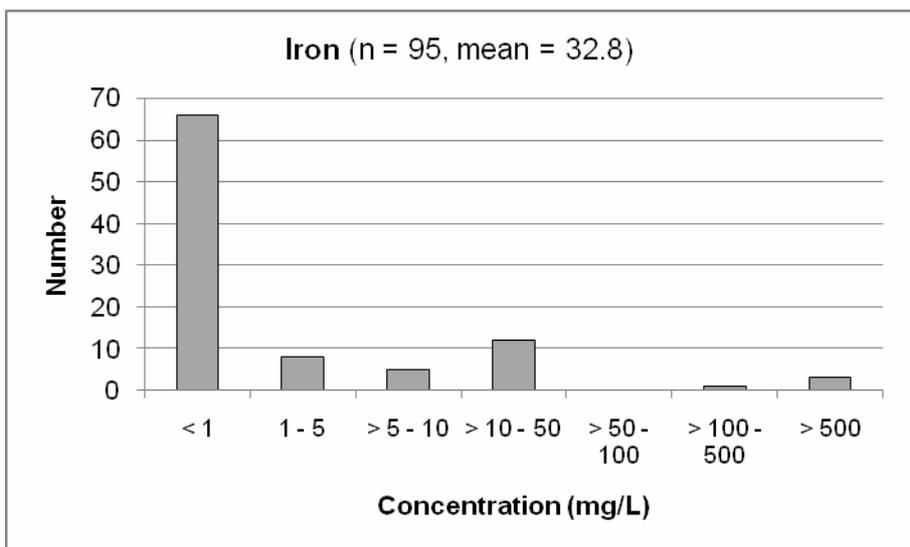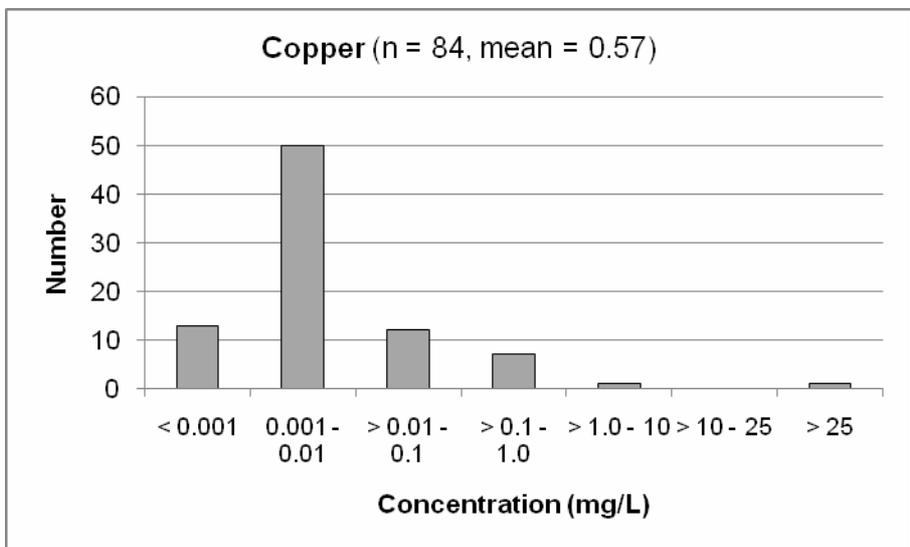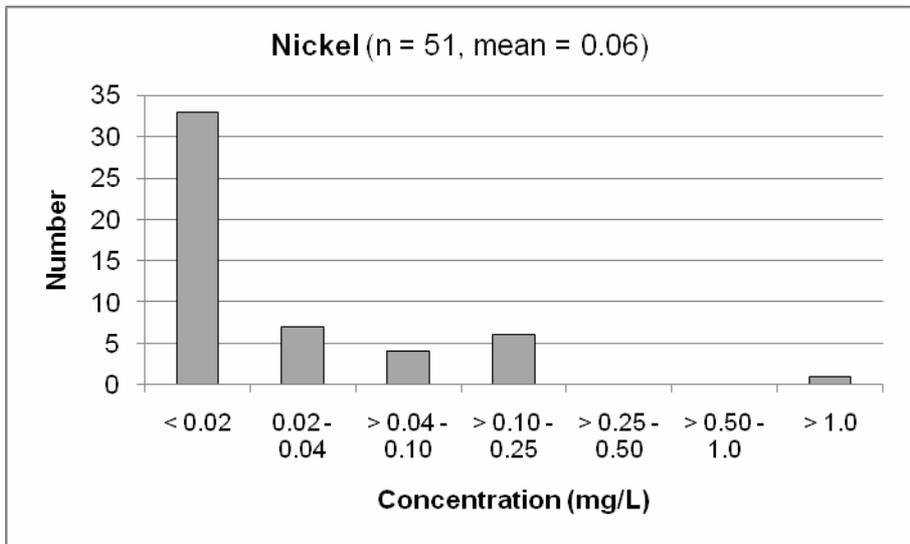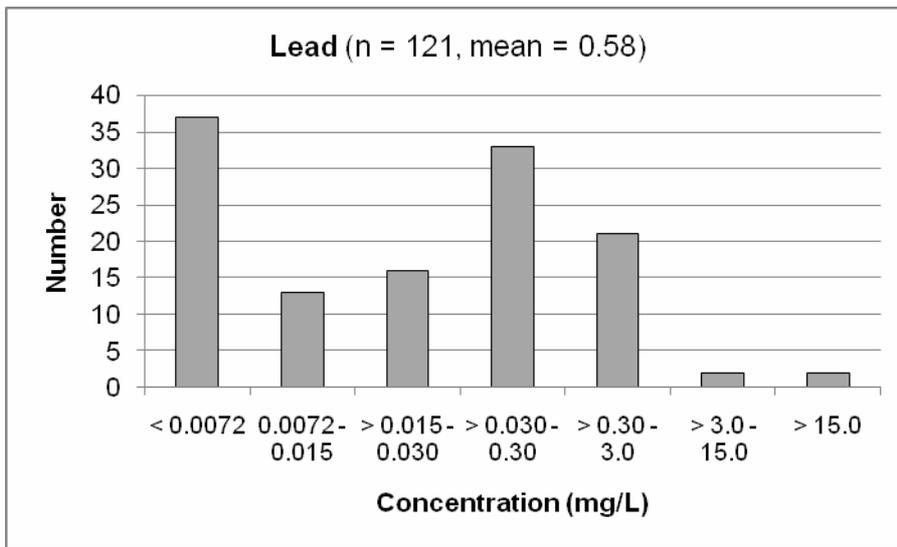
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